

SIMULATION AND OPTIMIZATION STUDIES OF $IC_{60}BA$ BASED
BULK HETEROJUNCTION ORGANIC SOLAR CELLS

A **THESIS**

submitted to the



University of Calicut

for the award of the degree

of

Doctor Of Philosophy

in Physics

by

RAMABADRAN C D



Opto-electronic Device Simulation Research Lab

Post Graduate And Research

Department of Physics

Christ College (AUTONOMOUS), Irinjalakuda

January 2026

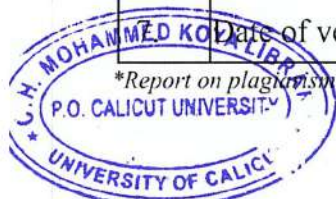


UNIVERSITY OF CALICUT

CERTIFICATE ON PLAGIARISM CHECK

1.	Name of the Research Scholar	Ramabadran. C. D	
2.	Title of thesis / dissertation	Simulation and Optimization Studies of IC60BA Based Bulk Heterojunction Organic Solar Cells	
3.	Name of the Supervisor	Dr. Sudheer Sebastian. K	
4.	Department/Institution	Department of Physics, Christ College(Autonomous), Irinjalakuda	
5.	Similar content (%) identified	Non Core	Core
		Introduction/ Theoretical overview/Review of literature/ Materials & Methods/ Methodology	Analysis/Result/Discussion / Summary/Conclusion/ Recommendations
		3	3
		10	10
6.	Software used	iThenticate	
	Date of verification	20.01.2026	

*Report on plagiarism check, specifying included/excluded items with % of similarity to be attached.



SEEMA M.V.
Assistant Librarian
University of Calicut
Malappuram - 673 635

Checked by (with name , designation & signature) *Seema*

Name and signature of the Researcher *Ramabadran. C. D. : e. D. R. L.*

Name and signature of the Supervisor.

Dr. SUDHEER SEBASTIAN K
PROFESSOR
DEPARTMENT OF PHYSICS
CHRIST COLLEGE (AUTONOMOUS)
IRINJALAKUDA - 680125

The Doctoral Committee* has verified the report on plagiarism check with the contents of the thesis, as summarized above and appropriate measures have been taken to ensure originality of the Research accomplished herein.

Name & Signature of the HoD/HoI (Chairperson of the Doctoral Committee)

Fr. Dr. Jolly Andrews
Associate Professor -
In-Charge of Principal
Christ College (Autonomous)
Irinjalakuda

*In case of languages like Malayalam, Tamil etc..on which no software is available for plagiarism check, a manual check shall be made by the Doctoral Committee, for which an additional certificate has to be attached.



Dedication

Dedicated to my family

For their endless love and support

Certificate

This is to certify that the thesis titled **Simulation and Optimization Studies of IC₆₀BA based Bulk Heterojunction Organic Solar Cells**, submitted by **Ramabadran.C.D**, to the University of Calicut for the award of the degree of Doctor of Philosophy, is a bonafide record of the research work done by him under my supervision. The contents of this thesis, in full or in parts, have not been submitted to any other Institute or University for the award of any other degree.




Dr. Sudheer Sebastian K

Dr. SUDHEER SEBASTIAN K Research Guide
PROFESSOR
DEPARTMENT OF PHYSICS
CHRIST COLLEGE (AUTONOMOUS)
IRINJALAKUDA - 680 125
Professor
Department of Physics
Christ college (Autonomous) Irinjalakuda
Irinjalakuda North, Thrissur
680 125

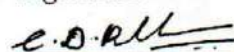
Place: **Irinjalakuda**

Date: **27-01-2026**

DECLARATION

I hereby declare that the work presented in the thesis entitled “**Simulation and Optimization Studies of IC₆₀BA based Bulk Heterojunction Organic Solar Cells**” is based on the original work done by me under the guidance of Dr. Sudheer Sebastian K and has not been included in any other thesis submitted previously for the award of any degree. The contents of the thesis are undergone plagiarism check using iThenticate software at C.H.M.K. Library, University of Calicut, and the similarity index found within the permissible limit. I also declare that the thesis is free from AI generated contents.

Signature



Ramabadran.C.D



Signature of the Supervising teacher



Dr.Sudheer Sebastain.K

Place: Irinjalakuda

Date: 27-01-2026

Dr. SUDHEER SEBASTIAN K
PROFESSOR
DEPARTMENT OF PHYSICS
CHRIST COLLEGE (AUTONOMOUS)
IRINJALAKUDA - 680125

Acknowledgements

It is with a profound sense of gratitude that I acknowledge my research guide, Dr. Sudheer Sebastian K. His invaluable mentorship, unwavering support, and consistent encouragement have been the cornerstone of this research endeavor. I am deeply indebted to him for motivating me to pursue this work with utmost dedication and sincerity. I am especially thankful for his exceptional care and commitment during the formative initial years of my Ph.D. journey, which provided the essential foundation for this thesis.

I express my sincere gratitude to Dr. Fr. Jolly Andrews, Principal-in-Charge, Christ College (Autonomous), Irinjalakuda, for his continuous encouragement and support throughout my research work.

I would like to express my sincere thanks to Dr. Antony Joseph, former Professor, Department of Physics, University of Calicut, for his invaluable support and insightful discussions during the course of this research. The knowledge and perspectives he shared significantly enhanced the quality and depth of this work.

I wish to express my profound gratitude to Dr. Pradyumnan P. P., Senior Professor, Head of the Department of Physics, University of Calicut, and member of the Research Advisory Committee, for his assistance and insightful recommendations

related to my research.

I extend my heartfelt thanks to Dr. Gopikrishnan, Associate Professor of Physics, Government Victoria College, for his extensive support during the writing of my thesis. I also take this opportunity to express my sincere thanks to Dr. Lakshmi, Head of the Department, Mercy College, Palakkad, for her support throughout my research work.

I am very thankful to my dear colleagues Dr. Nithya K. S., Dhandapani Sir, Shamna, Kavitha, Vijay Joy, Dennet Davis, Dr. Bindhu, Dr. Anju Sebastian, Dr. Sikha K. Simon, and Ms. Aswathy P. V. for their friendly cooperation and professional support.

I would like to express my sincere gratitude to my dear friend Dr. Richard Scaria, Associate Professor of Geography and Syndicate Member, University of Calicut, and to Anoop Sir, Assistant Professor of Commerce, for their valuable assistance in the submission of my thesis. I would like to express my sincere gratitude to my colleagues, Ajeesh P.V. and Dr. Arya S. of the Department of Physics, Government College Chittur, for their invaluable support and assistance throughout this research.

I wish to convey my profound appreciation to Prof. Burgelman, University of Ghent, for generously providing access to the SCAPS-1D simulation software. His dedication to supporting the academic and research community has been truly invaluable.

I would also like to express my gratitude to the office personnel, non-teaching staff, administrative staff, faculty members, and laboratory staff of the Department of Physics for their assistance and support.

I record my heartfelt gratitude for the blessings of my grandparents. I am extremely indebted to my wife, Lakshmi, who stood beside me through every storm and every sunshine. Her love and encouragement have been my greatest strength, and without her ceaseless care, I could not have completed this work. I am also sincerely grateful to my family members for their unconditional support.

Finally, I bow my head to God Almighty, who guided me through the hardest and toughest times of my life.

Ramabadrán.C.D

Abstract

Global energy consumption is increasing as the population grows and living standards improve. Unless energy-harvesting technologies are developed and deployed, the world will face a serious energy crisis. Of all physical resources, the solar energy incident to Earth is the most significant resource, and it is imperative to discover ways to convert it into usable forms suitable for the modern requirements of our society .

Despite a myriad of photovoltaic technologies that exist, organic solar cells (OSCs) stand out because of unique properties that include the availability of materials, light-weight, mechanical flexibility, low cost, roll-to-roll production, and ease of fabricating into a variety of products.

Over the last two decades, OSCs have improved significantly, particularly in bulk heterojunction architectures using polymer donors and fullerene or non-fullerene acceptors. To maintain the smooth operation, export stability, and efficiency of multilayer OSCs, it is essential to select appropriate charge-transport layers. A deeper understanding of the mechanics of fullerene bulk heterojunctions is therefore necessary to drive a rapid further rise in organic photovoltaics in the near future. Device modelling and simulation serve as powerful tools for understanding solar cell operation and optimizing the active layer in combination with the most efficient

transport-layer configurations.

This thesis, entitled “Simulation and Optimization Studies of $IC_{60}BA$ Based Bulk Heterojunction Organic Solar Cells,” examines the performance of fullerene-based multilayer organic solar cells incorporating P3HT: $IC_{60}BA$ as the active layer and a variety of hole and electron transport layers. To achieve the best possible outcome, the solar structure is modeled, and its performance is examined for various hole and electron transport layers. The impact of various material parameters on the output performance of the solar cell is also thoroughly explored, and a new design for a fullerene multilayer organic solar cell is proposed. The primary objectives of this study are to use simulation tools to design a highly efficient P3HT: $IC_{60}BA$ fullerene-based multilayer organic solar cell with optimally matched transport layers, and to propose novel device architectures that can support future advancements in organic photovoltaics. The careful design of multilayer structures with optimized transport layers holds substantial promise for accelerating the near-term commercialization of organic photovoltaic technologies.

Key words : Numerical simulation ,Organic Photovoltaics , Bulk heterojunction, Indene-C60 Bisadduct ($IC_{60}BA$), Power Conversion Efficiency (PCE)

സംഗ്രഹം

ജനസംഖ്യ വർദ്ധിക്കുകയും ജീവിത നിലവാരം മെച്ചപ്പെടുകയും ചെയ്യുന്നതിനനുസരിച്ച് ആഗോള ഊർജ്ജ ഉപഭോഗം വർദ്ധിച്ചുകൊണ്ടിരിക്കുകയാണ്. ഊർജ്ജ വിളവെടുപ്പ് സാങ്കേതികവിദ്യകൾ വികസിപ്പിക്കുകയും വിന്യസിക്കുകയും ചെയ്തില്ലെങ്കിൽ, ലോകം ഗുരുതരമായ ഊർജ്ജ പ്രതിസന്ധിയെ നേരിടേണ്ടിവരും. എല്ലാ ഭൗതിക വിഭവങ്ങളിലും, ഭൂമിയിലേക്ക് വീഴുന്ന സൗരോർജ്ജം ഏറ്റവും പ്രധാനപ്പെട്ട വിഭവമാണ്, നമ്മുടെ സമൂഹത്തിന്റെ ആധുനിക ആവശ്യങ്ങൾക്ക് അനുയോജ്യമായ ഉപയോഗയോഗ്യമായ രൂപങ്ങളാക്കി മാറ്റുന്നതിനുള്ള വഴികൾ കണ്ടെത്തേണ്ടത് അത്യാവശ്യമാണ്. നിരവധി ഫോട്ടോവോൾട്ടെയ്റ്റ് സാങ്കേതികവിദ്യകൾ നിലവിലുണ്ടെങ്കിലും, വസ്തുക്കളുടെ ലഭ്യത, ഭാരം കുറഞ്ഞത, മെക്കാനിക്കൽ വഴക്കം, കുറഞ്ഞ ചെലവ്, റോൾ-ടൂ-റോൾ ഉൽപ്പാദനം, വിവിധ ഉൽപ്പന്നങ്ങളാക്കി മാറ്റാനുള്ള എളുപ്പം എന്നിവ ഉൾപ്പെടുന്ന അതുല്യമായ ഗുണങ്ങൾ കാരണം ഓർഗാനിക് സോളാർ സെല്ലുകൾ (OSC-കൾ) വേറിട്ടുനിൽക്കുന്നു.

കഴിഞ്ഞ രണ്ട് പതിറ്റാണ്ടുകളായി, OSC-കൾ ഗണ്യമായി മെച്ചപ്പെട്ടിട്ടുണ്ട്, പ്രത്യേകിച്ച് പോളിമർ ദാതാക്കളെയും ഫുള്ളറിൻ അല്ലെങ്കിൽ നോൺ-ഫുള്ളറിൻ സ്വീകാര്യരെയും ഉപയോഗിക്കുന്ന ബൾക്ക് ഹെറ്ററോജംഗ്ഷൻ ആർക്കിടെക്ചറുകളിൽ. മൾട്ടിലെയർ OSC-കളുടെ സുഗമമായ പ്രവർത്തനം, കയറ്റുമതി സ്ഥിരത, കാര്യക്ഷമത എന്നിവ നിലനിർത്തുന്നതിന്, ഉചിതമായ ചാർജ്-ട്രാൻസ്പോർട്ട് ലെയറുകൾ തിരഞ്ഞെടുക്കേണ്ടത് അത്യാവശ്യമാണ്. അതിനാൽ, സമീപഭാവയിൽ ഓർഗാനിക് ഫോട്ടോവോൾട്ടെയ്ക്സിൽ കൂടുതൽ ദൃഢതയിലുള്ള ഉയർച്ചയ്ക്ക് കാരണമാകുന്നതിന് ഫുള്ളറിൻ ബൾക്ക് ഹെറ്ററോജംഗ്ഷനുകളുടെ മെക്കാനിക്സിനെക്കുറിച്ച് ആഴത്തിലുള്ള ധാരണ ആവശ്യമാണ്. സോളാർ സെൽ പ്രവർത്തനം മനസ്സിലാക്കുന്നതിനും ഏറ്റവും കാര്യക്ഷമമായ ട്രാൻസ്‌പോർട്ട്-ലെയർ കോൺഫിഗറേഷനുകളുമായി സംയോജിപ്പിച്ച് സജീവ പാളി ഒപ്റ്റിമൈസ് ചെയ്യുന്നതിനുമുള്ള ശക്തമായ ഉപകരണങ്ങളായി ഉപകരണ മോഡലിംഗും സിമുലേഷനും പ്രവർത്തിക്കുന്നു.

" $IC_{60}BA$ അടിസ്ഥാനമാക്കിയുള്ള ബൾക്ക് ഹെറ്ററോജംഗ്ഷൻ ഓർഗാനിക് സോളാർ സെല്ലുകളുടെ സിമുലേഷൻ ആൻഡ് ഒപ്റ്റിമൈസേഷൻ സ്റ്റഡീസ്" എന്ന തലക്കെട്ടിലുള്ള ഈ പ്രബന്ധം, $P3HT:IC_{60}BA$ സജീവ പാളിയായി ഉൾപ്പെടുത്തിയ ഫുള്ളറിൻ അധിഷ്ഠിത മൾട്ടിലെയർ ഓർഗാനിക് സോളാർ സെല്ലുകളുടെയും വിവിധതരം ഹോൾ, ഇലക്ട്രോൺ ട്രാൻസ്പോർട്ട് ലെയറുകളുടെയും പ്രകടനം പരിശോധിക്കുന്നു. സാധ്യമായ ഏറ്റവും മികച്ച ഫലം കൈവരിക്കുന്നതിന്, സൗരോർജ്ജ ഘടന മാതൃകയാക്കുകയും വിവിധ ഹോൾ, ഇലക്ട്രോൺ ട്രാൻസ്പോർട്ട് ലെയറുകൾക്കായി അതിന്റെ പ്രകടനം പരിശോധിക്കുകയും ചെയ്യുന്നു. സോളാർ സെല്ലിന്റെ ഔട്ട്പുട്ട് പ്രകടനത്തിൽ വിവിധ മെറ്റീരിയൽ പാരാമീറ്ററുകളുടെ സ്വാധീനവും സമഗ്രമായി പര്യവേക്ഷണം ചെയ്യപ്പെടുന്നു, കൂടാതെ ഒരു ഫുള്ളറിൻ മൾട്ടിലെയർ ഓർഗാനിക് സോളാർ സെല്ലിനായി ഒരു പുതിയ രൂപകൽപ്പന നിർദ്ദേശിക്കപ്പെടുന്നു. ഒപ്റ്റിമൽ പൊരുത്തമുള്ള ട്രാൻസ്പോർട്ട് ലെയറുകളുള്ള ഉയർന്ന കാര്യക്ഷമമായ $P3HT:IC_{60}BA$ ഫുള്ളറിൻ അധിഷ്ഠിത മൾട്ടിലെയർ ഓർഗാനിക് സോളാർ സെൽ രൂപകൽപ്പന ചെയ്യുന്നതിന് സിമുലേഷൻ ഉപകരണങ്ങൾ ഉപയോഗിക്കുക, കൂടാതെ ഓർഗാനിക് ഫോട്ടോവോൾട്ടെയ്ക്സിലെ ഭാവി പുരോഗതിയെ പിന്തുണയ്ക്കാൻ കഴിയുന്ന നൂതന ഉപകരണ ആർക്കിടെക്ചറുകൾ നിർദ്ദേശിക്കുക എന്നിവയാണ് ഈ പഠനത്തിന്റെ പ്രാഥമിക ലക്ഷ്യങ്ങൾ. ഒപ്റ്റിമൈസ് ചെയ്ത ട്രാൻസ്പോർട്ട് ലെയറുകളുള്ള മൾട്ടിലെയർ ഘടനകളുടെ ശ്രദ്ധാപൂർവ്വമായ രൂപകൽപ്പന, ജൈവ ഫോട്ടോവോൾട്ടെയ്റ്റ് സാങ്കേതികവിദ്യകളുടെ ഹ്രസ്വകാല വാണിജ്യവൽക്കരണം ത്വരിതപ്പെടുത്തുന്നതിന് ഗണ്യമായ വാശനങ്ങൾ നൽകുന്നു.

പ്രധാന വാക്കുകൾ : സംഖ്യാ സിമുലേഷൻ, ഓർഗാനിക് ഫോട്ടോവോൾട്ടെയ്ക്സ്, ബൾക്ക് ഹെറ്ററോജംഗ്ഷൻ, ഇൻഡീൻ-C60 ബിസാഡക്സ് ($IC_{60}BA$), പവർ കൺവേർഷൻ എഫിഷ്യൻസി (PCE)

Contents

Dedication	i
Thesis Certificate	iii
Declaration	iv
Acknowledgements	vii
1 Introduction	1
1.1 Solar Energy	1
1.2 Fundamentals of Photovoltaics	7
1.2.1 Absorption of Light	7
1.2.2 Charge Carrier Generation	8
1.2.3 Separation and Movement of Charge Carriers	8
1.2.4 Collection of Charge Carriers	9
1.2.5 Recombination	9
1.3 The Evolution of Solar Cell Technology	9
1.4 Growth of solar cell Technology	11

1.4.1	First-Generation Solar Cells	11
1.4.2	Second-Generation Solar Cells	12
1.4.3	Third-Generation Solar Cells	16
1.5	Physics of Solar Cells	21
1.5.1	The operation of a p-n junction	21
1.5.2	P-N junction under illumination	24
1.5.3	Solar cell equivalent circuit	24
1.5.4	Basic solar cell output parameters	27
1.5.5	Solar cell recombination mechanisms	31
1.5.6	Basic Semiconductor Equations	35
1.6	Organic Solar Cells: A New Frontier	39
1.6.1	The evolution of organic solar cells	40
1.6.2	Advantages of organic solar cells	41
1.6.3	Organic semiconductor materials	43
1.7	Diverse structural designs of organic solar cells	44
1.7.1	Single-Layer (Schottky) Structure	44
1.7.2	Bilayer (Planar Heterojunction, PHJ) Structure	45
1.7.3	Tandem (Multi-Junction) Structure	46
1.7.4	Inverted Structure	47
1.7.5	Bulk Heterojunction (BHJ) Structure	48
1.8	Thesis Outline	59
2	Simulation tool and Device modelling	61
2.1	Overview of the Simulation Methodology	62

2.1.1	Reasons for using SCAPS-1D	63
2.2	Simulation study workflow	64
2.3	SCAPS-1D Software: Theoretical Structure	65
2.3.1	SCAPS 1D Software Background (version 3.3.08, Capabilities)	65
2.3.2	Solved Basic Semiconductor Equations	66
2.3.3	Boundary Conditions	67
2.4	Material Characteristics: Parameters Required for SCAPS-	
	1D Analysis	68
2.4.1	General Parameters	69
2.4.2	Layer-Specific Parameters	70
2.5	Interface Modelling in SCAPS 1-D	75
2.5.1	Modelling of D/A Interface	76
2.5.2	Modeling of Active Layer/Transport Layer and Transport Layer/Electrode	
	Interfaces	77
2.5.3	Energy Level Alignment at Interfaces	78
2.6	Simulation Methodologies and Result Evaluation:	79
2.6.1	Optical Generation Profile Calculation	79
2.6.2	Electrical Simulation Settings	80
2.6.3	Key Output Characteristics to be Analyzed	81
2.7	Basic Operations of SCAPS 1D	82
2.7.1	Comprehending the operational mechanisms of SCAPS	88
2.7.2	Sampling of the structure in SCAPS	89
2.7.3	How to Reach the Solution in SCAPS	89

2.7.4 SCAPS output parameter extraction	90
3 Simulation Studies of P3HT:IC₆₀BA Organic Solar Cells using inorganic CuI as HTL	92
3.1 Standardization	93
3.2 Optimization of the cell structure ITO/CuI/P3HT:IC ₆₀ BA/ZnONPs/Al	98
3.2.1 Influence of Active Layer Thickness	98
3.2.2 Effect of HTL (CuI) properties	100
3.2.3 Effect of ETL (ZnO NPs) properties	107
3.2.4 Effect of defect density at HTL/Active Layer interface	113
3.2.5 Effect of defect density at Active Layer/ETL interface	115
3.3 Conclusion	117
4 Simulation Studies of P3HT:IC₆₀BA Organic Solar Cells without ETL	119
4.1 Standardization	120
4.2 Optimization of the cell structure ITO/PEDOT:PSS/P3HT:IC ₆₀ BA/Al	121
4.2.1 Effect of Active Layer properties	121
4.2.2 Effect of HTL properties	126
4.3 Effect of temperature	136
4.4 Conclusion	138
5 Simulation studies of P3HT:IC₆₀BA Organic Solar Cells with additional P3HT Layer	139
5.1 Introduction	140

5.2 Optimization of ITO/PEDOT:PSS/P3HT:IC ₆₀ BA/P3HT/ZnONPs/Al	141
5.2.1 Effect of Active Layer Properties	141
5.2.2 Effect of ETL Properties	150
5.2.3 Effect of Additional P3HT Layer Properties	157
5.2.4 Effect of PEDOT:PSS [HTL] Properties	162
5.3 Conclusion	165
6 Simulation studies of P3HT:IC₆₀BA Organic Solar Cells using diverse	
HTL and ETL Layers	166
6.1 Introduction	166
6.2 Standardization	168
6.3 Simulation studies of P3HT:IC ₆₀ A organic solar cell with different HTL	
and ETL Layers	173
6.4 Optimization of ITO/Cu ₂ O/P3HT:IC ₆₀ BA/IGZO/Al	174
6.4.1 Effect of Active Layer Properties	174
6.4.2 Effect of HTL Properties	177
6.4.3 Effect of Electron affinity of ETL	179
6.4.4 Influence of HTL/active layer interface defect density	181
6.4.5 Influence of Active Layer/ETL interface defect density	182
6.5 Conclusion	184
7 Conclusion	185
7.1 Important Findings	186
8 Recommendations and Scope for Future Work	188

List of Figures

1.1 Global energy potential of renewable sources in terawatts(TW) and non-renewable sources showing their estimated reserves in terawatt-years(TW _y)	5
1.2 Basic Structure of a Solar Cell	8
1.3 Basic structure of a silicon solar cell	12
1.4 Basic structure of a thin-film solar cell	13
1.5 Structure of an amorphous silicon solar cell	14
1.6 CdTe Solar Cell	15
1.7 CIGS Solar Cell	16
1.8 Dye-Sensitized Solar Cell	17
1.9 Organic Photovoltaic Cell	18
1.10 Perovskite Solar Cell	19
1.11 Quantum dot solar cell	20
1.12 Tandem Solar Cell	21
1.13 Creation of P-N Junction	23
1.14 Characteristics of P-N Junction	23
1.15 Solar Cell Equivalent Circuit	26

1.16 I-V characteristics of a solar cell	27
1.17 Quantum Efficiency Graph of a Solar Cell	31
1.18 Single-layer OSC	45
1.19 Bilayer OSC	46
1.20 Tandem [Multijunction] OSC	47
1.21 Inverted OSC Structure	48
1.22 Bulk heterojunction OSC	49
1.23 P3HT	52
1.24 MDMO-PPV	52
1.25 PTB7	53
1.26 PC ₆₀ BM	55
1.27 PC ₇₀ BM	56
1.28 IC ₆₀ BA	56
2.1 SCAPS 1D Action Panel	84
2.2 SCAPS 1D Simulation Panel	85
2.3 SCAPS 1D Layer Definition Panel	87
2.4 SCAPS 1D Result Panel	91
3.1 P3HT :IC ₆₀ BA Bulk Heterojunction Polymer Solar Cell Structure	94
3.2 Simulated Polymer Solar Cell Structure	95
3.3 Energy Level Band Diagram	95
3.4 Simulated J-V Curve	97
3.5 Influence of Active Layer thickness	100

3.6 Influence of thickness	102
3.7 Influence of doping density	103
3.8 Influence of Hole Mobility	105
3.9 Influence of Electron Affinity	107
3.10 Influence of Thickness	109
3.11 Influence of Electron Affinity	110
3.12 Influence of Doping Density	111
3.13 Influence of Electron Mobility	113
3.14 Influence of HTL/Active Layer Interface Defect Density	115
3.15 Influence of Defect Density of Active Layer/ETL Interface	117
4.1 Simulated Polymer Solar Cell Structure	121
4.2 Influence of thickness	122
4.3 Influence of defect density	123
4.4 Influence of electron affinity	125
4.5 Influence of carrier concentration	126
4.6 Influence of thickness	127
4.7 Influence of Band Gap	128
4.8 Influence of electron affinity	130
4.9 Influence of effective density of states [CB]	131
4.10 Influence of effective density of states [VB]	132
4.11 Influence of acceptor density of states	134
4.12 Influence of defect density	136
4.13 Influence of temperature	137

5.1 Simulated Polymer Solar Cell Structure	141
5.2 Influence of Thickness	142
5.3 Influence of Bandgap	144
5.4 Influence of Electric Permittivity	146
5.5 Influence of CB density of states	148
5.6 Influence of VB density of states	149
5.7 Influence of Uniform Donor Density of states	150
5.8 Influence of Thickness	152
5.9 Influence of Electric Permittivity	153
5.10 Influence of Conduction Band Density of States	155
5.11 Influence of Valence Band Density of States	156
5.12 Influence of Donor Density	157
5.13 Influence of Thickness	158
5.14 Influence of Band gap	159
5.15 Influence of Electron Affinity	160
5.16 Influence of Valence Band Density of States	162
5.17 Influence of Bandgap	164
5.18 Influence of Electron Affinity	165
6.1 Generalised structure for simulations with various ETL and HTL . .	168
6.2 Comparison of experimental and simulated JV graphs	169
6.3 Energy level diagram of HTL and ETL layers	170
6.4 Energy level diagram of optimised organic solar cell structure	172
6.5 Influence of thickness	175

6.6 Influence of defect density	176
6.7 Influence of electron affinity	178
6.8 Influence of doping density	179
6.9 Influence of electron affinity	180
6.10 Influence of defect density of HTL/Active layer interface	182
6.11 Influence of defect density of Active layer/ETL interface	183

List of Tables

2.1 The input parameters in the layer definition panel	87
3.1 Evaluation of simulated parameters with experimental results	94
3.2 Simulation Parameters used	97
3.3 Optimized Device Performance Parameters	117
4.1 Evaluation of simulated parameters with experimental results	121
6.1 Device parameters of HTL layers	171
6.2 Device parameters of ETL layers	171
6.3 Device Performance of OSC with different combinations of HTL and	
ETL	172
6.4 Optimized material parameters	183
6.5 Comparison of organic solar cell with standard and optimized designs	183

Symbols

I_D	Diode Current
I_o	Reverse Saturation Current
V_T	Thermal voltage
η	Ideality Factor
V_D	Diode Voltage
I_{sh}	Current flowing through shunt resistance
R_{sh}	Shunt resistance
R_s	Series resistance
k	Boltzmann constant
V_{oc}	Open circuit voltage
J_{sc}	Short circuit current
FF	Fill factor
PCE	Power conversion efficiency
n	Concentration of electrons
p	Concentration of holes
n_i	Intrinsic carrier concentration
τ_n	Effective lifetime of electrons
τ_p	Effective lifetime of holes
C_n	Auger recombination coefficient for electrons
C_p	Auger recombination coefficient for holes
R_{Auger}	Auger recombination rate
R_{rad}	Radiative recombination
R_s	Surface recombination
S	Surface recombination velocity

n_{eq}	Equilibrium electron concentration
p_{eq}	Equilibrium hole concentration
ρ	Charge density
ϵ	Permittivity
N_D	Ionized donor density
N_A	Ionized acceptor density
G_n	Generation rate of electrons
G_p	Generation rate of holes
R_n	Recombination rate of electrons
R_p	Recombination rate of holes
J_n	Electron current density
J_p	Hole current density
E	Electric field
q	Electric charge
μ_n	Electron mobility
μ_p	Hole mobility
D_n	Diffusion coefficient for electrons
D_p	Diffusion coefficient for holes
dn/dx	Electron concentration gradient
dp/dx	Hole concentration gradient
E_g	Bandgap
χ	Electron affinity
ϕ	Work function
N_c	Density of states in conduction band
N_v	Density of states in valence band

n	Refractive index
K	Extinction coefficient
Ψ	Electrostatic potential
E_{Fn}	Quasi Fermi level for electrons
E_{Fp}	Quasi Fermi level for holes

Chapter 1

Introduction

This chapter aims to provide a brief introduction to the thesis. The importance of solar energy and the basic principles of how a solar cell works are discussed. The basic concepts of semiconductors that underlie the mechanism of the solar cell are described. The chapter also reviews the history of solar cells with a specific focus on organic solar cells, especially bulk heterojunction devices, and their significance.

1.1 Solar Energy

Solar energy represents the most abundant energy supply on earth and is a sustainable energy source to supply the energy needs of our globe [1]. The sun behaves as a nuclear reactor through thermonuclear fusion, where hydrogen nuclei are turned into helium nuclei under conditions of extreme temperature and pressure. The energy derived from this reaction results as electromagnetic radiation. The blackbody spectrum will yield an approximation of its radiation spectrum at 5,760 K [2]. The photovoltaic effect was discovered by Alexandre-Edmond Becquerel in 1839 in an Electrolyte solution [3].

The improvements made by Bell Labs in 1954 ushered in the present-day solid-state solar cell technology [4]. In view of the current international energy crisis, solar energy and photovoltaic (PV) technology have assumed prominence as a renewable energy form.

More specifically, global energy generation is anticipated to reach approximately 31,525 TWh by 2025, driven by an estimated 92.0 TWh from wind and 63.2 TWh from solar contributions [5]. Fig 1.1 presents a graph that compares the total global energy resources on Earth, highlighting both non-renewable and renewable energy sources [6]. The average solar radiation flux incident on the surface of Earth estimates the amount of energy that is potentially available worldwide (1.2×10^{17} W) [7]. In a single hour of solar radiation from the sun, an adequate amount of energy is generated to meet the total annual energy consumption of the world [8].

Organic photovoltaic (OPV) has emerged as a strong and unique candidate among the forms of solar cell technology being developed. Organic photovoltaics are exciting for their potential to enable cheap manufacturing methods, mechanical flexibility, and compatibility with large-area processing methods.

P3HT:PCBM has for many years been the active layer benchmark for bulk heterojunction (BHJ) organic solar cells, and is used in devices due to its excellent processability, allowing for scalable, solution-based manufacturing, and its ability to form a well-dispersed interdigitated morphology due to thermal annealing that allows for efficient exciton dissociation and charge transport. The main downside to P3HT:PCBM is the low open-circuit voltage (V_{oc}), in the range of 0.55 V to 0.65 V. This phenomenon arises due to the presence of a diminished LUMO energy level

on the acceptor side, which inherently constrains the open-circuit voltage (V_{oc}) in accordance with the principle that V_{oc} is restricted by the energetic parameters of the contact, specifically defined as the energy differential between the donor's highest occupied molecular orbital (HOMO) and the acceptor's LUMO. The low LUMO of PCBM results in quite a sizeable energy loss on charge separation that is responsible for limiting the power conversion efficiency (PCE) of P3HT:PCBM devices to some 3–4%.

The adoption of a P3HT:Indene-C60 Bisadduct ($IC_{60}BA$) blend is thus a direct strategy to mitigate the limitation imposed by the use of PCBM. The model system used to enhance the BHJ organic photovoltaic (OPV) is based on the polymer poly(3-hexylthiophene) (P3HT) as the donor and indene-C60 bisadduct ($IC_{60}BA$) as the fullerene acceptor. The strategy is to use the fullerene acceptor $IC_{60}BA$ instead of PCBM with two different approaches. The LUMO energy level of P3HT: $IC_{60}BA$ is in the order of 0.17 eV above that of PCBM and when applied in PV will increase the open circuit voltage of the P3HT derivatives and accordingly increase the power conversion efficiency of P3HT based devices. Therefore, the transition from PCBM to $IC_{60}BA$ is a deliberate material engineering choice focused on minimizing the energy loss during charge separation, thereby unlocking the full photovoltaic potential of the P3HT donor polymer [9].

The system P3HT: $IC_{60}BA$ exhibited good photovoltaic performance when combined with morphology control (processing conditions). An increase of some 20% in the power conversion efficiency has also been found with additive functionality and ratio of donor:acceptor blend of P3HT: $IC_{60}BA$ [10]. Hence, this combination of

materials has found to be a model system not only for the study of the basic events in photophysics, charge formation, transport and collection to a OPV device, but also to probe the effect of nanoscale morphology on performance. Hence, this thesis aims to provide a full characterisation and optimization of the BHJ organic photovoltaic device using the active layer P3HT:IC₆₀BA and study the link between structure and properties associated with the PV performance of this donor-acceptor system.

The solar constant denotes the comprehensive total solar energy across all wavelengths that is generally received per unit time on a unit area at a distance of one astronomical unit (AU), which represents the mean separation between the Sun and the Earth, possessing a power approximately quantified at 1370 W/m² [11],[12]. A range of phenomena, comprising scattering and absorption, functions to lower the intensity of incoming sunlight as it navigates the Earth's atmosphere. Air mass (AM) serves as a metric for assessing the extent of attenuation caused by the light that is scattered and subsequently absorbed, which can be expressed in equation (1.1).

$$AM = \frac{\text{Optical path length to incident sunlight}}{\text{Optical path length when the sun is incident directly overhead}} \quad (1.1)$$

In addition to the direct rays from light that are scattered as it interacts with the Earth's surface, solar radiation also interacts with the Earth's surface at many angles, creating diffuse radiation. The diffused light intensity variation has been a major issue with photovoltaic systems as the angle of incidence varies with latitude. For an incident power of 1000 W/m² and an elevation angle of 42 degrees, the air mass value is known as AM 1.5 G (Air Mass 1.5 Global Spectrum), which contains both direct and diffused radiations [2].

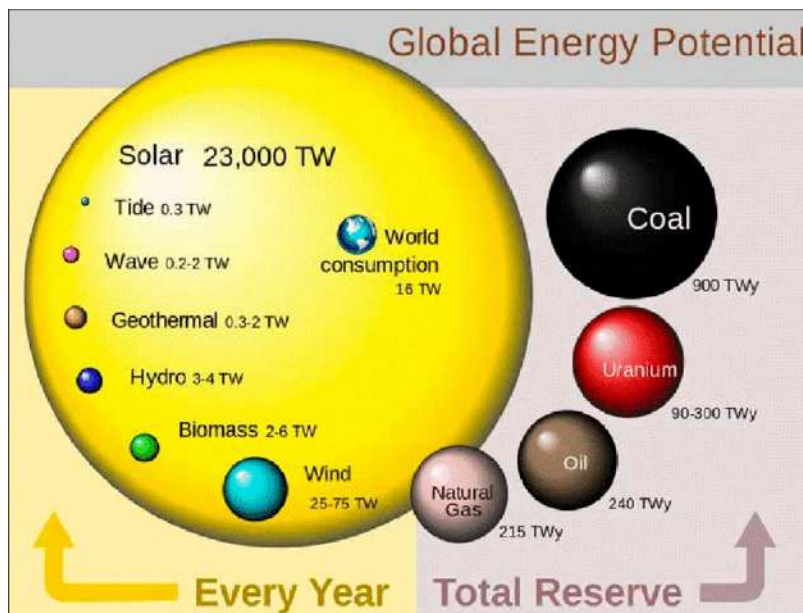


Figure 1.1: Global energy potential of renewable sources in terawatts(TW) and non-renewable sources showing their estimated reserves in terawatt-years(TWy)

Despite recent progress, carbon dioxide emissions from fossil fuels are on track to hit another all time high in 2025- a reminder of the challenges posed by fossil fuel burning. After the figure of 37.8 gigatons (Gt) CO_2 for 2024 (up 0.8% from 2023), the Global Carbon Budget 2025 expects them to increase by around 1.1% in 2025 for a total of around 38.1 Gt of fossil CO_2 emissions. The resultant increase in atmospheric carbon dioxide concentrations peaked at 422.5 ppm—50% higher than pre-industrial concentrations and had climate-related consequences [13].

Fossil fuels and fossil fuel extraction also pose considerable problems regarding resource depletion. Current global estimates suggest that current reserves will last about 50 years for oil, 52 years for natural gas, and 139 years for coal, assuming current consumption is maintained. Fossil fuel markets are also extremely economic

volatility markets, as evidenced by natural gas prices tripling as a result of the Russian invasion of Ukraine in 2022, and the projected natural gas prices in the U.S. are expected to double from 2024 to 2026. These result in excessive destabilizing, economic volatility, markets that threaten energy security [14].

Renewable energy, solar energy in particular, is a viable solution for all of these scenarios. Solar energy is essentially inexhaustible, and it is abundantly available worldwide [15]. Further, solar energy is increasingly becoming a beneficial economic case, where solar prices are down to prices that fell to \$876/kW from \$2,652/kW in 2014, and this zero-century pricing will follow Wright's Law again; for every doubling of global capacity, the price will decline by 20%. [16].

The historical continuous improvement in technology and pricing has resulted in structural/behavioural changes in the global energy markets. By year end 2025, virtually all newly installed renewable energy capacity will be lower cost than fossil fuel use. Also, solar and wind will be the lowest-cost new electricity generation mix in the near future [14]. Much has been said, as global solar photovoltaic (PV) installations are climbing by 29% from the previous year [17]. Solar energy comprises over 80% of all new renewable energy capacity in 2025. Now, what is intriguing is the solar energy transition from alternative new electricity generation to economic predominance—this solves environmental degradation, the resource depletion horizon line, and economic volatility issues with fossil fuel dependence, and allows for abundant, clean, and increasingly more affordable energy for global development.

1.2 Fundamentals of Photovoltaics

Solar cells (photovoltaic cells) are a direct, instantaneous means of converting solar energy into electricity. With respect to the photovoltaic effect, the conversion efficiency of a solar cell is fundamentally dependent upon the action of light photons upon semiconductor materials. The action gives rise to pairs of electrons and holes, and produces electrical current in the photovoltaic cell. The electrical production in a photovoltaic cell is brought about by virtue of the fact that there is charge carriers, that is, electrons and holes, as in the general form of structure shown in Fig. 1.2, and the basic structure of components is classified under:

- Light Absorption
- Charge Carrier Generation
- Charge Carrier Separation and Transport
- Recombination

1.2.1 Absorption of Light

When sunlight enters a semiconductor, the energy of a photon transfers to a valence band electron which can escape from the valence band and go to the conduction band. The band gap of every semiconductor determines whether a photon can excite the valence band electron and generate electron-hole pairs.

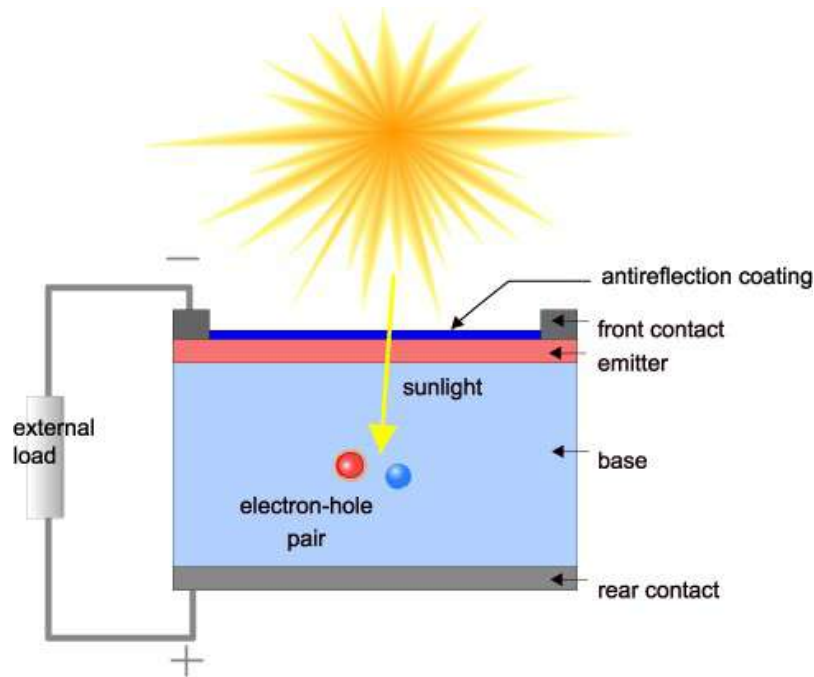


Figure 1.2: Basic Structure of a Solar Cell

1.2.2 Charge Carrier Generation

The absorption of photons produces free charge carriers or holes in the valence band and electrons in the conduction band. The generation mechanism is located in the region very close to the surface of the cell since practically all of the light is absorbed here. The charge carriers drift through the semiconductor and take an active part in the generation of an electric current.

1.2.3 Separation and Movement of Charge Carriers

After creation, the free electrons must be separated from the holes to prevent recombination and to establish light-losing bonds which would be a non-conservation of energy. The electric field established by the p-n junction separates these charges.

In the n-type region where free electrons are a greater concentration, the electrons are free. In the p-type region, where the holes are a greater concentration, the holes are free. This movement automatically produces some electrical energy, acting as an electrical current through an external circuit.

1.2.4 Collection of Charge Carriers

Metal contacts on the solar cell serve to collect the electrons moving from the n-type layer, thus allowing for the use of the electrical current that is generated. Those electrons act to power electric devices as they leave the solar cell and move through a connected circuit. A photovoltaic cell works like a battery in the sense that it develops a potential difference through the accumulation of electrons and holes [18].

1.2.5 Recombination

Although separation of charge carriers is intended, recombination can occur. Charge carriers (electrons) can also recombine with holes prior to collection. The result of this process is losses in solar cells. To improve cell performance, recombination processes must be minimized through design and material optimization. The time that is available for separated charge carriers to exist is related to the minority carrier lifetime.

1.3 The Evolution of Solar Cell Technology

Alexandre-Edmond Becquerel, a physicist, found the photovoltaic effect in 1839 and invented a new type of solar cell while working on an electrochemical cell. He found

out that some materials made a small electric current when they were lit up [19]. Selenium, which exhibits photoconductivity, indicates that exposure to light alters electrical resistance [20]. This finding led to the creation of the first solar cells.

In 1883, American Charles Fritts made the first real solar cell by putting a thin layer of gold on selenium. Even though it's efficiency is 1%, it showed that sunlight can be used as an energy source [21]. Albert Einstein's description of how light energy can free an electron from a material is a groundbreaking work on the photoelectric effect. So, this is why solar energy conversion is so important. It won a Nobel Prize in 1921 [22]. Russell Ohl found the p-n junction in silicon at Bell Laboratories. This was the key to all modern solar cells. His discovery was patented in 1941 and helped silicon solar cells become better [23]. The work of Daryl Chapin, Calvin Fuller, and Gerald Pearson at Bell Laboratories in 1954 on the first silicon solar cell was a big step forward in photovoltaic technology. It had a conversion efficiency of 6%, which was amazing compared to selenium solar cells [24].

The U.S. also launched Vanguard 1 in 1958, the first satellite to use a solar cell. This was the start of solar power being used in space [25]. In 1982, Hesperia, California, became the first place to generate solar energy on a large scale. In 1985, scientists at the University of New South Wales (UNSW) made a big step forward by making a silicon solar cell that was 20% efficient [26]. Perovskite solar cells became commercially viable in 2006 [27].

In 2016, multi-junction solar cells had lab efficiencies of over 40%. As of the year 2023, a total of six junction cells, along with highly efficient commercial cells, have been developed [28]. The cost of generating electricity from solar power dropped

so dramatically between 2010 and 2020 that it fundamentally changed the energy market, making solar a much more competitive and mainstream option. The bulk heterojunction photovoltaic cell idea [29] has led to a new type of organic optoelectronics. In the 2000s, organic solar cells were made with fullerenes, non-fullerene acceptors, and polymers as donor materials [30]-[31]. The tandem concept, which allows several solar cells to be connected in parallel or in series, opened a new area of research for photovoltaics.

1.4 Growth of solar cell Technology

1.4.1 First-Generation Solar Cells

First-generation solar cells, also known as crystalline silicon (c-Si) solar cells, are the most common and, indeed, conventional in photovoltaic technology, as they have existed since the 1950s thanks to their high efficiency, reliable performance, and longevity [32]. They incorporate monocrystalline, polycrystalline and multicrystalline silicon solar cells.

Monocrystalline silicon photovoltaic cells are derived from a single, continuous crystalline lattice of silicon, which is usually manufactured using the Czochralski method. There are fewer grain boundaries, which disrupt electron mobility and thus the greater efficiency (18-22%) of these cells as opposed to the polycrystalline [33]. The latter type of cell consists of a silicon melt of several silicon crystals and further cooling of such a crystal in a mould. These cells have a bluish, tessellated appearance that, by itself, reveals key information about their nature. The efficiency is somewhat lower than the case of monocrystalline cells, as a greater loss of electron

mobility occurs at the grain boundaries (15-18% efficiency) [34]. The process is straightforward and generates minimal waste, thereby facilitating an economical fabrication of polycrystalline cells, which subsequently enhances their worth. The structure of a silicon solar cell is given in Fig. 1.3.

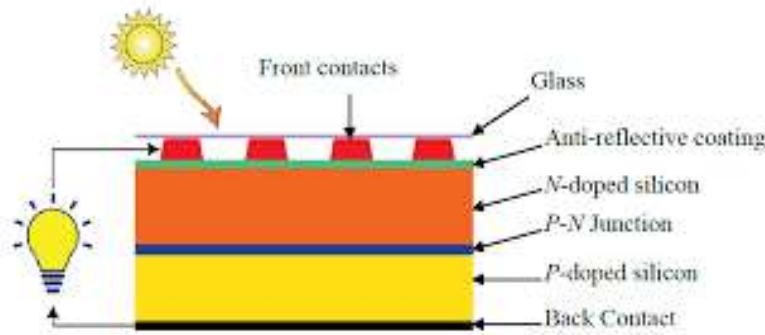


Figure 1.3: Basic structure of a silicon solar cell

1.4.2 Second-Generation Solar Cells

Second-generation solar cells based on thin-film technology represent a major improvement over first-generation crystalline silicon cells [34]. These solar cells contain thin layers of semiconductor materials with many advantages and uses. The thin layers of semiconductor materials, only a few microns thick, are the basis for the lightweight, flexible, low-cost nature of solar cell films. The almost new concept allows for smaller amounts of materials being used, which may mean lower manufacturing and production costs than the first-generation technologies, and will make them suitable for the large-scale future production of solar energy. Compared to the thick crystalline silicon solar cells, which rely on thick silicon wafers, the thin film solar cells

are much easier to work with and allow for the easy deposition of thin lightweight layers of semiconductor materials directly on a variety of substrates, which include glass, plastic, metal etc. The essential structure of the thin film solar cells is shown in Fig 1.4.

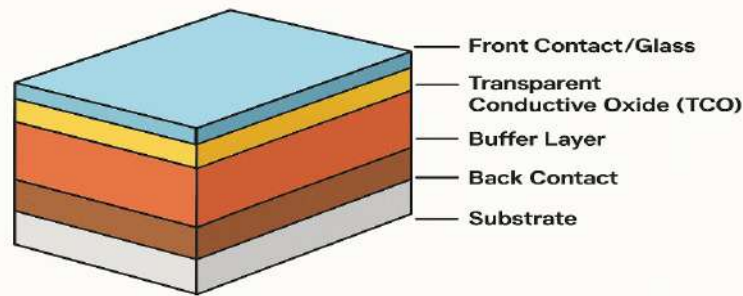


Figure 1.4: Basic structure of a thin-film solar cell

1.4.2.1 Amorphous Silicon (a-Si) Solar Cells

Amorphous silicon (a-Si) cells are made from non-crystalline silicon which has the advantage of more flexibility, lower production expenses, lesser weight, and better performance in relatively poor lighting. The chief disadvantages of this type of material are lower efficiency (6% - 12%) [35] compared to crystalline silicon (20%) and photodegrading. Thus, while a-Si has a broader band gap of 1.7 eV, say, compared to that of crystalline (c-Si) cells with a band gap of 1.1 eV, it has better absorption in the visible spectrum. Thus, amorphous silicon thin films could take the place of c-Si, which would give rise to a lighter and perhaps more flexible type of solar cell. Amorphous silicon solar-cell structure is represented by Fig. 1.5.

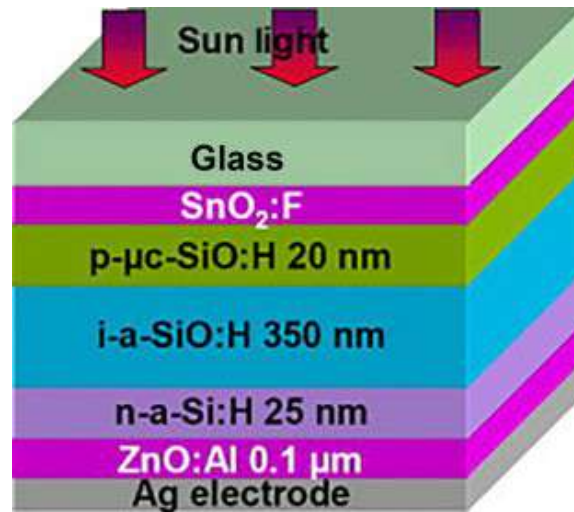


Figure 1.5: Structure of an amorphous silicon solar cell

1.4.2.2 Cadmium Telluride (CdTe) Solar Cells

CdTe, or cadmium telluride, is probably the most famous thin film technology, best known for its low cost and high efficiency. Presently, CdTe has the majority of the market share of all thin film technologies. At the same time, the toxicity of cadmium and the great scarcity of tellurium present serious problems with large scale sustainable production. The main advantages of CdTe solar cells are the high efficiency (18% - 22%) [36], low manufacturing costs and great durability and longevity, as shown in Fig1.6.

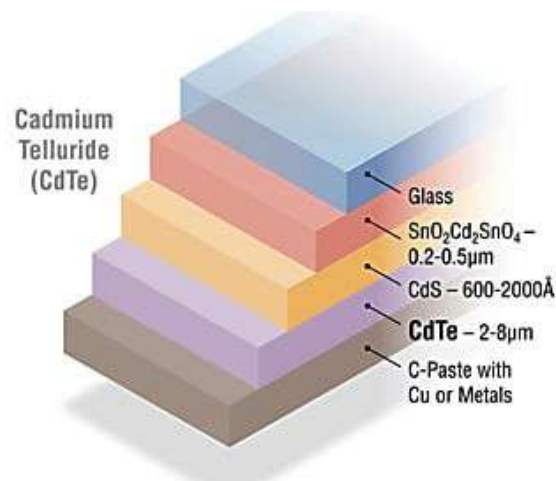


Figure 1.6: CdTe Solar Cell

1.4.2.3 Copper Indium Gallium Selenide (CIGS) Solar Cells

CIGS solar cells are the highest performing of the thin film solar cells even greater than that of the efficiencies of commercial crystalline silicon. The efficiency value of CIGS solar cells surpasses organic photovoltaic cells (OPV) at a value of 22.6%. CIGS cells have the benefit of being tuned for the band gap of the semiconducting material by varying the ratios of indium: gallium and selenium: sulfur. The study reveals a significant potential for the optimization of band gaps, indicating that these cells may be employed in tandem cell configurations in future. The worst attribute of CIGS is cost, which can be attributed to the use of indium and the special assembly of the integration. The optimization of CIGS itself is also quite difficult, based on the area of stoichiometry and the several step-by-step processing, which can be a hindrance to the scalability commercially. A CIGS solar cell is depicted in the illustration of Fig.

1.7

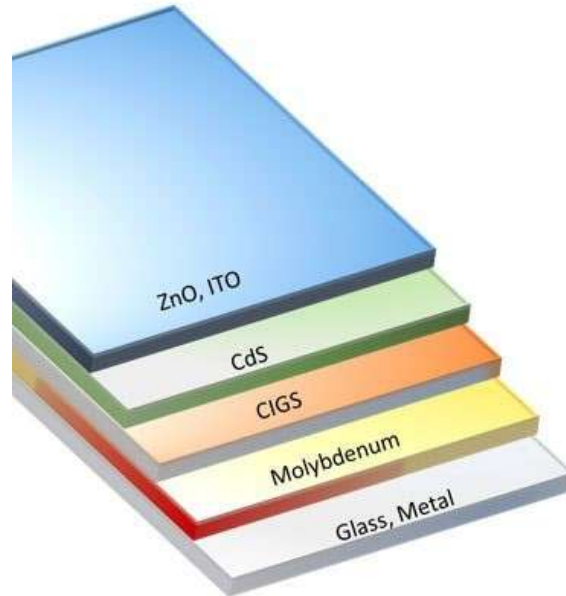


Figure 1.7: CIGS Solar Cell

1.4.3 Third-Generation Solar Cells

Third-generation solar cells were discovered to overcome the shortcomings of 1st- and 2nd-generation solar cells. The materials used in this generation of solar cells include organic polymers, perovskites, quantum dots, and dye-sensitized materials to increase efficiency, reduce production costs, and improve flexibility [37]. The main cutting-edge technologies represented in third-generation solar cells are dye-sensitized solar cells (DSSCs), organic photovoltaic cells (OPVs), perovskite solar cells (PSCs), quantum dot solar cells (QDSCs) and tandem multijunction solar cells.

1.4.3.1 Dye-sensitized solar cells [DSSCs]

In dye-sensitized solar cells (DSSCs), a solar energy-absorbing photoactive dye generates electrons, which are transferred through a nanostructured layer of titanium dioxide (TiO_2) [38]. The system architecture includes a photoanode, which commonly uses a semiconductor surface coated with dye molecules that can absorb light. The photonic energy excites the electrons in the dye, moves them to the semiconductor, and collects them as an electric current. The foundation of these systems uses a liquid electrolyte that contains organic dye absorbers, such as the N719 dye, and this technology utilizes abundant solution-processable materials and allows for the possibility of creating flexible and transparent electronic devices. An example of a dye-sensitive solar cell is shown in Fig 1.8.

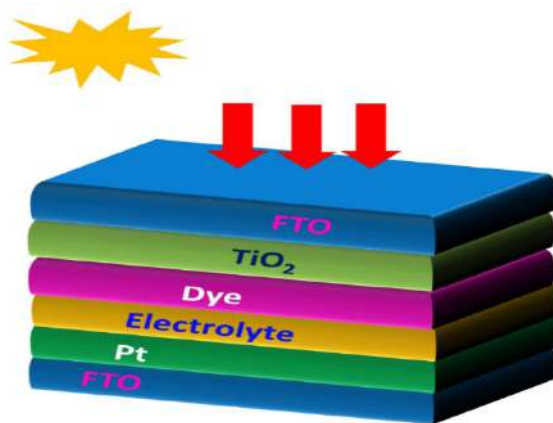


Figure 1.8: Dye-Sensitized Solar Cell

1.4.3.2 Organic Photovoltaic Cells (OPVs)

Organic compounds, or polymers, that operate as the active photovoltaic material make up OPVs. To create an electric current, these cells use light absorption to

create excitons, or bound electron-hole pairs, which are subsequently separated at donor-acceptor interfaces [31]. They may be printed, are flexible, and are lightweight. Generally speaking, they have shorter operational lifetimes and worse efficiency than their inorganic equivalents. The structure of an organic solar cell is given in Fig 1.9.

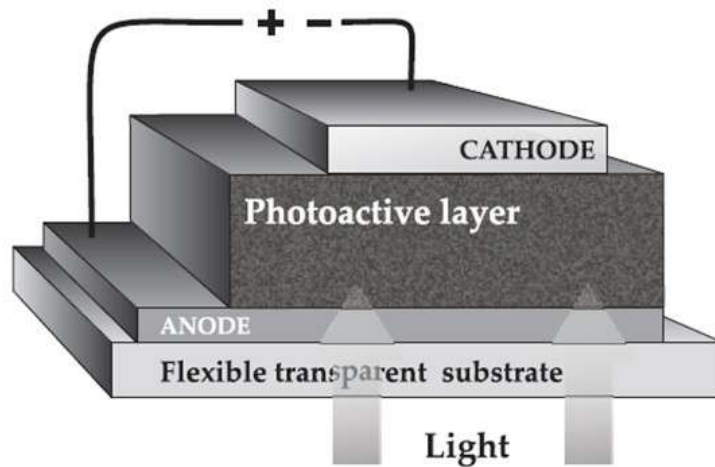


Figure 1.9: Organic Photovoltaic Cell

1.4.3.3 Perovskite Solar Cells (PSCs)

Perovskite solar cells (PSCs) rely on hybrid organic-inorganic lead halide perovskites (e.g. $CH_3NH_3PbI_3$) as the primary light-absorbing material that has high charge-carrier mobilities [39]. Within the active layer of these devices, materials of the perovskite family are used for light harvesting. The materials exhibit high light absorptive properties as well as moderate charge transport efficiencies. Their tunable band gaps facilitate the absorption of a wider range of the solar light spectrum in comparison to silicon photovoltaics. Over the last several years, PSCs have shown

phenomenal improvement in their efficiencies, and some corresponding devices have demonstrated PCEs over 25%. These efficiencies, however, occur at the cost of the materials' low stability in the structure of their devices, especially in high moisture environments, which may inhibit their prolonged use. The cell structure of PSCs is represented in Fig [1.10](#).

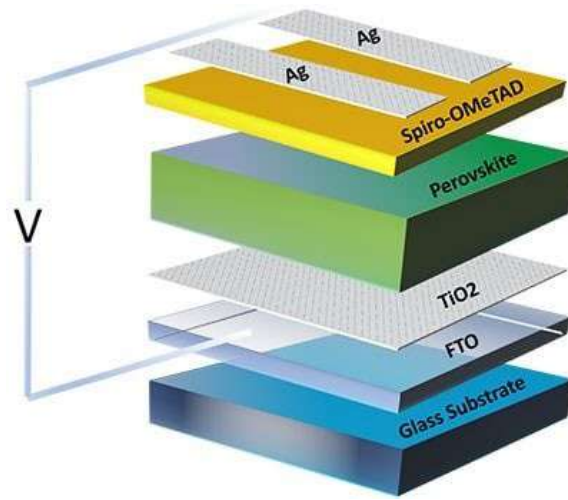


Figure 1.10: Perovskite Solar Cell

1.4.3.4 Quantum Dot Solar Cells (QDSCs)

Quantum Dot Solar Cells (QDSCs) utilize semiconductor nanocrystals (i.e., CdSe, PbS) to tune band gaps using quantum confinement effects [\[40\]](#). They use semiconductor quantum dots as light-absorbing entities. The size of nanocrystals can be tuned to selectively absorb certain wavelengths of light, meaning a wide range of solar radiation can be harnessed. The structure of a quantum dot solar cell is illustrated in Fig. [1.11](#).

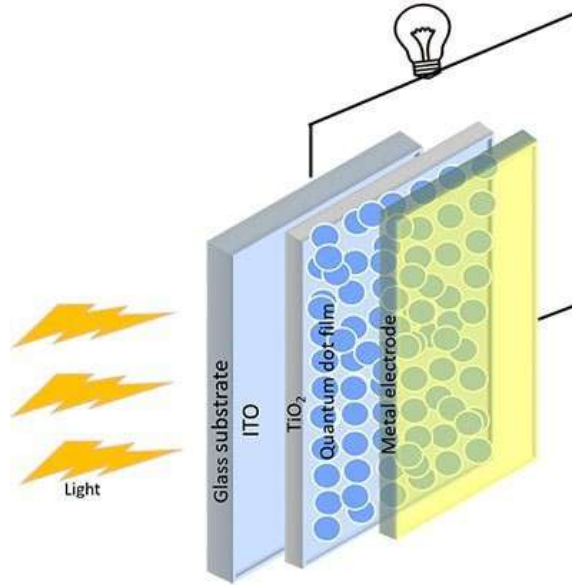


Figure 1.11: Quantum dot solar cell

1.4.3.5 Tandem and Multi-Junction Solar Cells

Tandem solar cells (TSCs) utilize two or more layers of PV materials with differing band gaps to effectively absorb a larger amount of solar irradiance [41]. Each absorber layer is specialized to absorb a specific region of the sunlight spectrum and contribute to the overall conversion efficiency. By stacking different absorber layers, we can theoretically utilize different wavelengths and thus bypass the 60% efficiency barrier, exceeding the Shockley-Queisser limit in principle. Fig 1.12 shows the tandem solar cell structure.

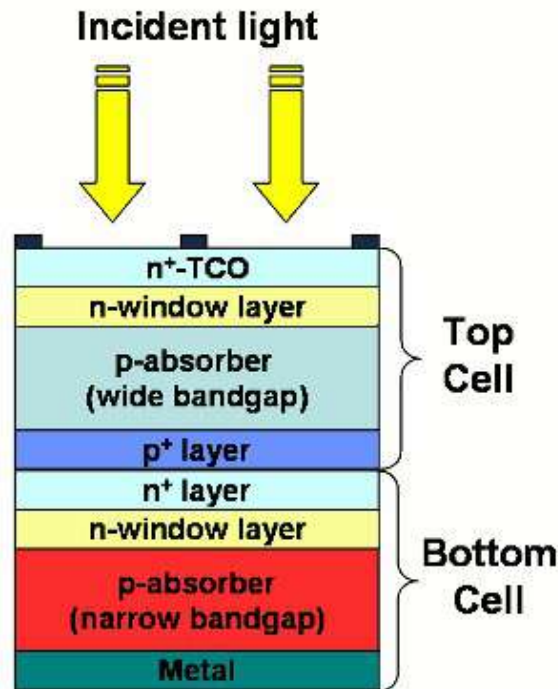


Figure 1.12: Tandem Solar Cell

1.5 Physics of Solar Cells

1.5.1 The operation of a p-n junction

Fundamental to the knowledge base of semiconductor physics and electronics is that the function of p-n junctions is based upon diffusion. As in both types of semiconductive materials with charged carriers, n-type, which has primarily electrons present, corresponding to the negatively charged carriers, the implementation of these carriers results from the inclusion of a pentavalent element. The p-type semiconductor primarily contains holes, or positively charged carriers, which are introduced by including a trivalent element. As far as holes are concerned, the holes on the p-side

are to diffuse to the n-side, and the electrons from the n-side diffuse into the p-type side. This results in a depletion region at the junction, which has a fixed number of ionized donor and acceptor atoms, which has been depleted of free charge carriers. For current to flow, the potential barrier produced has to be overcome, which is equal to the value of the electric field in the depletion region. Fig 1.13 indicates the formation of the p-n junction.

The conductance of a p-n junction is different under bias conditions. Under forward bias conditions, the negative terminal of the direct current power supply connects to the n-region, and the positive terminal connects to the p-region. This allows the charge carriers to migrate more easily to equalize the potential barrier at the junction. Conduction of current becomes evident and indicates that the diode is in a conductivity condition as the voltage increases beyond some threshold limit, which is approximately 0.7 volts with respect to a silicon diode. Under reversed bias, the terminal that connects to the positive side connects to the p-side, and the n-side is connected to the negative terminal. This increases the depletion region and increases the potential barrier produced, which diminishes the amount of current the junction can support. The only current flow that takes place through the junction is the reverse saturation current or leakage current, which flows as a result of minority carriers.

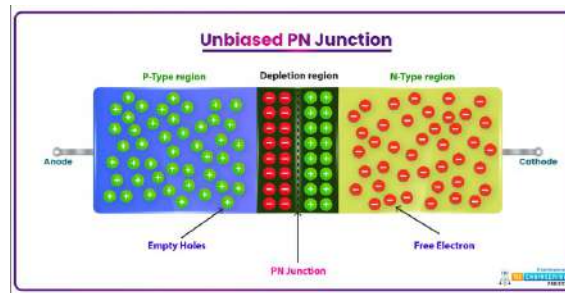


Figure 1.13: Creation of P-N Junction

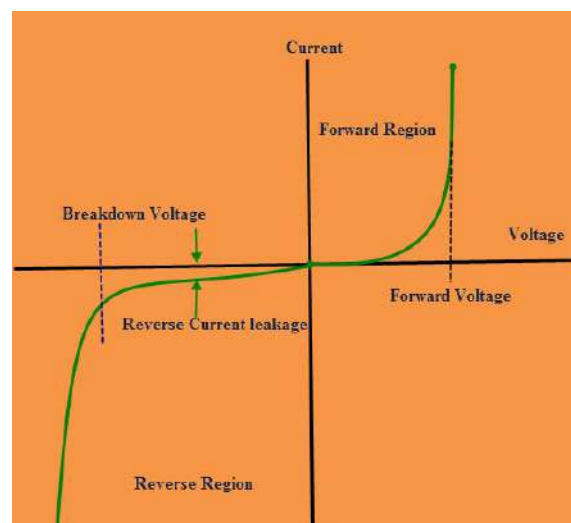


Figure 1.14: Characteristics of P-N Junction

The forward-bias region of a p-n junction diode's current-voltage (I-V) characteristics shows that, above the threshold voltage, the current increases significantly, resulting in a steep upward slope on the graph. The current in the reverse-bias region is quite small until breakdown, when the majority minority carrier flow causes it to increase dramatically. Fig 1.14 displays the I-V characteristics of a p-n junction semiconductor diode.

Current and voltage have a nonlinear connection, as evidenced by the forward bias

region's exponentially rising I-V curve. By expressing the current flowing through the diode as a function of the applied voltage, the diode equation provides a mathematical formulation of this behavior. In general, the following formula provides the diode current.

$$I = I_o \left[\exp \left(\frac{V_D}{\eta V_T} \right) - 1 \right] \quad (1.2)$$

where V_T stands for the thermal voltage, η for the ideality factor, V_D for the diode voltage, I for the diode current, and I_o for the reverse saturation current.

1.5.2 P-N junction under illumination

A photon entering the semiconductor will be absorbed if it has energy at least equal to that of the band gap of the material. This interaction promotes an electron from the valence to the conduction band, producing an electron-hole pair. The built-in field of the p-n junction separates these charge carriers and directs the electrons to the n-side and the holes to the p-side. As electrons accumulate at the cathode and holes at the anode, a potential difference arises. If a load is then connected between the two terminals, the circuit is completed, and current will flow, producing an output voltage.

1.5.3 Solar cell equivalent circuit

Photovoltaic cells, which convert photon energy into electrical energy, are the fundamental components of photovoltaic systems. To properly analyze and ultimately predict their operation in electrical circuits, it is necessary to model solar cells using an equivalent circuit. The equivalent circuit model helps understand the interrela-

tionships of the solar cell and shows how all components operate together to provide electrical energy. The standard equivalent circuit model for a solar cell consists of a diode, a series resistance, a shunt resistance, and an ideal current source. The current resulting from absorbed light energy is associated with the intensity of solar radiation on the solar cell as represented by the ideal current supply [42]. In parallel with the ideal current supply, the diode allows current to flow in the forward direction and blocks it in the reverse direction [43].

The series resistance is analogous to an internal resistance to sinusoidal current, serving to limit losses due to inefficient charge transport through the layers of the solar cell and providing resistance that allows the current from the cell to transfer to the electrical load without loss of power. The series resistance is critical for any solar cell, as it allows the charge produced by the solar cell to cross the boundary to the electrical load without losing power. Lastly, the shunt resistance is usually associated with the semi-conduction diffusion from impurities or defects in the solar cell material to the p-n junction. The effect of shunt resistance is low due to normal loads or a possible low level of piping to the junction defect in the material. Solar cells with high shunt resistance values are beneficial because they can cause significant power loss and hinder overall efficiency. The following equation describes the combined total current (I) supplied to the solar cell in equivalent current supply mode.

$$I = I_L - I_D - I_{sh} \quad (1.3)$$

where I_D is the current flowing through the diode, I_{sh} is the current flowing through the shunt resistance, and I_L is the benefited current from incident light. This

is a representation of the net current available at the output terminal, which includes the contributions and losses from the individual elements. A solar cell will generate I_L when under light exposure, and the I_L is reduced based on the shunt resistance and diode resistance. The diode is important since it limits the output voltage of a solar cell and begins to conduct when the output voltage exceeds a certain value. The existence of the shunt resistance is a detriment to the efficiency of the solar cell by conducting leakage current and diverting some of it from the load. The solar cell equivalent circuit is shown in Fig 1.15.

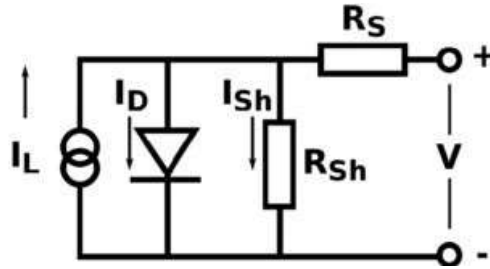


Figure 1.15: Solar Cell Equivalent Circuit

V_D can be written as

$$V_D = V + IR_s \quad (1.4)$$

As already mentioned in section 1.5.1, the diode equation is given by

$$I_D = I_o[\exp(V_D/\eta V_T) - 1] \quad (1.5)$$

The shunt resistance current can be written in the form

$$I_{sh} = V_D/R_{sh} \quad (1.6)$$

Thus, the modified diode equation becomes

$$I = I_L - I_0 \left[\exp \left(\frac{V_D}{\eta V_T} \right) - 1 \right] - \frac{V + IR_s}{R_{sh}} \quad (1.7)$$

The solar cell's I-V characteristics are essentially the photovoltaic device's superposition in both dark and light environments. Fig 1.16 shows the graphical representation. Under dark conditions, a solar cell's I-V properties are the same as those of a p-n junction semiconductor diode. The I-V curve shifts into its fourth quadrant when the device is illuminated, and the magnitude of the shift varies with the intensity of the incident light on the solar cell.

1.5.4 Basic solar cell output parameters

This section defines the fundamental output parameters that describe a solar cell's performance

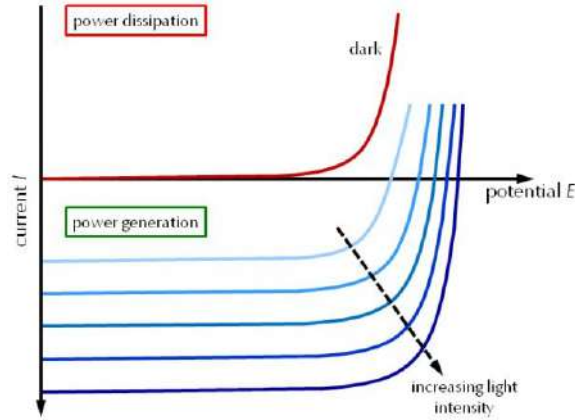


Figure 1.16: I-V characteristics of a solar cell

1.5.4.1 Open circuit voltage [Voc]

The Open Circuit Voltage (Voc) is the maximum voltage across the terminals of an illuminated solar cell with no load on its circuit - that is, with zero current flowing through it. This value is vital to most aspects of photovoltaic system design and performance analysis, and is taken into account even in lifecycle assessments, e.g., for carbon footprint calculations. The actual working voltage under load conditions is always lower than the Voc. Voc is variable, but it is constrained by several other parameters. Firstly, it is inversely proportional to temperature: the hotter the cell, the lower the voltage. On the other hand, it is directly proportional to the intensity of the light incident on the active cell surface; more photons mean more charge carriers. The maximum theoretical Voc is, however, limited by the semiconductor's material properties, its microstructure, and the work functions of the electrodes, as indicated below

$$V_{oc} = \eta kT / q \ln[I_L / I_o] + 1 \quad (1.8)$$

where q is the electronic charge, T is the temperature, η is the ideality factor, k is the Boltzmann constant, I_L is the photocurrent, and I_o is the saturation current.

1.5.4.2 Short Circuit Current [Jsc]

The short circuit current (Jsc) is the peak current a photovoltaic cell can generate in a short circuit while the terminals are shorted together with no load. This is an important figure of merit for evaluating photovoltaic cell performance, and it is one of the main figures of merit for solar cell characterization. Jsc is helpful in understanding

the performance figures and efficiencies of a solar cell; it tells you the amount of energy a solar cell can generate under standardized conditions and acts as a metric for comparing solar cell efficiencies. For example, a larger magnitude of short circuit current shows an increased ability to convert solar energy to electrical energy. These factors—charge-carrier collection efficiency, light intensity, cell area, incident light spectrum, and film properties—work together to determine the short-circuit current.

1.5.4.3 Fill factor [FF]

The fill factor (FF) is an important parameter for assessing photovoltaic cell performance and applicability. It signifies how close the solar cell is to the theoretical maximum power. The fill factor is the ratio of a photovoltaic cell's peak power to the theoretical power calculated by $V_{oc} \times J_{sc}$. A higher fill factor indicates better performance, implying that the solar cell is operating more efficiently relative to the maximum possible efficiency. Formally, the fill factor is quantitatively described as follows:

$$FF = P_{max}/V_{oc} \times J_{sc} = V_{max} \times I_{max}/V_{oc} \times J_{sc} \quad (1.9)$$

where P_{max} is the highest power output that can be generated from the solar cell. V_{max} and I_{max} are the maximum voltage and current that are generated, respectively. A lower fill factor indicates possible inefficiencies that can arise from design flaws, manufacturing inadequacies, or poor environmental conditions. In contrast, a higher fill factor indicates that the solar cell is functioning in an optimal environment with minimal losses. Thus, to advance solar technology and enhance the overall efficiency

of a photovoltaic system, it is essential to increase the fill factor.

1.5.4.4 Power Conversion Efficiency [PCE]

Power conversion efficiency (PCE) is a primary measure of a solar cell's performance in converting the incoming solar radiation into usable electric energy. Power Conversion Efficiency (PCE) is the ratio of the electrical power generated by a solar cell to the amount of solar power incident upon it. PCE increases the fraction of incident solar energy delivered as electrical power, and PCE is vital for photovoltaic (PV) systems to be efficient. It is expressed as

$$P_{out}/P_{in} = V_{oc} \times J_{sc} \times FF/P_{in} \quad (1.10)$$

The power conversion efficiency is enhanced by minimizing energy loss. The efficiency of power conversion is affected by losses due to reflection. The use of anti-reflective coatings on photovoltaic cells, and the implementation of maximum power point tracking (MPPT) techniques, which regulate the load connected to the photovoltaic cell ensures optimal operational efficiency. (QE) of a solar cell is a key factor in determining the degree to which incident photons can produce charge carriers, mainly electrons. Thus, it is an important number for purposes of determining the degree to which a solar cell converts solar radiation into electrical energy potential and represents the upper limit of efficiency for photovoltaic solar cells (total efficiency).

The quantum efficiency is defined as the total number of electrons produced from the solar cell divided by the total number of light photons that interact with the solar cell. The higher the QE, the better the light-to-electric-current conversion efficiency,

usually expressed as a decimal. The QE of a photovoltaic solar cell is influenced by many factors. First and foremost, material properties: every semiconductor material has varied bandgap energies, which leads to varied potential absorption of photons and production of electrons. For example, optimized bandgap materials could enhance QE via increased photons and thus increased area of solar radiation. Finally, significant advancements in thin film processing and cell design enhance the bottom line, giving increasing light absorption and carrier collection. Fig 1.17 illustrates the quantum efficiency graph of a solar cell.

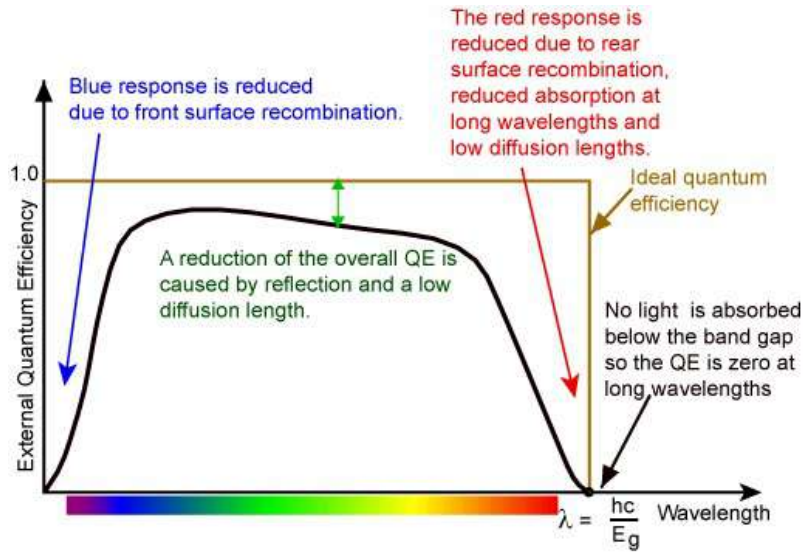


Figure 1.17: Quantum Efficiency Graph of a Solar Cell

1.5.5 Solar cell recombination mechanisms

Recombination in the semiconductor materials of photovoltaic applications is an important aspect of performance. Recombination is defined as the act of electron-hole pairs generated by photovoltaic absorption of solar energy recombining prior to

forming a flow of electric current, representing a loss of efficiency of the solar cell. To improve upon solar cell design and efficiency, we need to identify the different recombination mechanisms that may be present. This section will provide some of the relevant recombination mechanisms.

1.5.5.1 Shockley-Read-Hall Recombination

Shockley-Read-Hall (SRH) recombination, which is often known as defect-assisted or trap-assisted recombination, occurs when we have some impurities or defects within the bandgap of a semiconductor that introduce new energy levels. In this two-step process, an electron gets trapped at a defect level and eventually recombines with a hole in the valence band. These defects present additional routes for the recombination of electrons and holes that do not emit energy. This additional route ultimately has a large impact on the overall recombination rate. High defect density of materials such as silicon speeds up the recombination process and reduces carrier lifetimes, which can severely limit efficiency for solar cells. Therefore, improvements to solar cell efficiency must consider ways to reduce defect concentrations and improve crystalline structure quality. The general equation for the SRH recombination rate can be given by:

$$R_{SH} = (np - n_i^2) / [\tau_p(n + n_t) + \tau_n(p + p_t)] \quad (1.11)$$

where n and p are the concentrations of electrons and holes, n_i is the intrinsic carrier concentration, τ_n is the effective lifetime of electrons, τ_p is the effective lifetime of holes, and n_t and p_t refer to the trap defect concentrations [44].

1.5.5.2 Auger Recombination

The functioning of solar cells relies on a non-radiative recombination process called Auger recombination, where the energy lost during the recombination of an electron and hole is transferred to another carrier (electron or hole) instead of thermal energy release or light emissions.

The excess energy is transferred such that an additional electron (or hole) is promoted to a higher energy level instead of emitting a photon. Over time, the excited carrier eventually loses this excess energy through phonon interactions. Below is a mathematical relationship for the Auger recombination rate

$$R_{Auger} = C_n n^2 p + C_p p^2 n \quad (1.12)$$

The term R_{Auger} refers to the rate of recombination in carriers per unit volume per second, while C_n and C_p represent the Auger recombination coefficients for electrons and holes, respectively (with units of cm^6/s). Here, n denotes the electron concentration (in cm^{-3}), and p indicates the hole concentration [\[45\]](#).

1.5.5.3 Radiative Recombination

Through radiative combination, a photon is generated from the direct interaction of a conduction band electron and valence band hole. Radiative recombination is less prevalent in silicon solar cells because of their indirect bandgap; however, it can be an important consideration for applications in opto electronics that can benefit from it. Because many materials can exploit radiative efficiency to improve performance,

the effectiveness of radiative recombination is particularly relevant for multi-junction solar cells. The equation for the rate of radiative recombination is as follows:

$$R_{rad} = B \cdot n \cdot p \quad (1.13)$$

where R_{rad} represents the radiative recombination. The radiative recombination rate is denoted by B , which is the coefficient specific to the material and can vary based on factors such as temperature and doping levels; n represents the concentration of electrons in the conduction band, while p indicates the concentration of holes in the valence band [46]. Recombination is closely related to the semiconductor interface, which often has a higher concentration of defects than the bulk.

Defect states, especially larger ones, tend to have a higher probability of recombining minority carriers and therefore contribute to a lower effective minority carrier lifetime. Surface passivation techniques seek to reduce surface recombination by reducing the density of dangling bonds and other surface defects. High-quality surface passivation will improve the efficiency of solar cells by increasing both minority carrier lifetime and device efficiency. The surface recombination expression is given as

$$R_s = S \cdot (n - n_{eq}) \cdot (p - p_{eq}) \quad (1.14)$$

Where R_s is the surface recombination rate, S is the surface recombination velocity, which is a material-specific parameter that quantifies the rate at which carriers recombine at the surface, n is the concentration of electrons at the surface, n_{eq} is the equilibrium electron concentration, representing the concentration of electrons

under steady-state conditions, p is the concentration of holes at the surface, and p_{eq} is the equilibrium hole concentration [47].

1.5.6 Basic Semiconductor Equations

A group of basic semiconductor equations that describe the movement of charge carriers inside the device governs the physics of the solar cells. These basic equations, which include Poisson's equation, the continuity equations, and transport equations (also known as drift-diffusion equations), are used to understand and model the electrical behaviour of solar cells. These fundamental equations are used by many device simulators and can be solved numerically; under certain approximations, they can even be solved analytically. Let us now discuss these equations.

1.5.6.1 Poisson's Equation

Poisson's equation describes an important relationship between the charge density and the electric field in semiconductor materials. The electric field is responsible for moving and separating the electron-hole pairs created through photon absorption in the space charge region (also called the depletion region); thus, this equation is essential for understanding the distribution of electrostatic potential in solar cells (A Numerical Simulation and Modelling of Poisson Equation for Solar Cells). The link for the charge density (ρ) is grounded on the concentrations of electrons (n), holes (p), ionized acceptor atoms (N_A), and ionized donor atoms (N_D). It is assumed that N_D is constant in the n-region and negligible in the p-region, while N_A is constant in the p-region and zero in the n-region for the ideal diode derivation. This equation gives

the primary link from the charge density in the device to the electric field strength [48]. It can be expressed as :

$$\nabla^2 \psi = -\rho/\epsilon \quad (1.15)$$

where:

- $\rho = q(p - n + N_D - N_A)$ is the charge density
- ϵ is the permittivity of the semiconductor
- p and n are the hole and electron densities, respectively
- N_D, N_A are the ionized donor and acceptor densities, respectively

1.5.6.2 Continuity equations

Electron and hole transport within the semiconductor is described by the continuity equations. They take into consideration the diffusion, drift, recombination, and creation of carriers. [48].

Electron Continuity Equation

$$\frac{\partial n}{\partial t} = \frac{1}{q} \nabla \cdot J_n + G_n - R_n \quad (1.16)$$

Hole Continuity Equation

$$\frac{\partial p}{\partial t} = -\frac{1}{q} \nabla \cdot J_p + G_p - R_p \quad (1.17)$$

where:

1. G_n and G_p are the generation rates of electrons and holes (due to light absorption)
2. R_n and R_p are the recombination rates of electrons and holes
3. J_n and J_p are the electron and hole current densities

Under steady-state conditions ($\frac{\partial p}{\partial t} = 0$), the net charge carrier change remains constant.

1.5.6.3 Current Density Equations (Drift-Diffusion Equations)

Born of the basic continuity equations, these are the transport equations which form the basis of modelling the flow of current in a solar cell in terms of the two main methods by which carriers are driven in motion: diffusion driven by concentration differences, and drift driven by electric fields.

1.5.6.3.1 Drift current in the solar cell

Drift transport specifically refers to charge carriers in a semiconductor substrate moving due to an electric field (electrons move opposite to the electric field, and holes move with the electric field). The drift of the charge carriers results in a drift current. In order to consider the processes in which charge carriers are lost from the semiconductor material in a PV cell, it is important to recognize drift transport, which is explained through the drift equation. The one-dimensional drift equation is written as

$$J_x = q(n\mu_n + p\mu_p)E_x \quad (1.18)$$

where E_x is the electric field applied in the x-direction, q is the electron charge, n and p are the electron and hole concentrations, and μ_n and μ_p are the electron and hole mobilities. The electric field and the semiconductor's conductivity, which are influenced by the carrier concentrations and mobilities, are proportional to the drift current density, as demonstrated by this equation. The relationship between the electric field, carrier concentrations, and material parameters in determining the drift current is illustrated by equation [1.18](#).

Diffusion is the process by which charge carriers move randomly from a higher concentration to a lower concentration area. This will occur if the semiconductor has a non-uniform concentration gradient of carriers. Understanding how charge carriers move throughout the solar cell is an important factor, and this movement can be represented mathematically by the diffusion equation. The one-dimensional diffusion equations for electrons (n) and holes (p) are given by equations [1.19](#) and [1.20](#), respectively.

$$J_n = qD_n\left(\frac{dn}{dx}\right) \quad (1.19)$$

and

$$J_p = -qD_p\left(\frac{dp}{dx}\right) \quad (1.20)$$

where J_n and J_p are the diffusion current densities, q is the electron charge, D_n and D_p are the diffusion coefficients for electrons and holes, and $\left(\frac{dn}{dx}\right)$ and $\left(\frac{dp}{dx}\right)$ are

the electron and hole concentration gradients. According to these formulas, the diffusion current density is proportional to both the concentration gradient and the diffusion coefficient. The direction of the concentration gradient is opposed to the direction of the carrier motion, as shown by the hole diffusion equation's negative sign. Diffusion balances out carrier concentrations over time, which affects device performance. Therefore, as shown below, equations [1.21](#) and [1.22](#) represent the total current density inside the solar cell, which is the sum of the drift current and diffusion current [\[3\]](#).

$$J_n = qn\mu_n Ex + qDn\left(\frac{dn}{dx}\right) \quad (1.21)$$

$$J_p = qp\mu_p Ex - qDp\left(\frac{dp}{dx}\right) \quad (1.22)$$

1.6 Organic Solar Cells: A New Frontier

Organic solar cells (OSCs) have captured considerable academic interest in the last 20 years due to material availability, low cost of fabrication and processing, and sustainable ecological benefits. The ability for organic photovoltaics to be the solution to the world's energy needs will depend on how well we can optimize the resulting output performance of the device. Over the last 10 years, a lot of research has been conducted to understand the basic principles of organic solar cell technology and to understand OSCs' performance under different conditions. As a complete theoretical model of the operation of organic solar cells does not yet exist, research in organic photovoltaics would greatly benefit from numerical simulation studies and device model settings utilizing computational techniques.

1.6.1 The evolution of organic solar cells

The photovoltaic effect in organic materials was first observed in the 1950s, but early devices were not particularly efficient, with efficiencies of only $10^{-5}\%$ because of limited charge transport and substantial recombination losses [49]. In 1977, Tang made a big step forward by making the first bilayer organic photovoltaic cell, which had two separate organic semiconductor materials: a donor and an acceptor [50]. This device used copper phthalocyanine (CuPc) and a perylene tetracarboxylic derivative, and it was able to convert around 1% of the radiation it received. In the middle of the 20th century, scientists began to look into the photoconductive properties of polymers and organic materials. This was when the idea of using organic materials to convert solar energy started. In the 1970s, polymers were suggested as an active layer in prototype solar cells. However, these prototypes only achieved efficiencies of less than 1%, which showed that it was hard for excitons to break apart and for charges to go through organic materials.

The most important change was the introduction of the bulk heterojunction (BHJ) architecture in the middle of the 1990s. This arrangement made it easier for excitons to break apart since the donor and acceptor were mixed instead of stacked [51]. In 2001, Shaheen et al. made a significant breakthrough in BHJ photovoltaic devices when they combined the conjugated polymer poly(3-hexylthiophene) (P3HT) with [6,6]-phenyl- C_{61} -butyric acid methyl ester (PCBM) to get power conversion efficiencies (PCEs) of 2.5% [52]. This design is still the most popular choice today because it shows better separation and collection efficiency. Researchers have investigated the polymer ratio P3HT:PCBM a lot since it became the model system. They did this to

learn more about energy levels and morphology and improve performance [53] .

By the end of the decade, PCEs had risen to 5% thanks to better ways of making and processing donor materials, better control over the orientation of donor and acceptor materials, and the use of low band gap polymers to get even more out of the solar spectrum [54]. The most important improvement was the addition of non-fullerene acceptors (NFAs) like ITIC and Y6 [55]. NFAs have adjustable energy levels, absorb more light, and are more stable than fullerene derivatives. This means that the organic solar cell (OSC) quickly improved its power conversion efficiency (PCE). In 2020, Y6-based devices could reach over 18% PCE in the lab [56], showing that OSCs had made a platform transition and were now on par with some inorganic thin-film technologies. The tandem organic solar cells employ several absorbers with complementary band gaps, engineered to surpass the Shockley–Queisser limit for single-junction cells [57]. Research includes all-polymer solar cells, ternary blends, and hybrid organic-inorganic materials. New devices are also being built employing encapsulation and roll-to-roll production processes to meet standards for commercial usage. These devices are being made with the purpose of being stable and scalable for commercial manufacture.

1.6.2 Advantages of organic solar cells

Organic solar cells (OSCs) have several significant benefits that put them in a great position when compared to conventional silicon solar cells. Key benefits include:

- **Lightweight and Flexible:** OSCs are much lighter and more flexible than silicon solar cells, making them a perfect fit for applications where weight and

flexibility are important, such as building-integrated photovoltaics (BIPV) and wearable technologies.

- **Semi-Transparent:** OSCs can be manipulated to be semi-transparent, allowing them to be used in windows and other transparent surfaces, which cannot be done with conventional solar cells.
- **Environmentally Friendly:** Organic solar cells are made with organic materials and are generally more environmentally friendly and less toxic than the synthetic inorganic materials used in conventional solar technologies.
- **Thin and Material-Saving:** OSCs are about 1000 times thinner than their silicon-related solar cells, which provide material savings and cheaper production costs.
- **Short Energy Payback Time:** Organic solar cells will have shorter energy payback times than silicon solar cells due to lower energy to produce, making organic solar cells able to produce more energy than energy consumed during manufacturing time.
- **Simply Integrate and Versatile:** OSCs can simply integrate and be versatile, existing on their customized structure and flexibility, making them practical in various applications, from bendable panels to solar windows.
- **Lower Manufacturing Cost:** The cost of manufacturing organic solar cells is cheaper than standard, with the roll-to-roll printing process maximizing the efficiency in manufacturing and lowering manufacturing costs overall.

1.6.3 Organic semiconductor materials

Organic semiconductors are derived from π -bonded organic molecules or polymers, composed mainly of hydrogen and carbon, with heteroatoms such as sulfur or nitrogen sometimes added. In comparison to inorganic semiconductors, organic semiconductors are typically either highly amorphous thin films or molecular crystals that behave like electrical insulators until charge carriers are introduced (e.g., by photoexcitation, electrical injection, or doping).

Organic semiconductors consist of conjugated molecules or polymers, where carbon atoms use sp^2 hybridized orbitals. In conjugated systems, each carbon atom forms three σ -bonds with its sp^2 hybrid orbitals. Two of these σ -bonds are formed with hydrogen or side-group atoms, while one involves another carbon atom. The remaining p -orbital is unhybridized and lies in a plane perpendicular to the molecular plane, establishing π -bonds with neighboring π -orbitals. These π -bonds form a delocalized π electron system, providing charge-carrier movement and developing a conduction and valence band similar to those in inorganic semiconductors [58]. Typically, the band gap is in the range of 1.5 eV to 3.5 eV, enabling absorption or emission of visible light. Charge transport may occur through either hopping or delocalized mechanisms, depending on the crystallinity and purity of the material. Their light weight and mechanical flexibility also make them ideal for wearable and flexible electronics [59].

Organic semiconductors differ from inorganic ones mainly in their reliance on localized electronic states and a hopping conduction mechanism, where charge carriers move through localized molecular orbitals assisted by thermal activation. The mobility of holes and electrons in organic materials ranges from 10^{-6} to $1 \text{ cm}^2/\text{V} \cdot \text{s}$, whereas in

inorganic semiconductors like silicon, electron mobility can reach about $1500 \text{ cm}^2/\text{V} \cdot \text{s}$ [60]. The use of photovoltaic polymers is rapidly increasing, primarily because they can be fabricated by methods such as spray coating, spin coating, and inkjet printing [61].

1.7 Diverse structural designs of organic solar cells

Organic solar cells (OSCs) offer a promising alternative to inorganic photovoltaics due to their lightweight nature, mechanical flexibility, and low-cost fabrication. Multilayer OSCs consist of several functional layers that play essential roles in charge generation, transport, and collection. The device structure strongly influences both the efficiency and long-term stability of OSCs. The following sections outline the key device configurations commonly used in multilayer organic solar cells:

1.7.1 Single-Layer (Schottky) Structure

The basic single layer structure of an organic solar cell (OSC) consists of a single layer of organic semiconductor placed between two different electrodes, which are typically defined as a front anode, which has a high work function, and a back cathode, which has a low work function. The organic layer absorbs the photons, forming excitons (bound pairs of electrons and holes), which then diffuse to the interface of the electrodes, where charge separation occurs, driven by the built-in electric field. The efficiency suffers both from large recombination losses and a limited diffusion length of the exciton of around 10-20 nanometers [50]. The structure of a single-layer organic solar cell is shown in Fig. 1.18.

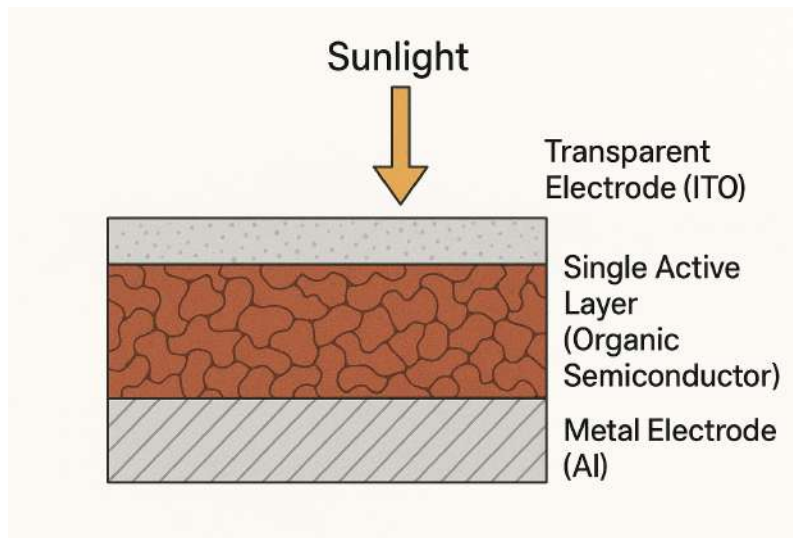


Figure 1.18: Single-layer OSC

1.7.2 Bilayer (Planar Heterojunction, PHJ) Structure

The PHJ structure was developed to maximize exciton dissociation, consisting of separate donor (D) and acceptor (A) layers, where excitons produced in the donor layer will diffuse towards the D/A interface to dissociate into free charge carriers due to the energy difference of the two materials. The bi-layer structure has superior charge separation than single-layer devices but will have an exciton diffusion length on the order of 10–20 nm, thus limiting the layer thickness and the amount of light absorption [62]. Fig 1.19 shows the structure of a bilayer organic solar cell structure

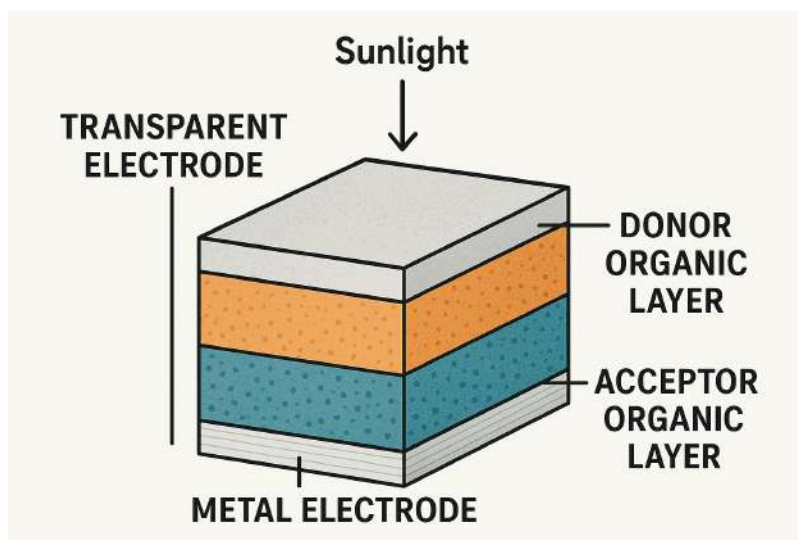


Figure 1.19: Bilayer OSC

1.7.3 Tandem (Multi-Junction) Structure

Tandem organic solar cells (OSCs) are composed of two or more subcells with complementary absorption spectra to optimize for light harvesting efficiency while also maximizing the open-circuit voltage. Each sub-cell can absorb a different portion of the solar spectrum to further contribute to the total photocurrent and voltage. The enhanced PCE of over 17% is attributed to the broadened spectral absorption and the reduction of thermalization losses. This technology is limited by two main factors: a complex fabrication process and the requirement for compatible interlayers. [63].

Fig 1.20 shows a tandem or multijunction organic solar cell.

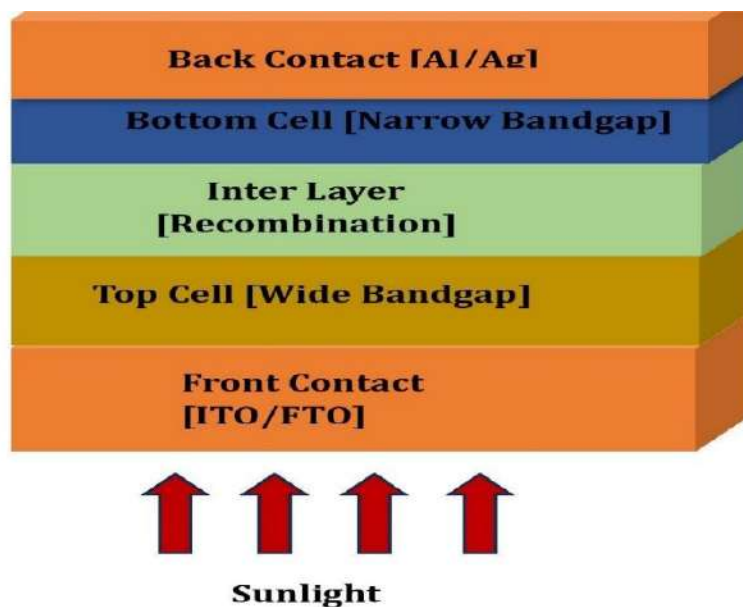


Figure 1.20: Tandem [Multijunction] OSC

1.7.4 Inverted Structure

Inverted organic solar cells (OSCs) change the normal layer stacking to improve stability by enabling air-stable electrodes. Unlike normal OSCs, electrons are collected at the cathode/electron transport layer (ETL), and holes travel through the hole transport layer (HTL) to the metal anode. Increased stability is obtained by avoiding reactive low-work-function metals, such as calcium (Ca) and aluminum (Al) [64]. Fig 1.21 illustrates an inverted solar cell architecture.

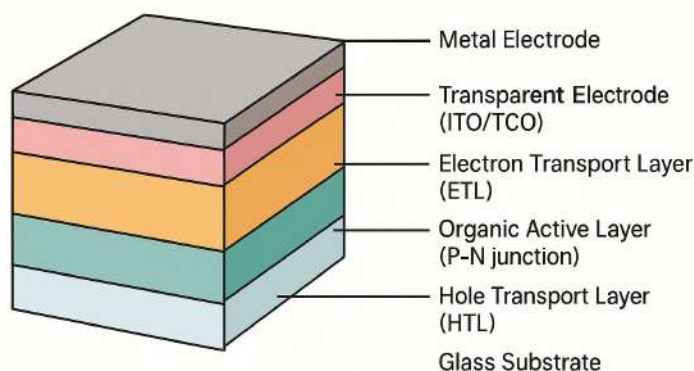


Figure 1.21: Inverted OSC Structure

1.7.5 Bulk Heterojunction (BHJ) Structure

The bulk heterojunction (BHJ) structure reduces the limitations of exciton diffusion by combining donor and acceptor materials into a single active layer and forming a nanoscale interpenetrating structure. When an exciton is formed in the composite blend, it can diffuse through the blend and get to a donor/acceptor interface within its diffusion length to facilitate charge separation. High power conversion efficiencies (PCEs over 10%) can be achieved based on the increased interface area. However, to control morphology, one must optimize phase separation for ideal charge transport [51]. The structure of the bulk heterojunction organic solar cell is shown in Fig. 1.22

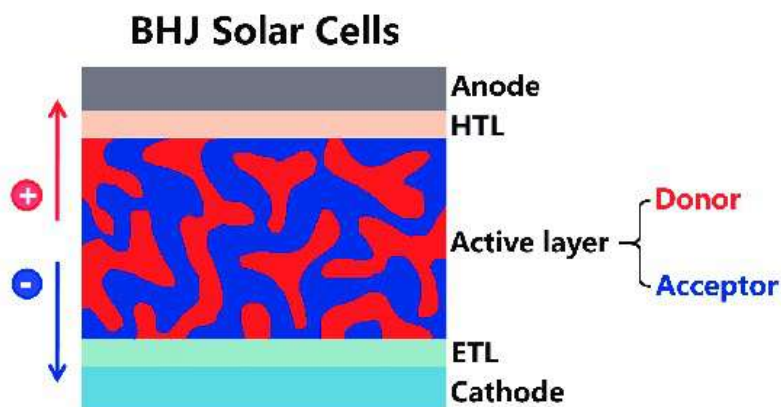


Figure 1.22: Bulk heterojunction OSC

1.7.5.1 Various layers within a multilayer BHJ OSC

Organic solar cells (OSCs) created with a new bulk heterojunction (BHJ) structure have gained enormous interest due to their low cost and potentially flexible or lightweight solar energy conversion. A multilayer BHJ OSC has multiple functional layers so that each layer is involved in the functional process of light absorption, charge generation, transport, or collection. A thorough description of each of these layers is given below:

1.7.5.1.1 Substrate Layer

The substrate provides the essential mechanical foundation for the solar cell, acting as the platform for the deposition of subsequent layers. Substrates may be rigid (e.g., glass) or flexible. For bottom-illuminated organic solar cells (OSCs), transparency is a mandatory characteristic. Glass is a widely used rigid substrate, while polymers such as polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) are

common choices for flexible applications [65, 66].

1.7.5.1.2 Transparent Conductive Electrode (TCE)

As previously mentioned, this superior layer allows light to be absorbed with care while collecting charges, whether used as the anode in conventional configurations or the cathode in the reverse configuration, and while doing so, allows the holes to move seamlessly through the material. The transparent conductive electrode must have excellent transmittance (85%) and low ($20\Omega/sq$) sheet resistance. The transparent conductive electrode must be Indium Tin Oxide (ITO), Fluorine-doped Tin Oxide (FTO), Conductive Polymers (PEDOT:PSS), or Metal Grids/Ag Nanowires [67, 57, 68].

1.7.5.1.3 Hole Transport Layer (HTL)

The HTL enhances hole transport to the anode while blocking electron transport, thereby enhancing charge selectivity and reducing recombination at the electrode interface. It optimizes the alignment of energy levels, while its modification produces vertical phase separation in the active layer, increasing device efficiency. The materials used in HTL typically are poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT: PSS), which is the most well-established. Possible inorganic materials that can be considered as HTLs are MoO_3 , V_2O_5 , and NiO [57].

1.7.5.1.4 Active Layer (Bulk Heterojunction, BHJ)

The active layer, or absorber, is the photoactive part of the device where photons are absorbed to create excitons, which then dissociate into free charge carriers that can produce a photocurrent. To achieve optimal performance, this active layer should have a low band gap, allowing it to absorb a wide spectrum of light, and a thickness optimised to that material's exciton diffusion length. Paired with other layers to allow charge extraction at the anode and cathode, the layer can be of three types: a single layer of one material, a bilayer heterojunction where there is a distinct donor and acceptor layer, or a so-called bulk heterojunction (BHJ) where the donor and acceptor materials are mixed into a composite film. The following sections discuss the donor and fullerene acceptor materials used in a bulk heterojunction organic solar cell.

1.7.5.1.4.1 Donor materials

The donor materials in organic BHJs are predominantly conjugated polymers. The most commonly used donor materials include P3HT (poly(3-hexylthiophene)), MDMO-PPV: Poly[2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylenevinylene], and PTB7 Poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]].

The acceptor materials are no less varied. There are both fullerene and non-fullerene acceptors. PC71BM and PC61BM are the two most common fullerene acceptors. Non-fullerene materials such as ITIC, PDI, IDTBR, IEIC, and others are widely used and account for a substantial share of the acceptor materials landscape.

Fig 1.23, Fig. 1.24 and Fig 1.25 represents the structure of donor materials.

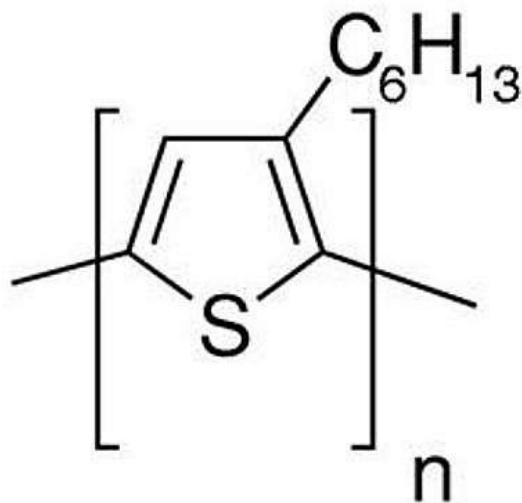


Figure 1.23: P3HT

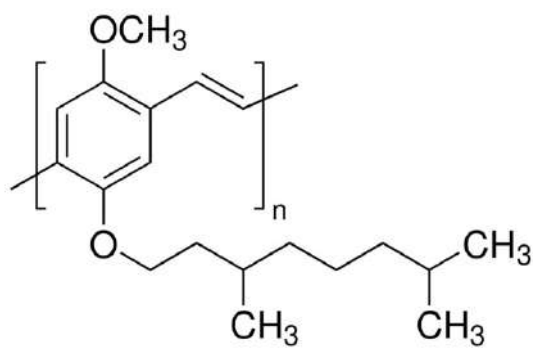


Figure 1.24: MDMO-PPV

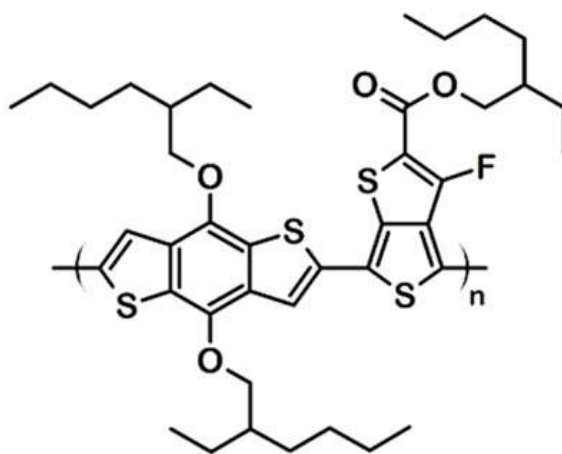


Figure 1.25: PTB7

1.7.5.1.4.2 Fullerene-Based Acceptors

For the past 20 years, the main area of research in the field of organic photovoltaics have been polymeric donors made with fullerene-based acceptors, because bulk heterojunction organic solar cells have been shown to achieve higher processability and stability in spite of the other donor-acceptor systems. The relatively enhanced device performance of fullerene-containing active materials in organic solar cells is associated with favorable interfaces created between donor and acceptor materials and tunable orbital energy levels of fullerene derivatives. Fullerenes have always been the most common type of acceptor materials of conventional organic solar cells due to their distinctive 3D nature, high electron mobilities, and tunable electron affinities [29, 30, 69].

The discovery of buckminsterfullerene, or C₆₀, in 1985 was one of the early devel-

opments of organic photovoltaics [70]. Fullerenes have a spherical design comprised of 30 conjugated double and single bonds with 12 pentagons and 20 hexagons [71]. The first bulk heterojunction organic solar cell had PC61BM as the acceptor and established fullerenes as the most prominent component of organic solar cells [51]. Fullerenes can also form advantageous interpenetrating networks with many donor materials, allowing organic solar cells with various donor polymers.

The efficiencies in OPV devices have been greatly enhanced through rigorous research and optimization of these materials. The electron acceptor is the most important component of the organic bulk heterojunction (BHJ) structure, with the electron donor (most often a conjugated polymer or small molecule) facilitating charge separation and transport. Fullerene, or in this case, fullerenes reached their zenith of consumption as electron acceptor materials around the mid-1990's, when C_{60} and C_{70} were first introduced as well as their soluble derivatives $PC_{60}BM$ (phenyl- C_{61} -butyric acid methyl ester) and $PC_{70}BM$ (phenyl- C_{71} -butyric acid methyl ester).

Because of their relatively high electron affinities (between 3.7 and 4.1 eV) and ability to extract electrons from numerous donor options quickly, fullerenes became very popular as electron acceptors. Additionally, fullerenes also exhibit nearly isotropic charge transportation characteristics, allowing the unequal charging of multiple layers of devices at once (better device performance). When fullerene is coupled with conjugated polymers, a desired nanoscale degree of phase separation in BHJ blends are developed, which is vital in converting light into electrical energy through the formation and transport of excitons and charge carriers in organic solar cells.

Fullerenes originated, in fact, from all-purpose testing in organic electronics devices.

In short, fullerenes were ultimately a harsh yet available resource (had capacity (high selective index of utility)), where $PC_{60}BM$ held the variety of prosthetic fullerene acceptors because solubility in organic polymer solvents was compatible with the donor through the use of P3HT (poly(3-hexylthiophene)). The molecular structures of $PC_{60}BM$ is given in Fig 1.26.

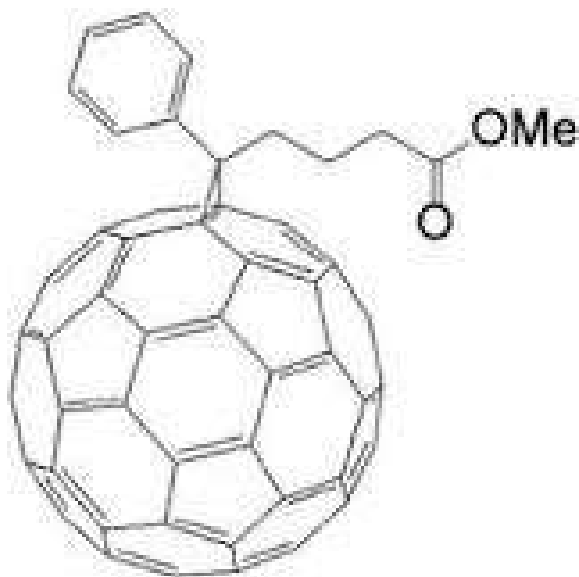


Figure 1.26: $PC_{60}BM$

Another operational performance advantage of $PC_{60}BM$ is in terms of a maximum operational performance average of 3–5% efficiencies. In terms of extended π -conjugation of C_{70} , derivative $PC_{70}BM$ (phenyl- C_{71} -butyric acid methyl ester), in fact, was recognized as having very closely the same absorption range allowable for the visible light region, and no incredible increases in J_{sc} were established for the operational achievement to extract any fresh incremental improvements for photovoltaic devices. The molecular structures of $PC_{70}BM$ is given in Fig 1.27.

Figure 1.27: PC₇₀BM

The operation had followed, mostly, a similar story; ICBA (Indene- C_{60} bisadduct) with operational advantages had a higher V_{oc} in comparison to PCBM due to the position relative to the lowest unoccupied molecular (LUMO) potential energy orbital [72, 73, 74]. The molecular structures of IC₆₀BA is given in Fig 1.28.

Figure 1.28: IC₆₀BA

A landmark in this area is that devices based on P3HT reached a PCE of nearly 6%. Organic photovoltaics based on fullerenes rely on certain photo-physical processes. Upon illumination, the donor polymer molecule absorbs photons and excites excitons, or bound electron-hole pairs, which diffuse towards the donor-acceptor interface. At this interface, dissociation occurs because fullerene molecules have a greater electron

affinity than the polymer donor, allowing the polymer to give up its excited electron to the fullerene acceptor, leaving a hole in it. The electron moves through the fullerene-rich domains and the holes through the polymer donor phase towards their respective electrodes. Fullerene acceptors need to have strong optical absorption, be miscible with a wide variety of donor polymers, and have tunable molecular orbital energy levels that correspond well with the donor materials.

Recent investigations have focused on ultra-low-electron-affinity fullerene derivatives, which are anticipated to exhibit novel optoelectronic properties, improved compatibility with donor materials, and potentially enhanced stability and performance in organic photovoltaic (OPV) devices. In addition, fullerene-based electron acceptors are entirely stable and photostable compounds and thus continue to be a large driver of future research, especially with multi-component and hybrid solar cells [75]. PCBM ([6] [6]-phenyl- C_{61} -butyric acid methyl ester) is the more popular fullerene derivative because of the following impactful factors: very high solubility, tunable energy levels, and high charge transfer efficiency [73]. Numerous structural derivatives of PCBM, like PC71BM (which is optically derived from C_{70}) and ICBA (indene- C_{60} bisadduct), may offer greater light harvesting and perhaps higher open-circuit voltage [53].

Indene- C_{60} bisadduct ($IC_{60}BA$) is a fullerene derivative widely used in high-performance organic solar cells (OSCs) due to its advantageous electronic and structural properties, including high electron mobility and optimal energy level alignment with common donor materials, thus ultimately simplifying $IC_{60}BA$ as an applicable electron acceptor material. $IC_{60}BA$ has a C_{60} fullerene core and 2 indene groups

attached at a bisadduct.

There are four possible regioisomers for $IC_{60}BA$ where indenenes can occupy different positions and polarities that can affect the photovoltaic performance of organic solar cells. Through single crystal x-ray diffraction (XRD) characterization, we can characterize the structural configuration of $IC_{60}BA$, which is important to understand because the location of the indene groups all relies on the arrangement of indene fully, which depends on their structural configuration above the C_{60} cage, which will dictate where the indene groups will lie.

$IC_{60}BA$ has a higher lowest unoccupied molecular orbital (LUMO) energy level than the popular fullerene derivative PCBM. Higher LUMO levels provide higher open-circuit voltage (V_{oc}) in organic solar cells (OSCs), ultimately leading to higher power conversion efficiency (PCE). The energy-level alignment of $IC_{60}BA$ with polymer donors is also critical for charge separation and for improving the cell's efficiency [76, 77, 78].

$IC_{60}BA$ is highly soluble in organic solvents (e.g., chlorobenzene, o-dichlorobenzene) and can be processed from solution [76]. Compared to PCBM, $IC_{60}BA$ is also much more amorphous and more miscible with polymeric donors on average, which limits the extent of large-scale phase separation. $IC_{60}BA$ was developed to strategically improve upon many of the deficiencies of fullerene acceptors, such as the very low open-circuit voltage (V_{oc}) and poor tunability. As a new family of $IC_{60}BA$ developments, it represented an important milestone to improve V_{oc} of fullerene-based acceptors with significant increases in optoelectronic efficiency in organic solar cells (OSCs) (maximum efficiencies on the order of 6.5%) [71]. $IC_{60}BA$ has also been used

as a performance additive in binary, ternary, and multi-component blends to control morphology in the photoactive layer of devices.

The photovoltaic performance of OSCs may also be improved with $IC_{60}BA$, as it likely has more symmetric charge transport properties. In comparison with non-fullerene binary OSCs, another important advantage of $IC_{60}BA$ is that it can enhance active layer morphology (as a third component). Organic solar cells (OSCs) rely on small, mostly pure domains to achieve efficient charge separation and transport. In summary, $IC_{60}BA$ represents an essential milestone in the development of fullerene-based acceptors for high-efficiency organic solar cells [79].

1.8 Thesis Outline

The primary aim of this thesis is to develop highly efficient multilayer bulk heterojunction organic solar cells utilizing fullerene acceptor materials in conjunction with the most compatible transport layers. The device architectures of fullerene-based organic solar cells are modeled using simulation tools, which have been calibrated by comparing their outputs with experimental data from existing literature. The subsequent chapters provide an in-depth analysis of the simulation studies conducted on multilayer fullerene organic solar cells.

Chapter 1 presents an introduction to solar cells and a summary of the history and operation of organic solar cells, with a specific focus on fullerene-based organic solar cells, including $IC_{60}BA$. In Chapter 2, the SCAPS 1D simulation tool was used to assess this study, and the fundamental principles of device modelling were discussed. In Chapter 3, the role of CuI as the hole-transport layer was evaluated in

fullerene-based organic solar cells with the P3HT:IC₆₀BA active layer. Chapter 4 used simulation results of fullerene organic solar cells and their optimization without an electron transport layer. Chapter 5 assessed how the P3HT additional layer affected the performance of fullerene-based organic solar cells. Chapter 6 followed the analysis and simulation of multilayer fullerene organic solar cells with diverse electron and hole transport layers. In Chapter 7, a summary of the study is done and potential next steps are proposed. Finally, chapter 8 provides recommendations for future study.

Chapter 2

Simulation tool and Device modelling

The development of organic solar cell (OSC) technology, particularly based on promising materials like $IC_{60}BA$, relies on our ability to understand and predict device performance. While experimental production and characterisation remain critical, they can be expensive and time-consuming, particularly when optimising across a broad parameter space. Computational simulation offers a viable and complementary pathway, allowing for rapid prototyping, extensive examination of internal device physics, and systematic assessment of how various material and structural parameters contribute to overall efficiency. The fascinating topics of simulation tools and device modelling are the focus of this chapter in the context of $IC_{60}BA$ -based organic solar cells.

It aims to give a broad overview of the theoretical background and practical applications of computational techniques for modelling and examining the behaviour of photovoltaic devices. The main emphasis of this chapter will be the Solar Cell Capacitance Simulator in one dimension (SCAPS-1D). This popular software package, including OSCs, is a highly valuable research resource for the thin-film solar cell com-

munity. Its value is partly in its strong numerical solution of the basic semiconductor equations (namely, Poisson’s equation and the continuity equations for electrons and holes) with drift-diffusion transport and the associated generation-recombination models. This chapter will outline the rationale for selecting SCAPS-1D as a research tool, describe its structure, detail the relevant physical models it employs for OSCs, and specify the input parameters required to accurately simulate IC₆₀BA-based device structures. The remaining sections will assist in establishing the context regarding the simulation results provided later in this thesis. These sections will first explore the general principles of device modelling as applied to solar cells, then demonstrate how to use the SCAPS-1D environment and describe the configuration and validation process undertaken for the specific complexities and features of the organic solar cells being studied, before interpreting the optimisation studies and design rules derived from simulation work.

2.1 Overview of the Simulation Methodology

This section explains the use of a computational simulation method in this thesis as part of the optimization studies of IC₆₀BA-based organic solar cell devices. The advantage of numerical simulation (in terms of cost and speed) over experimental methods (which can be difficult to do, or take longer) for studying device physics, optimizing performance parameters and evaluating the effects of different materials or structural characteristics are very significant. This thesis utilized the 1-D version of the program SCAPS. The following two subsections detail the rationale for selecting SCAPS-1D as the simulation tool and outline the systematic methodology used for

the simulations [80].

2.1.1 Reasons for using SCAPS-1D

The choice of SCAPS-1D (Solar Cell Capacitance Simulator – 1 Dimension) for this investigation is based on a number of very particular advantages of the software package for the optimization studies of organic solar cells based on IC₆₀BA. SCAPS-1D is a mature and widely accepted numerical simulation program for thin film solar cell devices and possesses the following advantages:-

1.In-depth Physical Modelling: SCAPS-1D can numerically solve the governing equations of the semiconductors (Poisson’s Equation and continuity equations for holes and electrons) to allow in-depth modelling on important phenomena such as charge transport, recombination and generation for solar cell devices [81].

2.Flexibility: SCAPS-1D can simulate many device types (thin film, crystalline, heterojunction) and therefore may be used for many different research applications [82].

3.Ease of Use: SCAPS-1D programs are very easy to use with both a graphical interface and script language, and allow for simple and advanced simulations.

4.Parameter Flexibility: The program allows the user to input material parameters (thickness, band gap, defects, doping), which allows an enormous variety and detail, allowing researchers to simulate the results of optimisation studies and sensitivity analysis [82].

5.Widespread Use and Community Acceptance: SCAPS-1D is widely used and accepted by the PV research community to the extent that many simulation results

show similar trends and values to those seen experimentally, thus giving researchers the confidence they need that the results of their simulations and predictive modelling techniques are accurate [83].

2.2 Simulation study workflow

The simulation study, using SCAPS-1D, has a pre-established workflow, which can be described as follows to complete a thorough and reproducible study:

a) Define the Problem: Define the device structure in terms of layers (how many layers are present in the device) and identify the material property of each of the layers (thickness, band gap, doping concentration, and defect densities) [80].

b) Define the Simulation Conditions: Define what conditions would apply to the simulation in terms of temperature and illumination spectra (AM 1.5G), including operating voltage/frequency ranges, to replicate realistic device operating conditions [80].

c) Define Measurable Parameters: The specific measurements selected to be simulated will differ in every research study, depending on the research objectives, for example, I-V (current-voltage), C-V (capacitance-voltage), C-f (capacitance-frequency), and Quantum Efficiency (QE) experiments [80].

d) Define Variations and Optimization: The user can incrementally vary and change the key device parameters and material property values, or utilise the batch calculation capability of SCAPS-1D, which also supports studies related to performance metrics such as Power Conversion Efficiency (PCE), open-circuit voltage (Voc), short-circuit current density (Jsc), and fill factor (FF) [82].

e) Running Simulations: Simulations can then be run for each set of defined parameters. SCAPS-1D allows for numerical computing-based analysis to examine energy band diagrams, carrier profiles, and I-V curves [81].

f) Analysis and Visualization: The simulation outputs are analyzed to identify performance trends, optimize device characteristics, and deepen the understanding of the underlying physics. SCAPS-1D facilitates this analysis by enabling users to visualize band diagrams and carrier densities. These visualizations allow for clear interpretation of the results, which is essential for reporting to funding agencies and for inclusion in scientific journals [83].

g) Final Documenting of Results: Data and graph results can be saved for processing and reports for publication. This process allows simulation studies to be thorough and reproducible, providing a solid foundation of reliable data to enable the design and optimisation of next-generation solar cell devices.

2.3 SCAPS-1D Software: Theoretical Structure

SCAPS-1D (Solar Cell Capacitance Simulator in 1D) is a numerical simulation tool developed for the study of photovoltaic systems. This section of the paper provides its theoretical foundation. Regardless of the circumstances, SCAPS-1D solves the basic semiconductor equations underpinning the operation of solar cells [80].

2.3.1 SCAPS 1D Software Background (version 3.3.08, Capabilities)

Developed at the University of Gent, Belgium, SCAPS-1D version 3.3.08 is a one-dimensional simulation tool [121][84]. The program was meant to help model thin

film solar cells including perovskite, CIGS, and CdTe based. SCAPS-1D offers the following features:

- Simulation of quantum efficiency (QE), capacitance-voltage (C-V), and/or current-voltage (J-V)
- Study of recombination processes and defect states
- Performance depending on temperature
- Optical modelling of multi-layer structures

2.3.2 Solved Basic Semiconductor Equations

SCAPS-1D numerically solves the following two linked partial differential equations:

1. Poisson's equation

$$\nabla^2 \psi = -\rho/\epsilon \quad (2.1)$$

where: • $\rho = q(p - n + N_D - N_A)$ is the charge density,

- ϵ is the permittivity of the semiconductor,
- p and n are the hole and electron densities, respectively,
- N_D, N_A are the ionized donor and acceptor densities, respectively

2. Equations of Continuity

Electron Continuity Equation

$$\frac{\partial n}{\partial t} = \frac{1}{q} \nabla \cdot J_n + G_n - R_n \quad (2.2)$$

Hole Continuity Equation

$$\frac{\partial p}{\partial t} = -\frac{1}{q} \nabla \cdot J_p + G_p - R_p \quad (2.3)$$

where:

- G_n and G_p are the generation rates of electrons and holes (due to light absorption).
- R_n and R_p are the recombination rates,
- J_n and J_p are the electron and hole current densities. The continuity equations indicate the time evolution of the concentration of the electrons, n , and holes, p , as well as the rules for generation (G) and recombination (R) of the carriers.

3. Drift-diffusion transport model

$$J_n = qDn\left(\frac{dn}{dx}\right) \quad (2.4)$$

and

$$J_p = qDp\left(\frac{dp}{dx}\right) \quad (2.5)$$

The drift-diffusion transport model includes the electric field E driving carrier drift, as well as carrier diffusion driven by the concentration gradient.

2.3.3 Boundary Conditions

The SCAPS-1D simulation environment allows for the implementation of several types of boundary conditions, which are essential for solving the governing semiconductor

equations [84]

Ohmic contacts are primarily defined by the high carrier concentrations achieved through donor or acceptor doping at the interface. In contrast, Schottky contacts are governed by properties like the metal work function and surface recombination velocity, which create a rectifying energy barrier. Boundary conditions are governed at the interfaces of the materials, including defect states and recombination bands.

Solving the above equations under specific boundary conditions helps determine the performance characteristics of solar cells—including efficiencies, fill factors, and recombination losses.

2.4 Material Characteristics: Parameters Required for SCAPS-1D Analysis

Numerical modeling via the SCAPS-1D (Solar Cell Capacitance Simulator in one Dimension) is fundamentally reliant on the appropriateness and accuracy of the input material parameters for their fidelity and prediction capability.[80]. SCAPS-1D numerically solves the basic equations of semiconductors (Poisson’s equation, continuity equations for electrons and holes and the drift-diffusion transport equations) in a 1D device structure. This section presents the material parameters used in the simulation of a bulk heterojunction (BHJ) solar cell, including the donor , acceptor, the hole transport layer (HTL), electron transport layer (ETL), and electrode contacts. The parameters are set out in sections detailing the electrical, optical, and defect properties, with the relevant experimental and theoretical work presented for validation.

2.4.1 General Parameters

These parameters establish the operational conditions for the simulated solar cell and are universally applicable to the device architecture.

2.4.1.1 Temperature

The operating temperature greatly influences the performance of a solar cell. SCAPS-1D can specify the device temperature in Kelvin (K). Many models consider a standard operating temperature of 300 K to analyze; however, SCAPS-1D can allow for modeling of the device, and a temperature-dependent analysis can be performed on the device to see the performance of the device in different temperature modes and where performance deviation of the device parameters will occur (Voc, Jsc, FF, etc.). Carrier mobility is affected by temperature (phonon scattering) and band-gap narrowing.

2.4.1.2 Illumination Spectrum (AM1.5G)

The Air Mass 1.5 Global (AM1.5G) spectrum is a standard solar irradiance spectrum used for terrestrial solar cell simulations. It is the industry/academic community standard for reporting solar cell efficiencies, and it helps provide comparability between simulated results from SCAPS-1D and experimental results. It can be thought of as sunlight after it has passed through Earth's atmosphere at an angle large enough to have 1-sun intensity. It is a key characteristic for simulating realistic operating conditions.

SCAPS-1D has this spectrum built in, which defines the incident light power and

wavelength distribution that SCAPS-1D uses to calculate photocurrent generation in the solar cell. The spectral range from 300 to 1200 nm is most relevant for organic semiconductors because below this range, photon energies are too high, leading to minimal absorption or generation. Above this range, photon energies are too low to generate electron-hole pairs. The light intensity is typically set to 1 sun (or 100 Mw/cm^2). The incident spectrum's intensity and the absorption coefficient of the active layers determine the rate of electron-hole pair generation; this rate is constant at every position and wavelength along the light path in the device. This feature is a principal driver for the photocurrent [85].

2.4.2 Layer-Specific Parameters

The parameters for each of the layers in the solar cell stack are independent. The composition of the active layer is a mix of P3HT and IC_{60}BA , and it is accepted that SCAPS-1D models the active layer as a single effective medium. However, other techniques such as co-simulations and multi-layers can be adopted if it is assumed that the parameters of the pure phases, and blend morphology, can be found. For each of the layers in a solar cell device SCAPS-1D requires a set of electrical, optical, and defect parameters. These parameters govern the behaviour of each of the materials in the device, and are needed for an effective simulation.

2.4.2.1 Electrical Parameters

- **Bandgap (E_g):**

The bandgap, E_g , is defined as the energy difference between the conduction

band minimum and the valence band maximum of the semiconducting material and fundamentally defines the wavelengths a material can absorb [86].

- **Electron affinity (χ):**

The electron affinity χ is the energy difference between the minimum of the conduction band(s) and that of the vacuum level. It is of interest because it serves to define electronic band arrangements that exist for any interfaces of the materials, and so the electron conduction [86].

- **Work function (ϕ)** The work function, ϕ , is the minimum energy required to take an electron out of a solid body (such as a metal) to a vacuum level just outside the solid surface. The work function is of particular interest in relation to metallic contacts, since this allows the height of Schottky barriers to be described [86].

- **Effective Density of States in Conduction/Valence Band (N_C, N_V) :**

The effective density of states is the number of available states at the top of the conduction band and at the bottom of the valence band. It is a very important factor in determining the carrier concentrations, and hence the conductivity, of any material.

- **Electron and hole mobility (μ_n, μ_p) :** The terms electron mobility and hole mobility denote the case of transporting electrons in the case of n-doped silicon under an electric field and holes in the case of p-doped silicon under an electric field. The improvement of charge carrier mobility should generally mean

increased charge carrier transportation and thus improvement of the device performance.

- **Dielectric Permittivity (ϵ_r):** It is a measure of a substance's ability to store electrical energy in an electric field. The dielectric permittivity enables one to determine the absolute capacity and thus the capacitance of the solar cell.
- **Doping Concentration (N_A, N_D):** The doping density indicates the number of impurities introduced into the semiconductor in order that the latter is able to show certain conductivity. N_A is the acceptor density (p-doping) and N_D the donor density (n-doping). This information is required to fix the electrical indication of the doped layers [86].

2.4.2.2 Optical Parameters

- ✓ **Absorption Coefficient of active layers :** The absorption coefficient indicates the amount of light that a substance absorbs of a certain wavelength, in relation to the incident light. This variable is an important parameter to take into account for all of the active layers, especially the perovskite layer, since the absorption coefficient basically says how much light generates electron-hole pairs.
- ✓ **Refractive index (n):** A refractive index indicates how light passes through a material, namely, the speed of light in a vacuum compared to that of light passing through some specific material.
- ✓ **Extinction coefficient (k):** The extinction coefficient indicates how much

light a material absorbs or scatters. The extinction coefficient is also defined in relation to the absorption coefficient. When performing detailed optical modelling, these parameters can be included in a simulation that more accurately examines how light propagates and is absorbed between the light source and the device.

2.4.2.3 Defect Parameters

2.4.2.3.1 Type:

- **Neutral:** These are traps that are electrically neutral when empty (not occupied by a charge carrier). When it captures an electron, the defect becomes negatively charged. When it captures a hole, the defect becomes positively charged.
- **Donor like:** These traps are neutral when filled with an electron and positively charged when empty (having donated their electron, which is equivalent to capturing a hole).
- **Acceptor like:** These traps are negatively charged when filled with an electron and neutral when empty.

2.4.2.3.2 Distribution

- **Single Level (Discrete):** Defects are defined as being at an energy level position within the bandgap.
- **Gaussian:** Defects are defined as being in a range of energies with a Gaussian-like distribution, characterized by a mean energy and standard deviation (char-

characteristic energy). This is usually more realistic for disordered organic materials.

- **Uniform:** Defects are defined as being distributed uniformly over a range of energy positions in the bandgap.

2.4.2.3.3 Energy Level (E_t):

Energy location of the defect state in the bandgap (usually described from E_V or E_C). Deep-level defects (close to mid-gap) are more effective as recombination centres than shallow-level defects (close to band edges), which act as recombination traps mostly

2.4.2.3.4 Density (N_t):

Number of defect states per volume. Higher defect densities mean more recombination.

2.4.2.3.5 Capture Cross-sections (σ_n, σ_p):

Effective areas depict the probability of a defect state capturing either an electron (σ_n) or a hole (σ_p) - to calculate the recombination rate with Shockley-Read-Hall (SRH) statistics.

2.4.2.4 Sources of Parameters:

The values for SCAPS-1D material properties are obtained from several sources. These are:

2.4.2.4.1 The literature

The major source of parameter values is literature values, which consist of both available experimental values and those resulting from the simulation of other studies.

2.4.2.4.2 Standard Values:

Certain materials have standard values for parameters such as electron affinity and dielectric permittivity, etc, which can be found in semiconductor physics textbooks & databases.

2.4.2.4.3 Educated Guesses:

In the case where little experimental data exists, educated guesses, based on the values of similar materials, may be employed

2.4.2.4.4 Parameters:

Certain parameters, such as the thickness of the active layer or the defect density, may be varied intentionally in the simulation to study their effect on the device performance.

2.5 Interface Modelling in SCAPS 1-D

Interfaces are important to the functionality of thin-film solar cells and thus influence charge-carrier transport, recombination, and device efficiency. SCAPS-1D allows different interfaces in a photovoltaic device to be modelled, including D/A (donor/acceptor) interfaces, active layer–transport layer interfaces, and transport

layer–electrode contacts. An accurate model of interfaces is important for understanding the contributions of recombination, charge trapping, and energy-level misalignment to losses. This section provides some important points on interface modelling in SCAPS-1D, covering interface types, defect states, recombination mechanisms, and energy level alignment.

2.5.1 Modelling of D/A Interface

The donor/acceptor (D/A) interface is an important part of the functionality of bulk heterojunction (BHJ) or planar heterojunction solar cells, as it strongly influences carrier separation and recombination. SCAPS-1D allows for the D/A interface to be modelled in depth by helping researchers define several parameters. The D/A interface is first modelled by defining its type, recombination rates and defect properties.

2.5.1.1 Interface Type:

SCAPS-1D enables the researcher to model either abrupt (sharp transition from one material to another) or graded (gradual change in composition from one material to another) interfaces. Abrupt interfaces will give rise to abrupt band offsets, and a graded interface will provide continuity of electric field changes to mitigate any discontinuities that may induce recombination losses. [\[86\]](#)

2.5.1.2 Interface recombination velocity:

The interface recombination velocity describes how fast carriers recombine at the interface. This is another adjustable parameter set by the researcher, where a higher

value indicates greater attraction to the recombination pathways. The value selected in SCAPS-1D will affect the experimental open-circuit voltage (Voc) and fill factor (FF) [86].

2.5.1.3 Interface defect states:

These defects are characterized by their density (cm^{-2}), their energy within the bandgap (eV), and their capture cross-sections (cm^2). For example, if a defect density of $10^{12}cm^{-2}$ is present at mid-gap energies, then it would trap a carrier from one of the active layers and cause non-radiative recombination [87].

2.5.2 Modeling of Active Layer/Transport Layer and Transport Layer/Electrode Interfaces

The active layer, transport layers (ETL and HTL), and electrode interfaces affect charge extraction efficiency and contact resistance. SCAPS-1D contains models to represent these interfaces, considering:

2.5.2.1 Contact Type (Ohmic vs. Schottky)

Ohmic Contact: This type of contact aims to have the least energy barrier for charge extraction, so it is modelled as having a small difference in work function, and as high doping at the interface [88]. This form of contact is certainly the type that would lead to effective solar cells.

Schottky Contact: This type of contact has a built-in potential barrier in the form of work function differences between the cathode and ETL or the anode and

HTL. This creates charge accumulation at the interface, which leads to increased recombination factors [2].

2.5.2.2 Interface defects and surface recombination velocity

As was noted for the D/A interfaces, defects located at the transport layer/electrode interface may trap carriers, leading to increased recombination [89]. SCAPS-1D contains a mechanism to set defect density and energy distribution.

The efficiency of charge collection is dependent on the surface recombination velocity (SRV) at the interface between the transport layer and the electrode. A high SRV at the ETL/cathode or HTL/anode interface leads to voltage losses. The higher the SRV the more likely electrons and holes will recombine at the interface and hence the scarcer the carriers to be collected.

2.5.3 Energy Level Alignment at Interfaces

The arrangement of energy levels (the conduction band minimum, E_c ; the valence band maximum, E_v ; and the Fermi level, E_F) at the interfaces of different materials controls all built-in electric fields, barrier heights, and charge transport pathways of the solar cell. The energy-level alignment at each interface is critical for charge injection and extraction; however, if the energy levels are misaligned, barriers can form and block charge flow. SCAPS-1D can model these energy-level alignments and optimise the interface, thereby minimising barriers. In an ideal situation, appropriate energy-level alignment will minimise charge-injection barriers and charge-recombination losses.

SCAPS-1D can be used to understand the equilibrium Fermi level at an interface

to determine a built-in potential and a barrier to carrier injection [89]. A 0.3 eV offset at the HTL/active layer interface, for instance, will reduce the Voc by about 150 mV. Defects at an interface will cause localised band bending in addition to the built-in potential, which complicates carrier transport. Including the defect charge contribution in Poisson's equation will mimic the localised band bending [90].

2.6 Simulation Methodologies and Result Evaluation:

This section describes the computational methods used to recreate the solar cell devices studied and how the results and key parameters were analysed. The aim is to develop a broad framework for understanding the device's physics, so that performance can be recreated and predicted, and its optimization enabled.

The simulation method will also be explained, providing a means to understand the numerical simulation of the photovoltaic device and the key output characteristics analysed, to characterise the performance of the devices studied. The general overall workflow for all the simulations would comprise two major work areas: (1) the calculation of the optical generation profiles and (2) the electrical simulation set-ups. The key output parameters collected or observed from the simulations will then be compiled and discussed.

2.6.1 Optical Generation Profile Calculation

The generation rate of carriers is an optically derived quantity that is an essential input to the simulated response, since it gives the density distribution in space of the photo-generated carriers and can be determined using the Beer-Lambert law, which

offers the exponential attenuation of the light intensity with depth, which depends upon the absorption properties. The expression is given in equation [2.6](#).

$$G(x) = \alpha\phi_0 \exp^{-\alpha x} \quad (2.6)$$

We can then separate it into those terms which depend on depth and those which are constant, and also from the Beer-Lambert law we can estimate the degradation in the optical generation rate through the measurement of the diminishing light intensity with penetration into the solar cell material. The full appreciation of the spatial distribution of the absorbed photons and the distinguishability effects of the various materials is of utmost importance in arriving to an understanding of the optical generation rate of the mobile electron-hole pairs. The inner workings of the SCAPS also can give further insight into the full optical model of the solar cell and of the varying layer effects of the incident light effects with regard to the surface of the solar cell [\[91\]](#).

2.6.2 Electrical Simulation Settings

Electrical simulations are complex processes involving settings for convergence criteria that will guarantee that the iterative methods used to solve the equations arrive at a stable solution. Voltage stepping is one of the critical simulation parameters. Voltage stepping refers to when the voltage is increased in increments when applying voltage to establish a simulation that reproduces the current-voltage (J-V) characteristics of the cell. This parameter represents the performance of the cell under different patterns of operation [\[92\]](#).

2.6.3 Key Output Characteristics to be Analyzed

2.6.3.1 Current-Voltage Test (J-V) Characteristics:

The J-V characteristics, both dark and illuminated are valuable for estimating V_{oc} , J_{sc} , FF, PCE, and primary metrics required to measure the overall performance of the solar cell [92].

2.6.3.2 Quantum Efficiency (QE) Spectra:

The QE spectra show how efficiently photons can be converted into charge carriers as a function of wavelength. This helps to understand how losses occur due to reflections, absorption, and recombination. [92]

2.6.3.3 Capacitance-Voltage characteristics (C-V):

C-V measurements reveal defects, built-in potential, doping, and carrier transport in the solar cell; therefore, it is essential to understand properties including defects and potential barriers to charge carrier transport [93].

2.6.3.4 Energy Band Diagrams:

Energy band diagrams show how the energy levels of the solar cell are impacted by voltage. This is important for visualizing the band bending, potential barriers and the ability to affect the charge carriers [91].

2.6.3.5 Electric field profiles:

These electric field profiles are important in responding how internal fields are a factor that separates carriers and how far the carriers travel. These fields are associated with the material behaviour of each component and processing that arises in the structural behaviour [94].

2.6.3.6 Carrier concentration profiles:

Carrier concentration profiles give an indication about charge carriers and how they will be distributed through the cell. This is significant in relating areas of higher recombination or poor carrier collection [94].

2.6.3.7 Recombination rate profiles:

Recombination rate profiles will include all of the different recombination mechanisms such as Shockley-Read-Hall (SRH) recombination, Auger recombination, and radiative recombination to help identify carrier loss mechanisms and regions of opportunity and/or photonic improvements.

2.7 Basic Operations of SCAPS 1D

The fundamental “Action window (Action panel)” of the simulation program is illustrated in Fig. 2.1. The window is divided into sections based on the functions involved in the simulation process. The tab on the left-hand side is called ‘set problem’, which allows you to access the ‘problem definition’ window shown in Fig. 2.2. This is the window where the device structure of the solar cell and associated material

parameters (listed in Table 2.1) are defined. There is an icon to the right-hand side at the bottom of the window for 'load' and 'save', where the 'load' icon is used to upload a file that has previously been saved with known structure and parameters.

By loading some examples that are already available in the SCAPS database, we can run some investigations into them. Using the save icon, we can also input the desired device structure and parameters into the database. When a solar cell device structure is defined in SCAPS, there are various layers in the device. Each layer can be thoroughly defined with material parameters and an absorption profile by clicking underneath the layers tab. The layer definition window opens as seen in Fig. 2.3.

On the bottom left-hand side, there is a tab called Set Absorption Model for defining the absorption profile of the material. There are some absorption profiles for certain materials in the SCAPS database. This data can be selected by selecting the option from file. Additionally, we can upload the absorption profile of a material externally as well by creating a new absorption profile. The absorption profile must be uploaded as a text file that meets specific requirements[i.e: . abs extension].

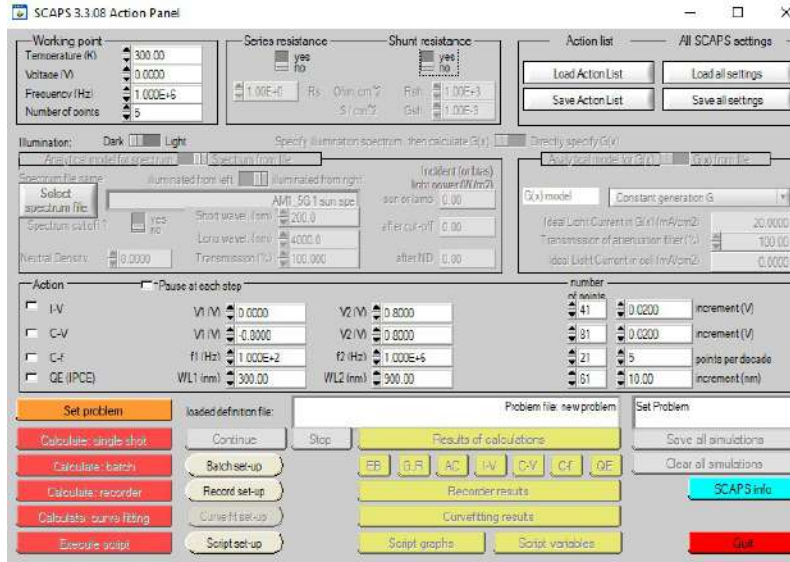


Figure 2.1: SCAPS 1D Action Panel

This is, for the most part, done by digitizing the absorption coefficient curves for the material at its wavelengths, which range from about 300 nm to 800 nm. The digitized values are converted into a text file for the absorption coefficient, with two columns: the first column corresponds to the wavelength in nm, and the second to the absorption coefficient in m^{-1} [95].

The defects and recombination in the respective layer are specified in the RHS of the layer definition window. SCAPS allows adding up to 7 defects per layer, and when you click the 'Add/Edit' tab, a new window opens. After saving the defect parameters, the layer definition window would display the respective carrier lifetimes and diffusion lengths for holes and electrons. There are provisions of giving the direct value of the capture coefficient corresponding to Auger recombination and the coefficient of radiative recombination in the top portion of the RHS of the layer

definition windows.

For organic solar cells, we kept these blanks since the dominating recombination has been the SRH recombination. All of the above specifications must be provided separately for each layer in the device structure. There are also provisions for adding, deleting, and duplicating any layers. Once all the layer parameters have been specified, the contact parameters for the front and back contacts should be provided. This can be done by clicking on the tab that means ‘front contact’, ‘back contact, which would initiate a new window for the contact parameters to be entered. The main parameters of a contact electrode is the work function, which can be directly entered in the panel for contact properties. Another means for getting the work function of the electrode is to make use of the ‘flat bands’ tab.

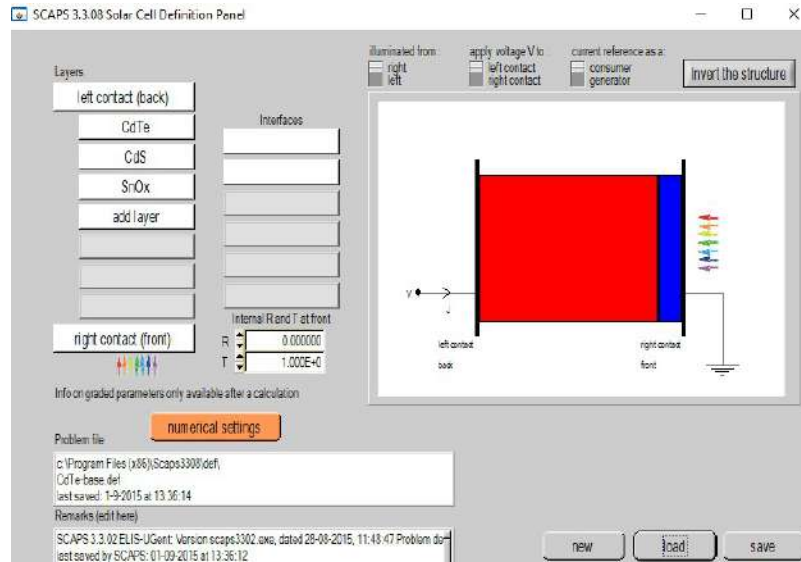


Figure 2.2: SCAPS 1D Simulation Panel

The final parameters to be specified in the layer definition panel concern the characteristics of interface defects. Upon clicking the tab next to layers, a box opens

where the data to be entered regarding the densities of the interface defects must be entered. All data concerning interface defects being encapsulated, the problem is defined. The device can be registered in SCAPS by clicking Save in the lower-right corner of the open box. The left-hand side concerns the simulation environment. The work point shows the conditions that must remain constant throughout the simulations. The temperature parameter must have constant values assigned across all simulations. The initial voltage in SCAPS is always 0 volts, which tends to increase to the specified value after a few iterations.

The frequency (f) is required to perform capacitance-voltage simulations, if required. The most important tab for the photovoltaic cell's working function is illumination conditions. The illumination may be specified as dark or light, and the spectrum may be selected accordingly. The illumination of the cell structure may be applied through the front or rear contacts. The standard spectrum used is the AM 1.5 G spectrum, with an incident beam power of 1000 W/m^2 . It is then necessary to specify what class of simulation is to be carried out.

SCAPS allows for Current-Voltage (I-V), Capacitance-Frequency (C-f), Capacitance-Voltage (C-V), and Quantum Efficiency (EQE) simulations. Following the selection of the box for the simulation type, depending on the need for the required simulation conditions, the box entitled calculate single shot must be pressed, so that the simulations can be entered. The action window also gives the possibility of batch simulations.

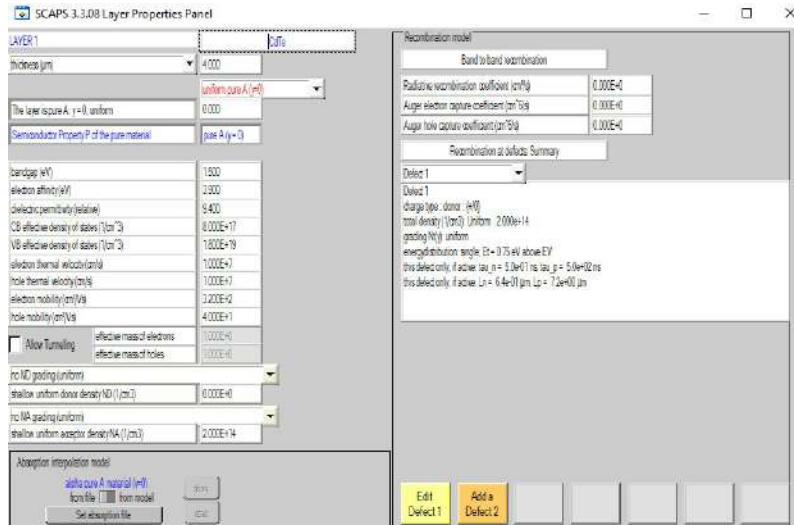


Figure 2.3: SCAPS 1D Layer Definition Panel

Sl No	Parameters	Unit
1	Thickness	nm
2	Band gap	eV
3	Electron affinity	eV
4	Dielectric permittivity	
5	CB effective density of states	cm^{-3}
6	VB effective density of states	cm^{-3}
7	Electron thermal velocity	cm/s
8	Hole thermal velocity	cm/s
9	Electron mobility	cm^2/Vs
10	Hole mobility	cm^2/Vs
11	Donor density	cm^{-3}
12	Acceptor density	cm^{-3}

Table 2.1: The input parameters in the layer definition panel

2.7.1 Comprehending the operational mechanisms of SCAPS

SCAPS 1-D solves the basic semiconductor equations given below in the bulk of the device structure. SCAPS forms a batch of coupled differential equations by applying suitable boundary conditions. Then, steady-state solutions of the system are calculated numerically.

$$\frac{\partial}{\partial x} \left(\epsilon_o \epsilon_r \frac{\partial \Psi}{\partial x} \right) = -q \left(p - n + N_D - N_A + \frac{\rho_{def}}{q} \right) \quad (2.7)$$

$$\frac{\partial J_n}{\partial x} - U_n + G = \frac{\partial n}{\partial t} \quad (2.8)$$

$$\frac{\partial J_p}{\partial x} - U_p + G = \frac{\partial p}{\partial t} \quad (2.9)$$

$$J_n = \mu_n n \frac{\partial E_{Fn}}{\partial x} \quad (2.10)$$

$$J_p = \mu_p p \frac{\partial E_{Fp}}{\partial x} \quad (2.11)$$

In this case, q stands for the charge and Ψ stands for the electrostatic potential. p , n stand for the free hole, free electrons. N_D and N_A are the concentrations of ionised donor-like and ionised acceptor-like doping, respectively. ϵ is the permittivity, and ϵ_0 is the permittivity of empty space. J_n and J_p are the current densities of electrons and holes, respectively. U_n is the rate at which electrons recombine, U_p is the rate at which holes recombine, G is the rate at which new particles are made, μ_n and μ_p are the rates at which electrons and holes move, and E_{Fn} and E_{Fp} are the quasi-Fermi levels for electrons and holes, respectively.

2.7.2 Sampling of the structure in SCAPS

The device architecture is sampled by passing through the computational mesh. The act of meshing at SCAPS has to comply with certain criteria. A coarse mesh will be located in the center of a layer, and a fine mesh will be initiated at the location of the interfaces of the device and the contact electrodes. During meshing, it will discretize two points at the structural interface. The software will optimize the mesh while conducting computations. There is also the option to optimize the mesh through each iteration by ticking the box worded 'recalculate the mesh during iterations' in the numerical interface [95].

If the option of mesh recalculation is selected in the numerical interface, the program will calculate the ratios of $\exp(q\phi/k_B T)$, $\exp(E_{Fn}/k_B T)$, and $\exp(E_{Fp}/k_B T)$, as well as the recombination rate (R) and generation rate (G) between adjacent mesh points. The numerical panel has a predetermined maximum ratio value. If a calculated ratio is larger than this value, the program will add mesh points to the simulation. Conversely, the program will remove mesh points if the calculated ratio is smaller than a predetermined minimum value. [95].

2.7.3 How to Reach the Solution in SCAPS

The computations can be initialized by clicking either on the calculate single shot tab to investigate how a parameter affects the action window or by clicking on the Batch tab to study the result of a range of values for a parameter and the influence on the action window after determining the manner of simulation (see section 2.7). First, in this instance, numerical methods are used to solve the equations in Section 2.7.1. The

well-known Gummel method (for the most part) & also the Newton-Raphson method (of which knowledge is secondary) form the basis of the more common numerical methods.

The numerical screen indicates the parameters of the numerical method. The greater the number of iterations employed, the quicker the calculation. The subject has to make a first approximation. The first approximation must be close to the real solution of the system; otherwise, if too far away, the iteration may be very long before the solution converges. Attention must be very carefully paid as to which first approximations are made, but the first approximation value does not enter the program. SCAPS is run through various values that are to be ascertained for the working point conditions, which dictate which reference is to be used for the calculation. SCAPS assumes that the quasi-Fermi levels in the device structure are all equal to 0 and that the potentials in the structure are also equal to 0 initially. Those have something to do with the first approximation when no external voltage is applied, and in the case of equilibrium in the dark. But, at the same time, SCAPS runs the short-circuit cases at one not too great from the first approximation value, so that an initial approximation value should be prepared [95].

2.7.4 SCAPS output parameter extraction

The output of the simulation can be assessed in the results panel from the action window itself. The most essential option for results analysis is the I-V panel. The current density-Voltage curve(J-V curve) is shown at the top LHS of the panel. The RHS has recombination currents. The output parameters of the solar cell are shown

below the J-V curve, with Voc, Jsc, FF, and PCE arranged as four columns. It is also possible to do a logarithmic scale for the J-V curve. The QE of the simulated device can then be analysed by clicking on the QE panel tab. Note that QE is external quantum efficiency (EQE). We can save the J-V curve and QE curve directly in any other folder by clicking on the 'save' tab on the top-right side of the window. The output / result window opens as seen in Fig. 2.4.

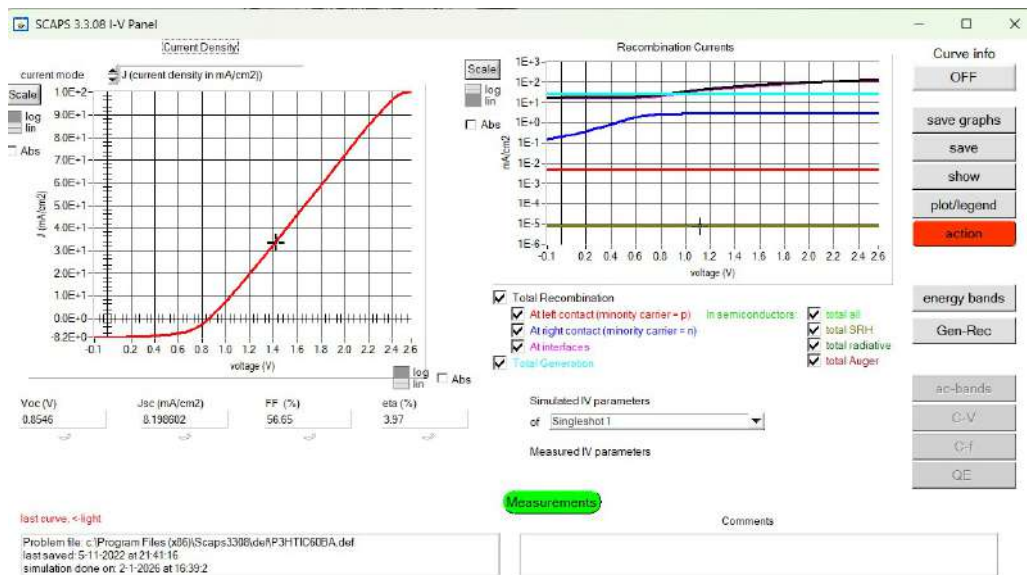


Figure 2.4: SCAPS 1D Result Panel

Chapter 3

Simulation Studies of P3HT:IC₆₀BA Organic Solar Cells using inorganic CuI as HTL

This chapter describes the effect of the inorganic hole transport layer CuI on bulk heterojunction polymer solar cells in which a fullerene IC₆₀BA acts as the acceptor. The various characteristics of the hole transport layer are studied in detail. Among the various systems investigated, bulk heterojunction polymer solar cells in which poly(3-hexylthiophene) (P3HT) serves as the donor and either phenyl C61 butyric acid methyl ester or one of its derivatives, i.e., IC₆₀BA, serves as the acceptor have attracted the most attention. However, one important challenge in optimizing cell operation is the design of suitable hole-transport layers that efficiently transport holes.

The non-crystalline polymer poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT: PSS) has been extensively studied as a hole-transport layer due to its high conductivity and transparency. However, owing to its hydrophilic and acidic nature, as well as the tendency of PEDOT: PSS to dissolve in water (which

may lead to device degradation), it becomes necessary to develop suitable alternative hole-transport layers to achieve long-term efficiency.

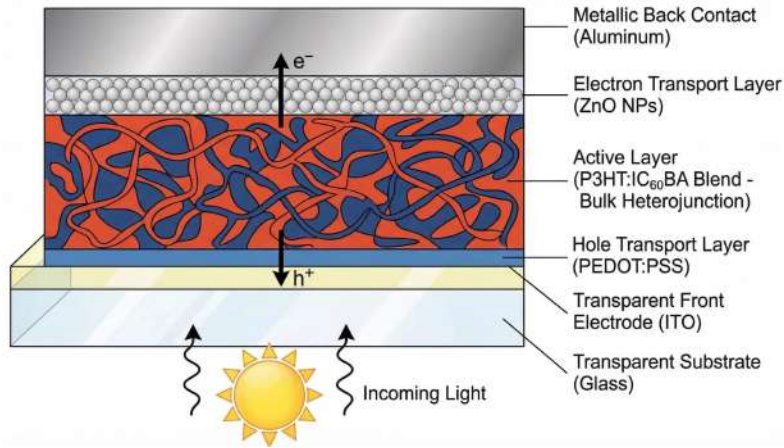
We will now make a direct comparison between the intriguing properties of CuI, such as its conductivity and solution-processability, with those of PEDOT:PSS and determine their relative effects in relation to the cell performance and stability of P3HT:IC₆₀ BA based devices. The work will encompass the study of the influence of copper iodide on power-conversion efficiency, fill factor, short-circuit current density, and open-circuit voltage, thus enabling a more thorough understanding of CuI's impact on solar cell performance.

3.1 Standardization

A stringent requirement on validation of a given software is a standardization with experimental works [10]. The device structure used for the purposes of standardisation is shown in Fig 3.1. The matching output parameters are given in Table 3.1 [experimental results and simulated results].

Parameters	Experimental	Simulated
Voc[V]	0.84	0.955
Jsc [mA/cm^2]	8.36	6.28
FF [%]	65	69.9
PCE[%]	4.22	4.19

Table 3.1: Evaluation of simulated parameters with experimental results

Figure 3.1: P3HT :IC₆₀BA Bulk Heterojunction Polymer Solar Cell Structure

The simulated bulk heterojunction structure of polymer solar cell analyzed by SCAPS 1-D, is given in Fig 3.2 [Glass/ITO/CuI/P3HT:IC₆₀BA/ZnO NPs/Al]. Fig. 3.3 represents the energy band alignment diagram. At the Active/CuI interface, the valence band offset is of +0.12 eV, allowing holes to be readily transmitted to the back metallic contact; while the conduction band offset is of + 0.5 eV, at the ZnO NPs/Active interface allowing the produced photoelectrons to readily move to the front contact.

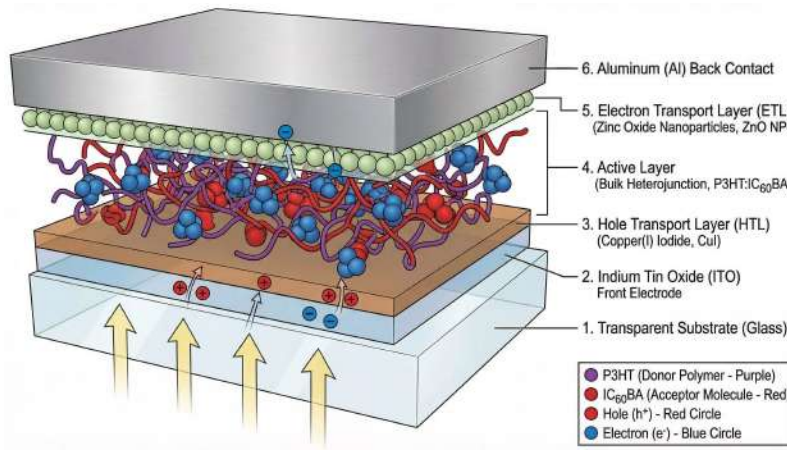


Figure 3.2: Simulated Polymer Solar Cell Structure

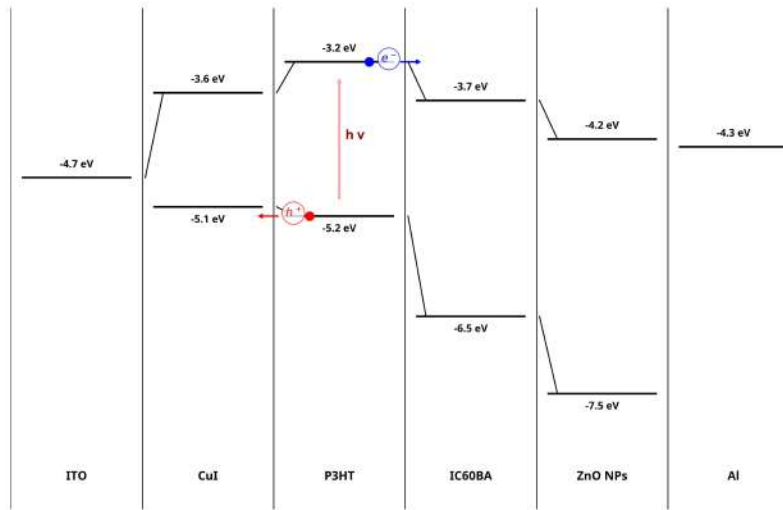


Figure 3.3: Energy Level Band Diagram

A summary of the simulation parameters for the configuration is shown in Table 3.2. The parameters are represented by the Energy bandgap [E_g], relative permittivity [ϵ_r], electron affinity [χ], electron and hole mobilities [μ_n , μ_p] and defect density terms [N_t]. The excitation densities for the conduction band and valence band are N_C and

N_V , respectively, while the densities of the acceptor and donor materials are N_A and N_D , respectively [96]. All the layers have the same values for the parameters not mentioned in Table 3.2. The average energy of the neutral Gaussian distribution defect is chosen to be 0.1 eV. The electron and hole capture cross-sections are adjusted to $9 \times 10^{-15} \text{ cm}^2$, and the thermal velocity of both carriers is fixed at 10^7 cm/s [97]. Two defect interfaces (HTL/Active and ETL/Active) are defined at a density of 10^9 cm^{-2} . The work function of the front electrode is set to 4.7 eV, and the back electrode has a work function of 4.125 eV. The absorption profile of the active layer and HTL was obtained from the literature data [98]. The simulation does not take into account optical reflectance, and to obtain the numerical parameters used in the studies, the control variable method was adopted.

The solar cell output parameters were obtained from the conditions reported in Table 3.2 as open circuit voltage (Voc), short circuit current density (Jsc), fill factor (FF), and power conversion effectiveness (PCE). By improving the characteristics of the polymer solar cells, the structure calibrated is employed in the formation of BHJ PSC with inorganic CuI as HTL.

Parameters	HTL	Active Layer	ETL
Thickness [nm]	27	250	46
E_g (eV)	2.98	1.68	3.2
χ (eV)	2.1	3.880	4.6
ϵ_r	6.5	3.3	9
$\mu_n(cm^2/Vs)$	10^7	10^7	10^7
$\mu_p(cm^2/Vs)$	10^7	10^7	10^7
$N_A(cm^{-3})$	2×10^{18}	0	0
$N_D(cm^{-3})$	0	3.2×10^{18}	1.1×10^{18}
$N_C(cm^{-3})$	10^{22}	1.6×10^{20}	2.2×10^{19}
$N_V(cm^{-3})$	10^{22}	10^{19}	1.8×10^{19}
$N_t(cm^{-3})$	10^{15}	10^{16}	10^{15}

Table 3.2: Simulation Parameters used

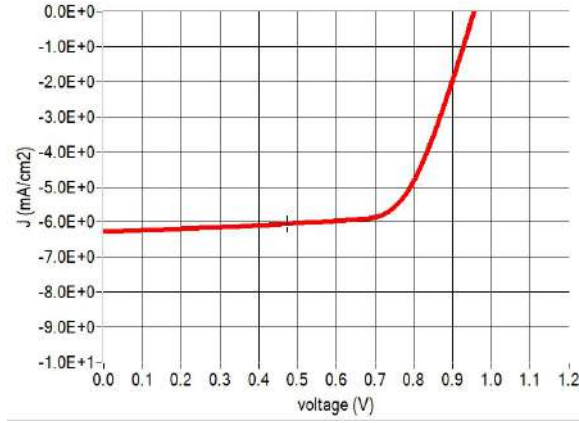


Figure 3.4: Simulated J-V Curve

3.2 Optimization of the cell structure ITO/CuI/P3HT:IC₆₀BA/ZnONPs/Al

3.2.1 Influence of Active Layer Thickness

A Polymer solar cell's [PSC's] performance is mainly determined by a complicated interaction of physical processes, such as light absorption, exciton dissociation, charge transport, and charge extraction. Of the important device parameters, the active layer thickness, which is that of a bulk heterojunction (BHJ, typically a polymer donor and fullerene or non-fullerene acceptor) may be the most important design factor since it governs the trade off between optical and electrical performance. The major challenge in PSC optimisation is how to exploit the inherent trade-off involved with increasing the active layer thickness, wherein increased light absorption is obtained at the expense of maximising current losses (recombination) associated with increased thickness.

The main purpose of the active layer is to absorb the incoming photons and produce excitons. It is obvious why the total light absorption is directly proportional to the thickness of the active layer, since the increase in thickness will lead to an increase in optical path length passing through the cell and thus to a greater number of photons absorbed and thus to a greater possibility of obtaining short circuit current density, J_{sc} . The relationship is, however, not a purely linear one owing to the optical environment within the device.

The PSC structure, being one of several layers of different refractive index (transparent electrode, active layer and metal back electrode for instance) acts like an optical cavity and the interference effects therein give rise to a non-uniform light

absorption pattern over the whole of the active layer. In very thin films (usually less than ~ 100 nm), with proportionately little light absorbed, optical losses due to interference effects become large, and Jsc is ultimately limited. Although increasing the thickness can lessen this, the optimal thickness for light absorption depends upon the particular material's absorption coefficient and also likely upon the optical design of the cell.

As illustrated in Fig. 3.5, the output is examined. Investigation of active-layer thicknesses from 50 to 400 nm reveals a direct correlation with performance. Jsc, PCE, and Voc all increase with thickness. Device performance is best at 400nm (max Jsc is 8.24 mA/cm², PCE is 4.13%, and Voc is 0.830 V). Increased thickness may be responsible for this by boosting light absorption and photogenerated carriers. As the thickness increases, more photons are absorbed, and hence, PCE increases. Jsc rises because of a higher likelihood of exciton dissociation or a lower rate of recombination of non-geminate charge couples. As thickness increases, FF first rises before beginning to fall. The maximum value of 62.29% occurs at a thickness of 150 nm. This could be due to the increase in series resistance with thickness.

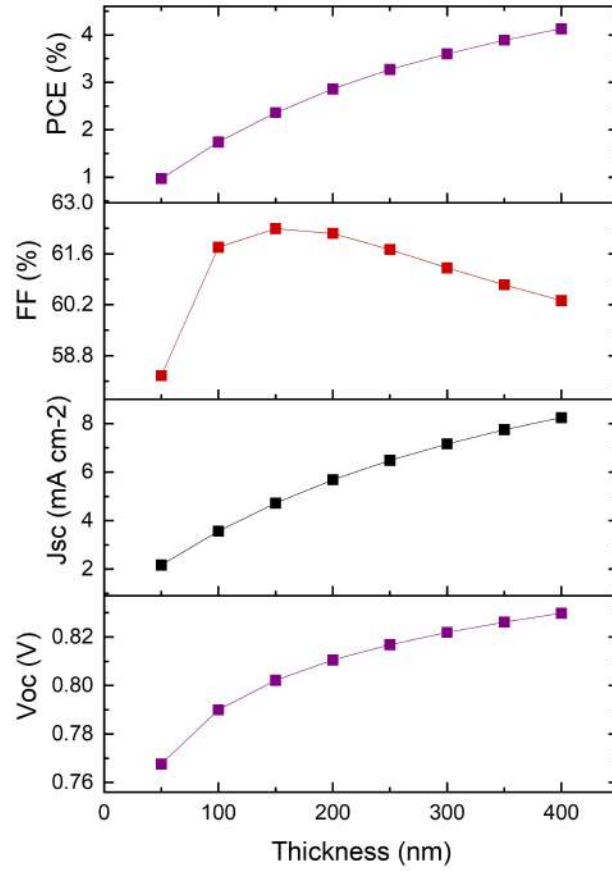


Figure 3.5: Influence of Active Layer thickness

3.2.2 Effect of HTL (CuI) properties

On solar cell performance, the effects of HTL layer [CuI] parameters, including thickness, doping concentration, mobility and electron affinity, were examined.

3.2.2.1 Influence of thickness

The thickness of the HTL is a critical parameter that must be carefully optimized, as it influences both the electrical and optical properties of the device. The output of BHJ PSC is heavily influenced by the hole-transporting layer. Better hole transport and charge collection at the electrodes are made possible by a superior HTL selection.

The HTL layer thickness is increased from 50 to 400 nm, and Fig. 3.6 illustrates the impact on output parameters. From the graph, we find that V_{oc} , J_{sc} , and PCE increase, while for FF it is found to decrease initially with HTL thickness and then increases.

The variation in V_{oc} is mainly due to improved interfacial quality and reduced recombination. J_{sc} varies due to enhanced photon absorption and, consequently, a higher rate of exciton generation. Variation in FF accounts for the increase in series resistance. PCE varies due to the cumulative effect of all other output parameters. The highest possible V_{oc} , J_{sc} , and PCE occur at a thickness of 250nm.

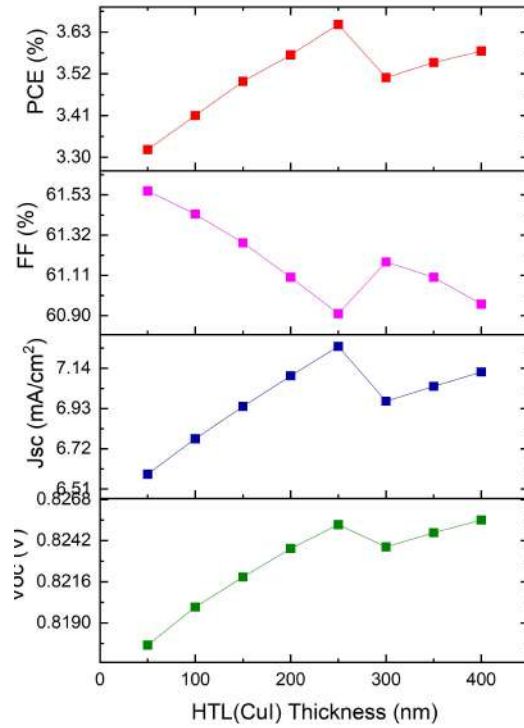


Figure 3.6: Influence of thickness

3.2.2.2 Influence of doping density

Another important HTL parameter is the doping density, which is preferably tuned for each device, as it also plays an important role in the device's electrical and optical properties.

Charge accumulation due to higher doping density in the HTL yields a stronger electric field at the HTL/active layer interface and reduced interface recombination, resulting in higher power conversion efficiency.

Also noted was a corresponding dependence of fill factor and power conversion

efficiency of devices. Fig. 3.7 depicts the effect of hole transport layer doping density on output characteristics.

The increase in V_{oc} is primarily an electrochemical effect driven by the shift in the HTL's Fermi level. The increase in fill factor accounts for a reduction in series resistance and increase in the extraction of holes. The decrease in J_{sc} is a consequence of charge transport and optical losses introduced by excessive doping.

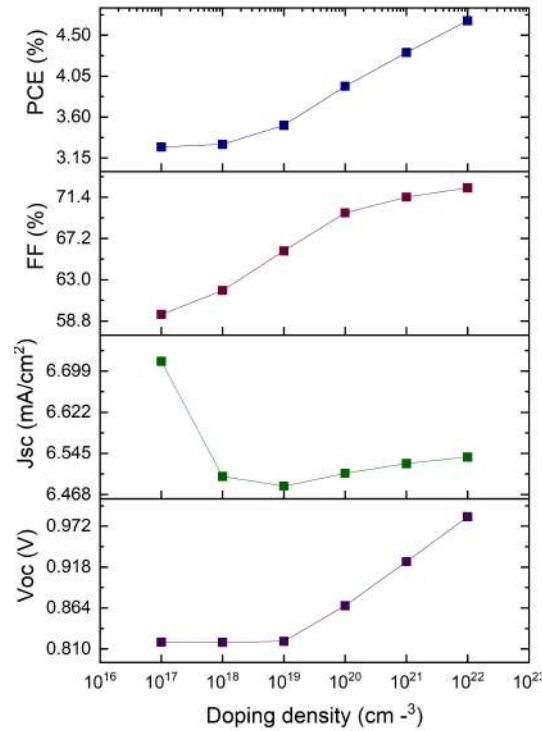


Figure 3.7: Influence of doping density

3.2.2.3 Influence of Hole Mobility

The hole mobility of HTL layer is the parameter that has the most direct effect on the performance parameters of the solar cell, in that it indicates the rate at which positive charge carriers (holes) can migrate through it. The HTL layer's hole mobility is raised from 10^{-6} to $1 \text{ cm}^2/\text{Vs}$, and the outcomes are investigated. The output appears to validate the material's increased conductivity due to the mobility effect.

The PCE is maximum at $10^{-3} \text{ cm}^2/\text{Vs}$. High hole mobilities lead to a higher V_{oc} due to enhanced hole extraction efficiency and a reduction in charge recombination. J_{sc} will reduce as a consequence of the balance of charge carriers and increased recombination at the interfaces.

The fill factor increases because of improved hole transport, which reduces the series resistance in the solar cell and enables more efficient power extraction. In general, high hole mobilities in the HTL lead to higher PCE in the solar cell due to efficient hole transport, leading to lower levels of charge recombination [99].

The optimum cell output is represented in the assessment, considering the work function of the metal contacts, defect states, and the amount of light transmitted or reflected from the front/back contacts [100]. Fig. 3.8 depicts the effect of hole mobility.

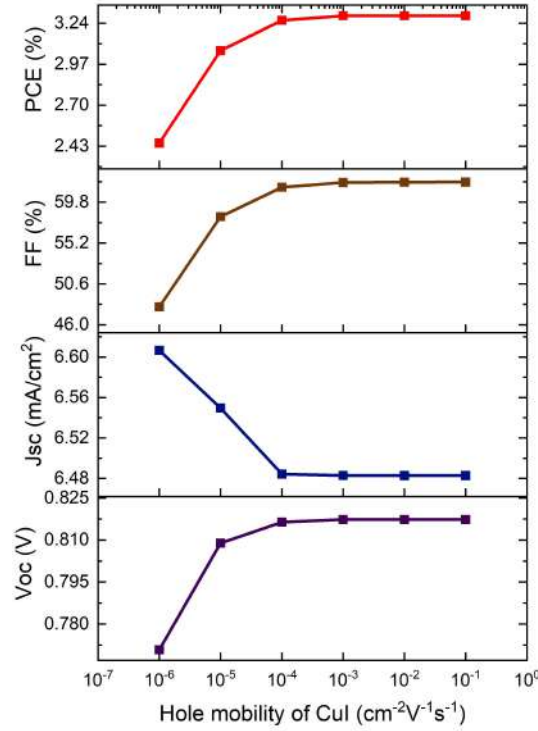


Figure 3.8: Influence of Hole Mobility

3.2.2.4 Influence of Electron Affinity

The electron affinity of the hole-transport layer plays a crucial role in determining the energy alignment at its interface with the active layer, which, in turn, significantly impacts the efficiency of charge extraction and, consequently, the overall performance of polymer solar cells [101].

This energy alignment is crucial because it helps eliminate energy losses and maximize the open-circuit voltage by promoting efficient hole extraction while preventing the simultaneous extraction of electrons [102]. An essential factor for proper electron

affinity in the hole-transport layer is that the polymer's ionization potential is now closer to the active layer's valence-band energy, which seems to favour improved hole extraction efficiency and open-circuit voltage [103]. For example, a higher electron affinity of the hole transport layer leads to a higher open-circuit voltage, up to an acceptable level, by reducing the valence band offset between the layers, thereby improving charge separation and reducing recombination losses.

Conversely, a greater electron affinity may also lead to a greater number of electrons being available, at least in the form of a higher concentration [83]. HTL's electron affinity is changed between 1.8 and 2.6 eV, and the influence on output performance is displayed in Fig 3.9. At 2.4 eV, the PCE of 3.56% and the Voc of 0.9234 V are achieved. At 2.8V, an FF of 61.82 % is obtained. The BHJ PSC's active and transport layers benefit from better energy alignment and improved charge transport when the best electron affinity reduces the band gap effect.

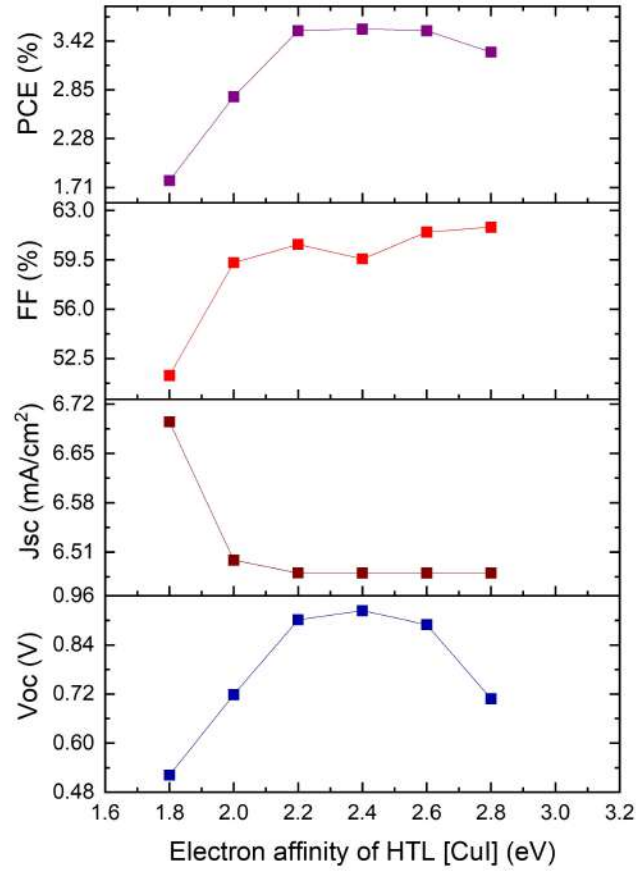


Figure 3.9: Influence of Electron Affinity

3.2.3 Effect of ETL (ZnO NPs) properties

3.2.3.1 Influence of Thickness

The thickness of ETL is varied from 2nm to 10nm. As a thicker ETL increases the series resistance, it is easier for the charge carriers to be lost before being extracted from the photoactive layer, and it results in the loss of a buffer layer to stabilize the

photoactive layer against degradation due to the influence of light on the structure [98].

A higher thickness of the ETL will result in a higher level of absorption of light by the ETL material than if the ETL were thinner, resulting in less light being absorbed by the photoactive layer and subsequently resulting in a reduction in the short-circuit current-density [104].

An overly thick ETL will result in poor electron transport through it and into the electrodes, because electrons have to travel a longer distance through the ETL to reach the electrodes, increasing the probability of charge-carrier recombination before extraction at the electrodes. In addition to the increase in the length of the electron-transport route, there will also be a potential drop along the length of the ETL due to the resistance of the ETL, resulting in a decrease in the open-circuit voltage.

The decrease in the output characteristics of the solar cells can be made worse by the fact that the band edges are not optimized at the interface regions, which will result in the possibility of negative charge-transfer dynamics and will create a greater issue of defects in the spectrum of the over-thick layer, which will act as charge-carrier-recombination centers. Additionally, the reduced ability of the solar cell to convert photons in the visible spectrum into electrical energy will result in an additional reduction in the short-circuit current density of the solar cells when they use an overly thick ETL, which will negatively affect the overall power-conversion efficiency of the solar cells [105]. Fig. 3.10 represents the graphical representation of the variation of output parameters with the thickness of the electron transport layer

of a polymer solar cell.

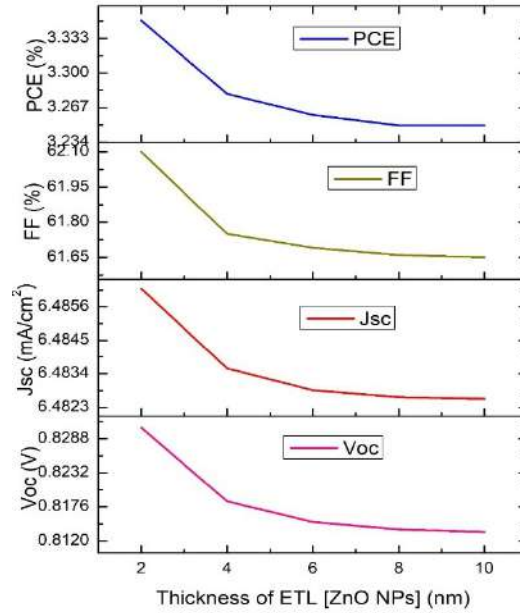


Figure 3.10: Influence of Thickness

3.2.3.2 Influence of Electron Affinity

The electron affinity is changed from 3.8eV to 4.4eV. A robustly excessive electron affinity in the electron transport layer negatively affects the open-circuit voltage, short-circuit current density, and power conversion efficiency by virtue of the large energy barrier it forms, which slows electron extraction and increases charge recombination at the interface. This also occurs because the excessive electron affinity leads to an unfavourable band alignment, which increases interfacial recombination and reduces the collection of charge carriers.

This also underscores the importance of tuning the electron affinity to achieve

an ideal balance between sufficient and efficient charge extraction and minimal recombination, leading to optimal device performance [106]. Fig 3.11 shows the graphical representation of the variation of output parameters with an increase in the electron affinity of the electron transport layer.

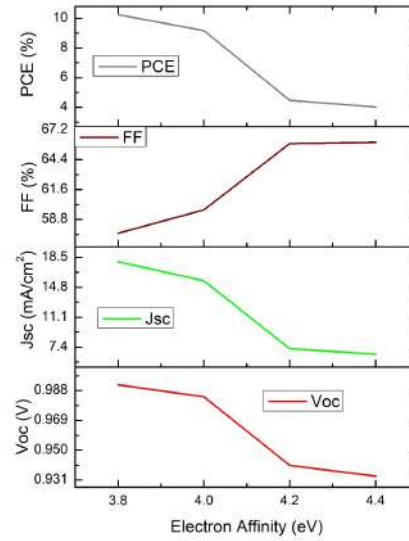


Figure 3.11: Influence of Electron Affinity

3.2.3.3 Influence of Doping Density

Doping level within the ETL is an important factor influencing the physical properties of polymer PVs. Screening of the donor-acceptor interface by a high doping density enhances the extraction of charge carriers and speeds up the transport process at the expense of recombination loss [107].

The doping density is varied from 10^{17} to 10^{20} cm^{-3} . The fact that the doping densities are such that the increased conductivity of the electron transport layer gives

rise to increased transport of electrons in the solar cell leads to decreased build-up of electrons in the solar cell, both of which lead to increased short circuit current density and fill factors for the solar cell [108]. Also, if the concentration of doping in the electron transport layer could be optimized and if the parameters of the electron transport layer were optimized, a more favourable high work function for use in the solar cell could be obtained, which would vastly improve electron extraction and could lead to increased open circuit voltage of the solar cell.

Ultimately, the increased enhancement of the transport of the charge carriers and the associated decrease in recombination would lead to increased efficiencies of conversion of the power of the solar cell [109, 110]. Fig 3.12 represents the respective graph.

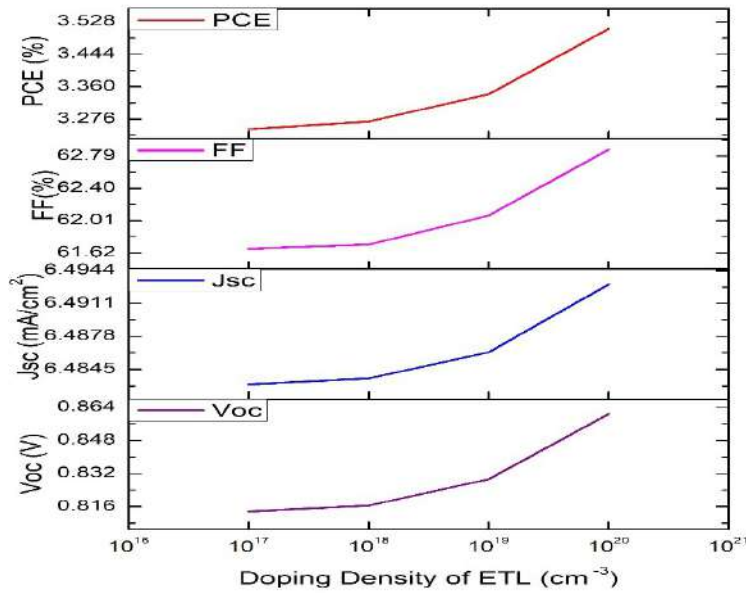


Figure 3.12: Influence of Doping Density

3.2.3.4 Influence of Electron Mobility

The electron mobility of ETL is changed from 10^{-6} to $10^{-2} \text{ cm}^2/Vs$. With elevated electron mobility in the electron transport layer of a polymer solar cell, both the open-circuit voltage and the power conversion efficiency are generally decreased. At the same time, the fill factor and the short-circuit current density increase. The latter two phenomena occur because of improved charge extraction and reduced recombination within the device. There is a complex relationship among these parameters such that optimized electron transport results in more efficient charge collection, which increases both the Jsc and FF, however, it also potentially alters the energy level alignment as well and creates additional nonradiative recombination pathways, ultimately decreasing both Voc and PCE. Electron mobility can improve charge-carrier separation and transport, thereby increasing Jsc and FF by reducing charge-carrier recombination [111]. However, high electron mobility may result in charge carrier transport imbalances that can elevate recombination at the back contact, thereby reducing Voc and PCE [112]. Fig3.13 shows the respective graph.

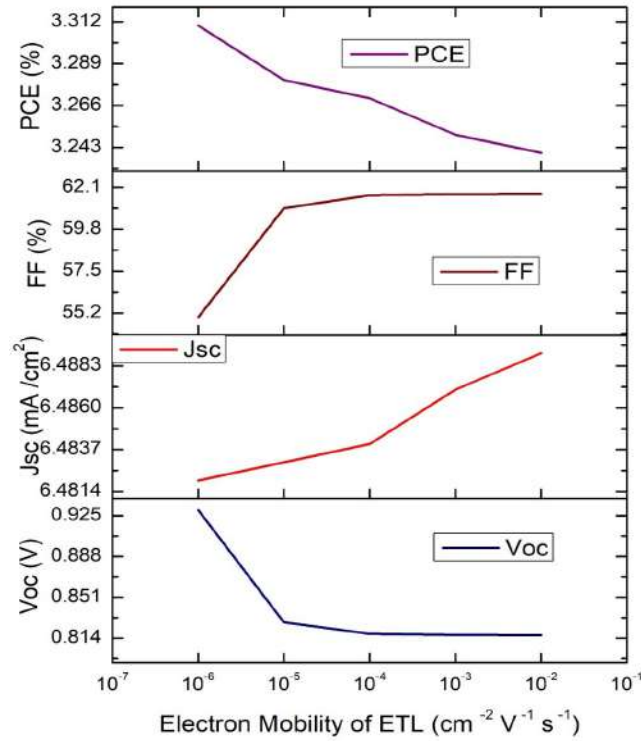


Figure 3.13: Influence of Electron Mobility

3.2.4 Effect of defect density at HTL/Active Layer interface

Defect density at the hole-transport layer and active-layer interfaces is significant for the electrical characteristics of organic polymer solar cells because it can influence charge-carrier recombination and charge-collection efficiency. If there are large numbers of defects at these critical interfaces, then they may facilitate trap-assisted recombination that will be detrimental to the open-circuit voltage and fill factor. The defects will also result in an increase in the leakage current and a decrease in the short-circuit current density because of poor charge separation and transport[113].

Additionally, defects at this interface can lower charge mobility, leading to charge accumulation.

Charge accumulation increases series resistance, reducing the fill factor. Thus, a high defect density at the interface can lead to increased charge-carrier recombination, resulting in a rapid decrease in J_{sc} . High defect density at the interface can make a fast drop in the fill factor. Defects act as traps for charge carriers, leading to energy loss and reduced efficiency. Therefore, defects arising from either unfavourable band alignment between the HTL and active layers or from the inherent properties of the materials used can significantly degrade the performance of the photovoltaic device by providing additional pathways for recombination at the HTL-active layer interface.

The defect densities of the HTL/Active layer contact are varied from $10^7 cm^{-3}$ to $10^{12} cm^{-3}$. Increased defect density results in more traps and lowers cell performance. For better output, low interface defect densities are desirable. The effects of changing defect density at the HTL/Active layer contact are depicted in Fig. 3.14. The optimized values are obtained at $10^7 cm^{-3}$.

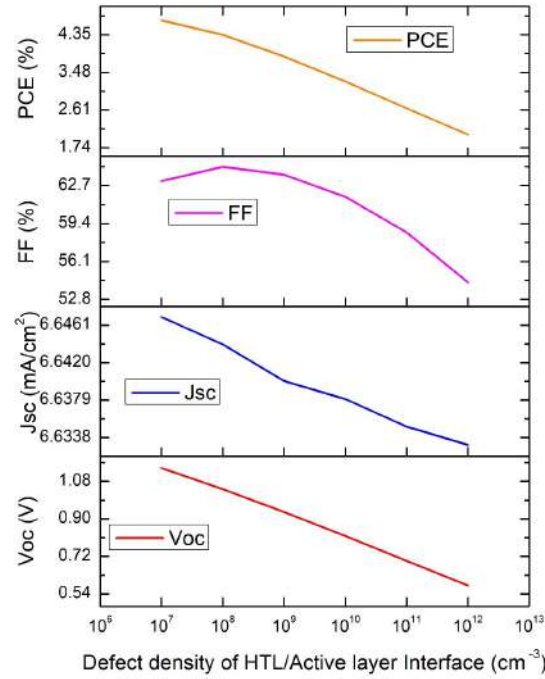


Figure 3.14: Influence of HTL/Active Layer Interface Defect Density

3.2.5 Effect of defect density at Active Layer/ETL interface

The interfacial disorder is a site for exciton recombination, thereby lowering the overall power conversion efficiency [114]. Here, the interface defect density is varied from 10⁸ to 10¹² cm⁻³.

Specifically, a high density of defects at the interface between the active material layer and the electron transport layer may be sites where charge carriers become trapped, thereby slowing their transport to the electrodes.

Thus, a greater opportunity for charge-carrier recombination occurs before extraction. This higher probability of recombination is likely caused by the presence of

pinholes within the solar cell, which would lead to further degradation of the solar cell's performance by increasing the diffusion length of the charge carriers [115]. Not only does the trapping action lead to fewer carriers available for extraction, but it also results in a substantial loss in the open circuit voltage because of reduced quasi-Fermi level separation, leading to less maximum potential difference across the solar cell [102].

Short-circuit current density is also negatively affected, since an increase in defect density increases the number of defects that capture photogenerated carriers, thereby decreasing the probability that carriers achieve sufficient mobility to reach the electrodes and ultimately decreasing the total current produced by the solar cell [116]. Non-radiative recombination occurring at defects on the interfaces decreases the power conversion efficiency of the solar cells due to decreased fill factor since the charge carriers are redirected to non-productive areas before they have an opportunity to perform useful external work [117]. Fig 3.15 shows the respective graph.

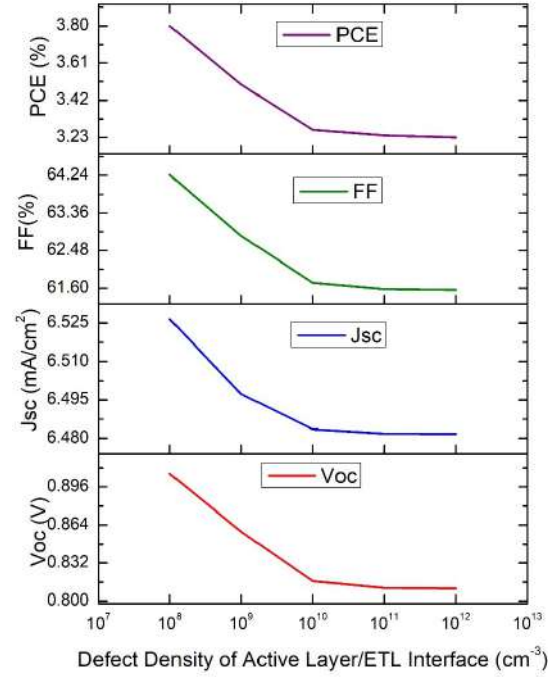


Figure 3.15: Influence of Defect Density of Active Layer/ETL Interface

Device Parameters	Thickness [nm]	χ [eV]	μ_n [cm^2/Vs]	Final
Voc[V]	0.8252	0.9234	0.8173	1.2170
Jsc [mA/cm^2]	7.2543	6.4802	6.4829	6.9243
FF [%]	60.91	59.57	62.01	73.95
PCE[%]	3.65	3.56	3.29	6.23

Table 3.3: Optimized Device Performance Parameters

3.3 Conclusion

Overall, performance of bulk-hetero junction (BHJ) polymer solar cells can be significantly enhanced by carefully optimizing several parameters of the hole-transport layer (HTL). Cell performance benefits from an increase in HTL thickness, which, for

the best results, was approximately 400 nm thick. Reduction of defect density in this layer was also important. The optimized device performance parameters are shown in Table 3.3 and the simulated J-V curve is shown in Fig 3.4. This work constitutes the first report of a high-efficiency BHJ solar cell with an inorganic Copper Iodide (CuI) hole-transport layer, demonstrating the great potential of CuI for application in next-generation polymer solar cells.

Chapter 4

Simulation Studies of P3HT:IC₆₀BA Organic Solar Cells without ETL

Following the extensive simulation study in Chapter 3 on the optimum P3HT:IC₆₀BA polymer solar cell, which employed a CuI hole-transport layer (HTL) together with a ZnO nanoparticle electron-transport layer (ETL), we now shift our focus from devices with interfacial layers to the underlying device physics in the absence of such interfaces. One of the key conclusions drawn from the comparative analysis in Chapter 3 is the superior performance achieved when CuI replaced PEDOT: PSS as HTL. But when Polymer SC structure is studied under simulation, it was found that PSC with CuI as HTL attained an efficiency of 10.10% when compared to PSC Structure with PEDOT :PSS [without ETL], showing a PCE of 10.83%. This observation underscores the crucial role of the HTL, even purely in terms of extraction and recombination dynamics, and also indicates that this behaviour is strongly material dependent. To establish a clear baseline for understanding these subtle interfacial effects, Chapter 4 presents a simulation study of the P3HT:IC₆₀BA bulk-heterojunction active layer

entirely without ETL. The aim is to examine the essential charge-generation, transport, and recombination processes within the active layer, allowing direct and quantitative comparison with the high-performance architecture in Chapter 3 and the intermediate device structures, thereby clarifying how the performance becomes limited and why selective contacts are required in this material system.

In polymer solar cells, electron transport layers (ETL) are used for collecting the electrons but could limit the performance of the device because of issues with the material such as TiO_2 degrading from UV light or trapping the electrons because of defects in the layer. Studies have shown that removing the ETL can create an increase in efficiency and lower the cost of production. In some cases, the performance of the ETL materials can be greatly affected by lowering the thickness of the layer, so the material becomes a scalability issue for mass production. An optimization of a standard structure replaced without ETL [ZnO nanoparticles] leads to a most significant enhancement of the efficiency from structures with ETL proving evidence to high efficiency polymer solar cells.

4.1 Standardization

The software is validated by comparing the simulated results to the experimental one. The standardization results are shown in the Table [4.1](#). The Simulated polymer solar cell [PSC] structure is represented by Fig. [4.1](#).

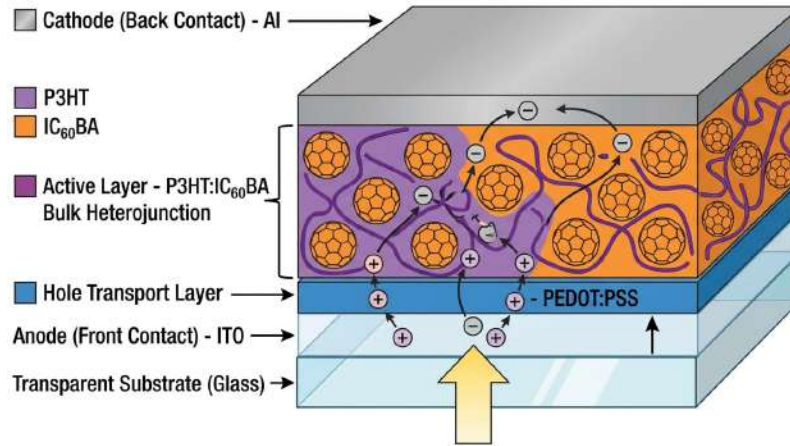


Figure 4.1: Simulated Polymer Solar Cell Structure

Parameters	Experimental	Simulated
Voc[V]	0.84	0.85
Jsc [mA/cm ²]	8.36	8.21
FF [%]	59	57
PCE[%]	4.1	3.97

Table 4.1: Evaluation of simulated parameters with experimental results

4.2 Optimization of the cell structure ITO/PEDOT:PSS/P3HT:IC₆₀BA/Al

4.2.1 Effect of Active Layer properties

4.2.1.1 Influence of thickness

The active layer thickness is varied between 300 nm to 700 nm. As thickness increases, Voc, Jsc and PCE increase, but FF decreases. The corresponding graph is given in Fig 4.2. Voc increases due to the production of more charge carriers. Variation in Jsc accounts for more light absorption and FF declines due to increase in series resistance

and recombination. Variation in PCE accounts for the combined effect of all other output parameters.

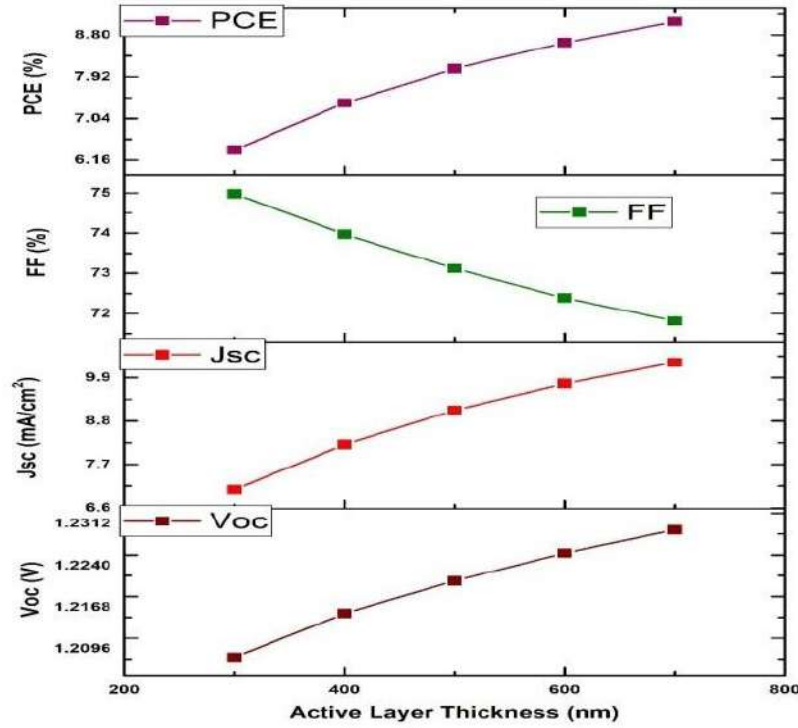


Figure 4.2: Influence of thickness

4.2.1.2 Influence of defect density

The defect density of the active layer is increased from 10^{16}cm^{-3} to 10^{20}cm^{-3} . Voc increases with defect density, while FF and PCE vary similarly. Jsc is found to decrease with increasing defect density. Optical techniques based on sub-bandgap absorption or emission can also provide insights into the defect density in the active layer. It was discovered that optimal performance occurs at 10^{17}cm^{-3} , with an efficiency of 5.87 %. These variances are shown in Fig 4.3.

Defect density can act as trapping sites, leading to non-radiative recombination which impacts the Voc. The Jsc is lowered by high defect density due to increased recombination and trapping of charge carriers. The lack of symmetry in the transit rate of electrons and holes will lead to a charge being built up in the device (termed a space charge), and this will inhibit further charge extraction and thus reduce the fill factor. Traps can serve to balance the transport, slowing down the more mobile carrier for a time and arranging for a more uniform transit of charge, and reducing the deleterious effects of space charge. Influence of Voc, Jsc, and FF results in the variation of PCE. Improved material purity, optimized fabrication processes, and careful control of the polymer morphology helps in reducing defect density for better performance.

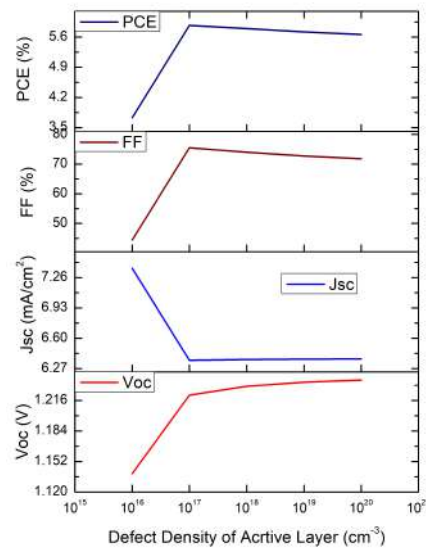


Figure 4.3: Influence of defect density

4.2.1.3 Influence of electron affinity

The electron affinity of the active layer ranges from 3.5 eV to 4 eV. Simulation revealed an ideal value of 5.75% efficiency at 3.7 eV electron affinity. This is the result of the enhancement in exciton dissociation and charge transport while reducing recombination losses. A graphical illustration is shown in Fig [4.4](#)

If the electron affinity of the acceptor is high, it ensures a bigger energy difference, which may increase Voc; however, if it is too high, it may also increase recombination rates, which may negate the advantage. If the electron affinity is increased the more it leads to better electron transport to the electrode, thus reducing recombination in the active layer. This leads to a better charge collection overall and thus higher Jsc and FF. If, however the electron affinity is not optimized, it leads to poor charge transport and losses on recombination. Proper electron affinity means ideal energy level alignment is achieved and charge transfer facilitated. Tuning the electron affinity ideally improves charge transport and reduces recombination, which improves Jsc and FF. Well-matched electron affinity helps in reducing losses by recombination, leading to better overall efficiency of the device.

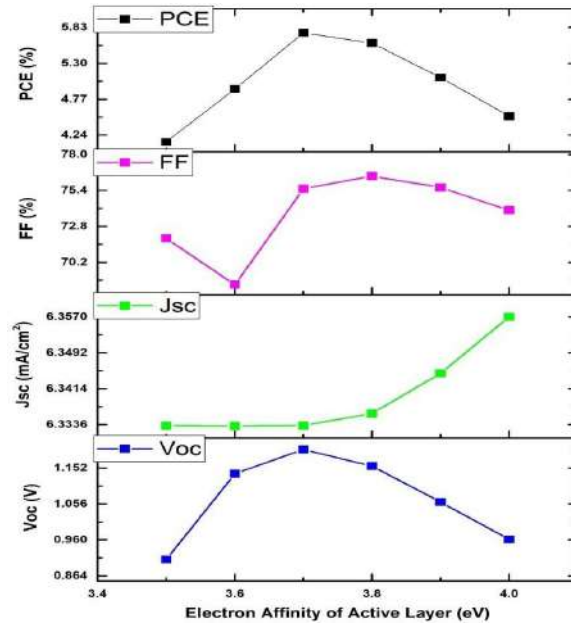


Figure 4.4: Influence of electron affinity

4.2.1.4 Influence of carrier concentration

The carrier concentration of the active layer is increased from 10^{17} cm^{-3} to 10^{21} cm^{-3} . In this simulation, the maximum Voc was 1.2013 V and the maximum PCE was 5.76 % at 10^{18} cm^{-3} . The maximum value of Jsc was found to be 6.333860 mA/cm^2 , and the maximum value of FF was 76.08 % at 10^{21} cm^{-3} . The respective figure is given in Fig 4.5. A higher doping concentration can provide more favourable energy level alignment within the layers to reduce recombination loss and possibly enhance Voc. Jsc increases with increasing doping due to improved transport of charge carriers to the electrodes. Increased carrier mobility, reduced trap-assisted recombination, and improved charge extraction lead to an increase in fill factor and power conversion efficiency.

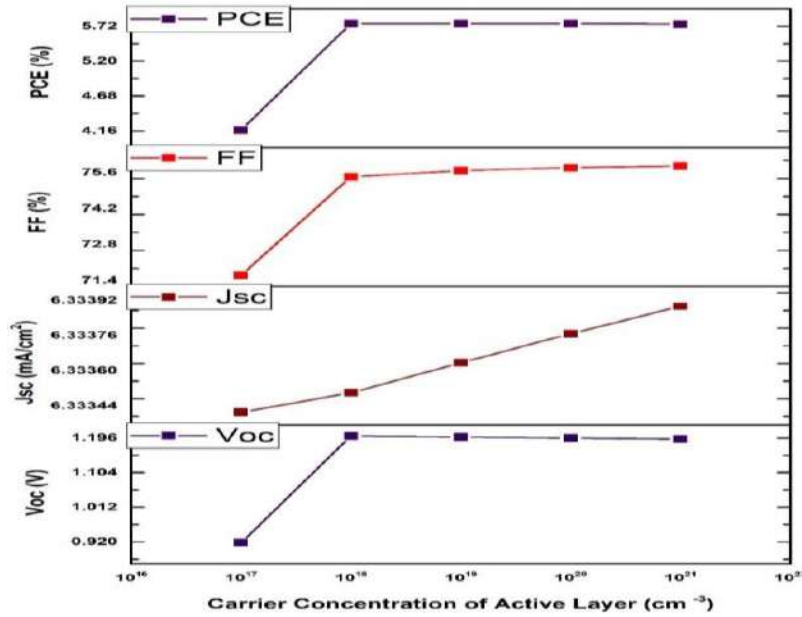


Figure 4.5: Influence of carrier concentration

4.2.2 Effect of HTL properties

4.2.2.1 Influence of thickness

The thickness of HTL is varied from 25 nm to 45 nm. The optimized values are obtained for the thickness of 30 nm. The corresponding diagram is shown in Fig. 4.6. An optimal HTL thickness allows for efficient hole transport and extraction, which helps to maintain a high built-in potential and thus results in a higher Voc. An appropriately thick HTL also allows for efficient hole transport to the anode without excessive recombination losses, resulting in a higher Jsc. A very thick HTL increases series resistance, which reduces the FF. An optimal HTL thickness increases the Voc, Jsc and FF, which results in a higher PCE.

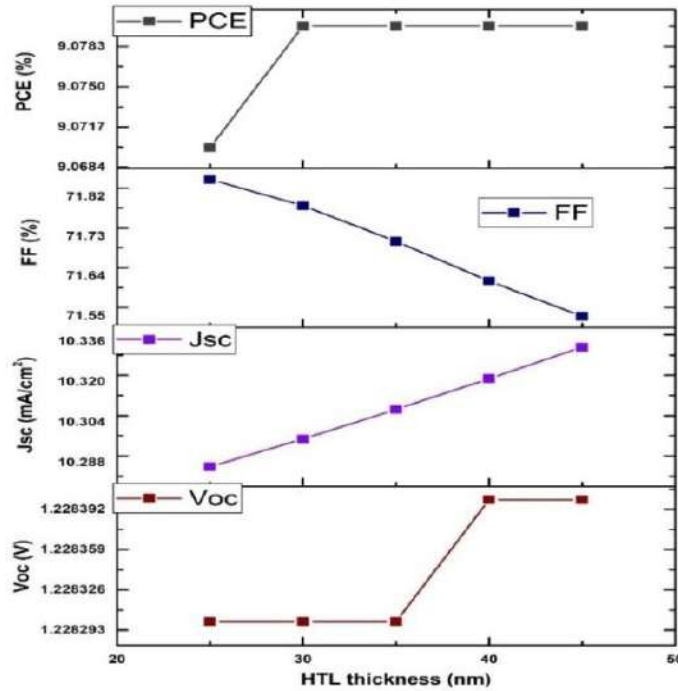


Figure 4.6: Influence of thickness

4.2.2.2 Influence of band gap

The band gap of HTL is varied from 1.4 eV to 1.8 eV. It was found that Voc, FF and PCE increase with the band gap and reach the highest at 1.8 eV. In Fig 4.7 the variation of the output parameters are shown. It follows from the bandgap that Voc is also directly proportional to bandgap; hence higher the bandgap higher will be Voc, and the lower will be the radiative recombination rate. Jsc tends to decrease with increasing bandgap because there are comparatively fewer electrons produced since few photons have enough energy. Having an HTL with appropriate band gap ensures that the energy level align very well with the active layer, and hence lowers the energy losses for hole extraction resulting in a higher Voc.

If the band gap is mismatched, it can create an energy barrier for holes, making hole extraction difficult and leading to higher recombination and lower Jsc. If the band gap is well-aligned, then recombination and series resistance are lowered resulting in higher FF. A proper band gap for an HTL ensures efficient extraction of charges and the re-combination is less, and hence higher Voc, Jsc, and FF, will equal maximum PCE. The HTL band gap should be optimized to align closely with the active-layer energy levels to enhance the performance of polymer solar cells.

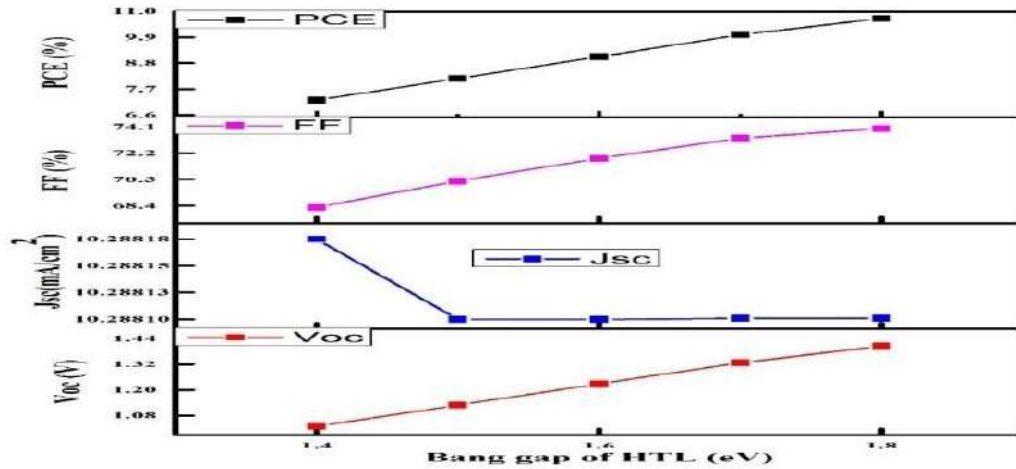


Figure 4.7: Influence of Band Gap

4.2.2.3 Influence of electron affinity

The electron affinity of the HTL is varied from 3.2 eV to 3.6 eV. The optimal performance is achieved at an electron affinity of 3.4 eV. Changes in the parameter outputs are shown in Fig. 4.8. An HTL with suitable electron affinity will match its energy levels with the active layer, hindering leakage and recombination at the HTL/anode interface.

If the electron affinity is mismatched, it may imply ineffective charge separation and therefore increased recombination losses, resulting in a reduced Jsc. Suitable electron affinity levels commensurately reduce the recombination losses as well as ensure efficient charge transport, which collectively lead to higher FF (i.e., 3.4 eV, which would correspond to an optimum value). Poor hole extraction, high series resistance, and hole pile-up lead to lower FF at 3.2eV, while efficient hole extraction, minimal recombination, lead to maximum FF at electron affinity of 3.4eV. At 3.5eV, due to the onset of electron leakage and interfacial recombination, FF decreases, while it increases further due to the blocking of electrons and electron leakage.

A suitable electron affinity ensures effective charge separation and efficient transport, thereby maximizing Voc and leading to a higher PCE at 3.4 eV. Discrepancies in electron affinities may result in suboptimal values of Voc, Jsc, and FF, thereby diminishing power conversion efficiency (PCE). To maximize charge transport in polymer solar cells, the electron affinity of the HTL can be adjusted to ensure suitable energy-level alignment with both the active layer and the anode.

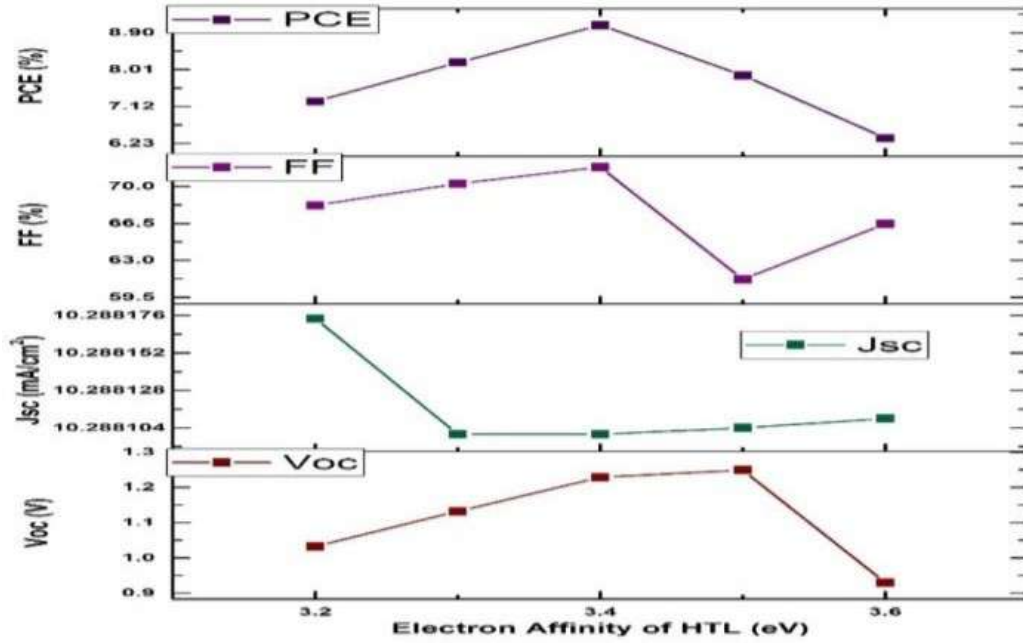


Figure 4.8: Influence of electron affinity

4.2.2.4 Influence of Effective density of states [CB]

The effective density of states of the conduction band of HTL [N_c] is changed from 10^{14} cm^{-3} to 10^{18} cm^{-3} . This primarily governs the HTL's ability to block electrons. The optimized performance is obtained at 10^{14} cm^{-3} . The corresponding graph is given in Fig 4.9.

In this case, Voc and PCE decrease as the density of states increases. Jsc increases, FF decreases, then increases again. A higher N_c in the HTL conduction band increases the likelihood that electrons from the active layer are thermally excited into the HTL conduction band. This probability leads to increased electron leakage (or back-diffusion) from the active layer to the anode, which results in an enhanced dark current and, in turn, a lower Voc.

Increasing the effective density of states in the HTL's conduction band enhances its ability to act as an effective electron-blocking layer. Raising the energy barrier for electrons reduces leakage current to the anode, ensuring that more of the photogenerated electrons contribute to the external circuit and thereby increasing J_{sc}.

The fill factor is therefore susceptible to losses due to recombination and series resistance. The increased electron leakage from the higher N_c results in significant recombination losses at the HTL/active layer interface. This poor charge transport and increased loss mechanism directly cause the fill factor to drop significantly. The Power Conversion Efficiency (PCE) decreases due to the combined effect of the three parameters V_{oc}, J_{sc} and FF [118, 119].

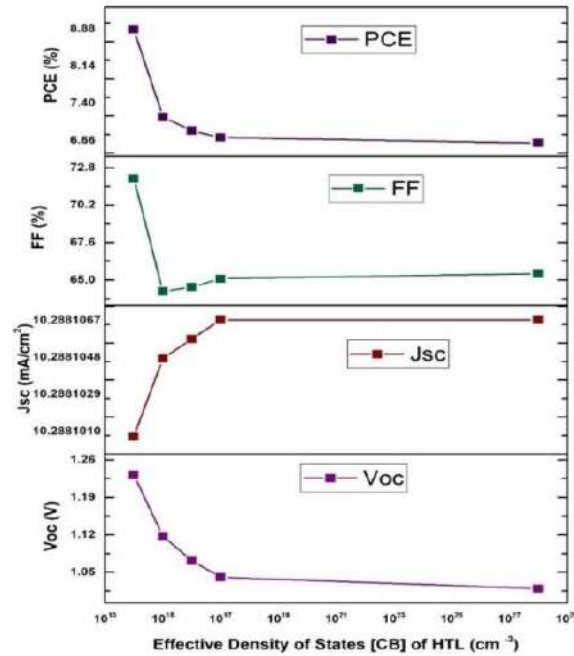


Figure 4.9: Influence of effective density of states [CB]

4.2.2.5 Influence of effective density of states [VB]

The effective density of states of the valence band of HTL is changed from 10^{16}cm^{-3} to 10^{20}cm^{-3} . Here, the Voc, FF, and PCE decrease as the density of states in the valence band of HTL increases. Jsc is found to increase with the density of states in the valence band of HTL. The optimized results are obtained at 10^{16}cm^{-3} . The results are shown in Fig. 4.10.

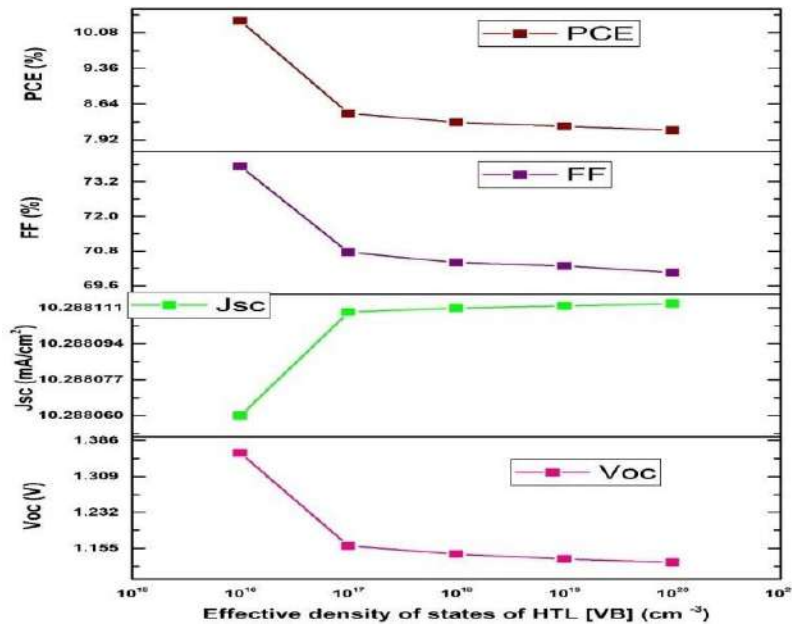


Figure 4.10: Influence of effective density of states [VB]

The effective density of states in the Valence Band (N_v) of the Hole Transport Layer (HTL) is a crucial parameter that directly influences the HTL's primary function: to efficiently transport holes from the active layer to the anode. In many practical solar cell systems, an excessively high doping concentration (which correlates with a high N_v) in the transport layer can lead to a significant increase in interfacial recombination

at the HTL/Active Layer boundary. Jsc increases due to enhanced charge transport and effective charge extraction FF decreases as a result of recombination. The decline in PCE is due to the combined effect of other output parameters [120, 121].

4.2.2.6 Influence of acceptor density of states

The acceptor density of states of HTL is varied from $10^{14}cm^{-3}$ to $10^{18}cm^{-3}$. In this case, it is found that Voc increases with increasing HTL acceptor density, whereas FF and PCE decrease with increasing HTL acceptor density. An acceptor density of $10^{14}cm^{-3}$ yields the best results. The graphs are displayed in Fig. 4.11. The Voc is enhanced by improving the built-in potential and reducing recombination losses. The Jsc is increased with efficient hole transport and minimal recombination. The FF is reduced due to increased series resistance and recombination. The PCE is maximized when Voc, Jsc, and FF are optimized through balanced HTL acceptor density. To maximize the performance of polymer solar cells, the HTL acceptor density should be optimized. This involves finding a balance where the HTL is doped enough to improve hole mobility and transport while minimizing recombination and resistive losses. Achieving this balance ensures that the solar cell operates efficiently with high Voc, Jsc, FF, and PCE.

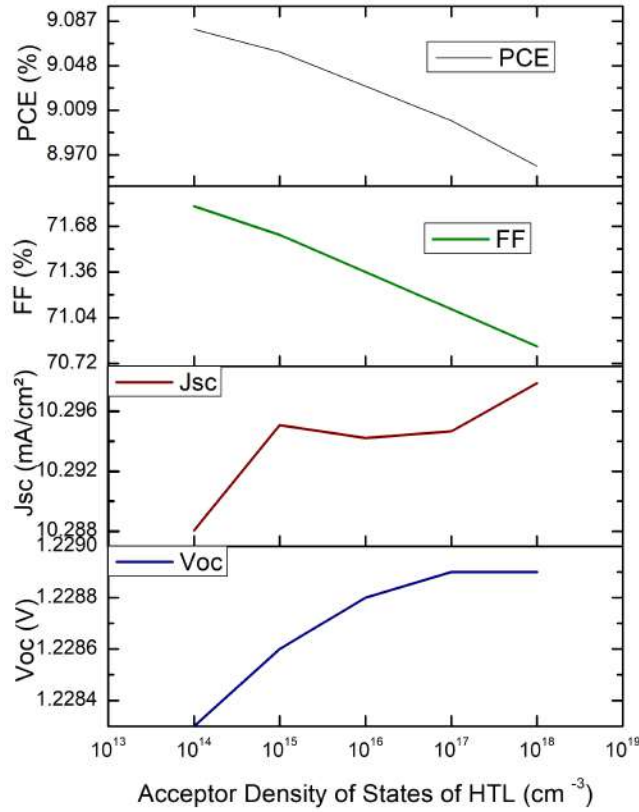


Figure 4.11: Influence of acceptor density of states

4.2.2.7 Influence of defect density

The total defect density of HTL is changed from 10^{14} cm^{-3} to 10^{18} cm^{-3} . Here, the Voc is found to increase, while the other output parameters are found to decrease as the total defect density of HTL increases. The optimized performance is obtained at a defect density of 10^{14} cm^{-3} . The corresponding graph is given in Fig 4.12.

In polymer solar cells, a defect in the HTL can act as a recombination center where charge carriers (electrons and holes) can recombine. When the defect density of

such defects increases, it can lead to effectively trapping the electrons (or holes) that have diffused into the HTL. This effective trapping can lead to a situation where the photogenerated charge carriers are trapped in the HTL and are less likely to recombine at the HTL interface. This results in a lesser overall rate of recombination of charge carriers for a given charge separation in the cell, i.e. a larger charge separation leads to an increased quasi-Fermi level splitting of the photon-to-current conversion process in the cell leading to an increase in V_{oc} .

Thus, a high defect density can trap our charge carriers, reducing their normal mobility and increasing the effective recombination rates, thereby lowering the J_{sc} . High defect density can increase the series resistance due to impeded charge transport and increase the shunt through providing more shunt paths in this way (also increases recombination) thereby also reducing the FF. Lower defect density can ensure efficient charge transport and collection (its actually preferential as far as charge carriers), minimal recombination, and optimal series and shunt resistances, maximising the V_{oc} , J_{sc} , and FF, and therefore the PCE. Higher defect density has the opposite effect, reducing these and in turn lowering the PCE. Careful material selection, processing conditions and interface engineering are adopted to reduce the defect density.

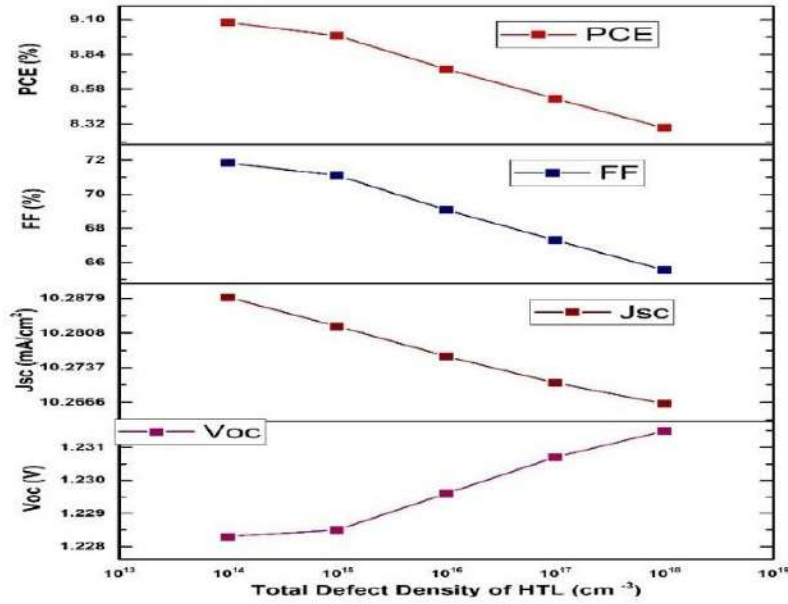


Figure 4.12: Influence of defect density

4.3 Effect of temperature

The temperature is changed from 300 K to 360 K. The optimal performance is achieved at 300 K. The variations of output parameters are shown in Fig 4.13.

At higher temperatures, the materials' carrier concentrations, mobility of charge carriers, resistance, and band gap change. As the temperature rises, the intrinsic carrier concentration in the semiconductor increases, reducing the separation of the quasi-Fermi levels. This results in a decrease in Voc. Additionally, higher temperatures can increase charge-carrier recombination rates, further lowering Voc. Jsc increases with increasing temperature due to enhanced mobility of charge carriers and a slight increase in photon absorption.

Elevated temperatures increase the series resistance and decrease the shunt resis-

tance in the solar cell, leading to a reduction in FF. The diode ideality factor also increases with temperature, negatively impacting the FF. Since PCE is a product of V_{oc} , J_{sc} , and FF, the overall efficiency decreases with increasing temperature. Maintaining the efficiency of polymer solar cells requires proper thermal management, including selecting materials with improved thermal stability to mitigate the negative effects of temperature rise, using heat sinks, and optimizing cell design. In polymers, thermally generated carriers increase with temperature, enhancing the overall carrier concentration.

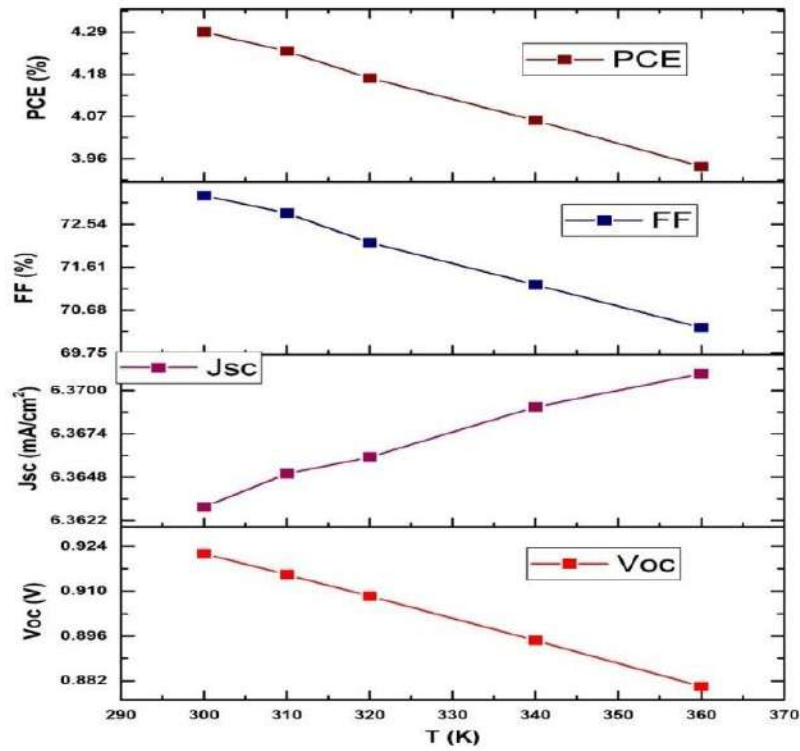


Figure 4.13: Influence of temperature

4.4 Conclusion

Here we explored how the performance of the ETL-free solar cell - depends on the parameters of the HTL as well as the properties of the active layer (its thickness, carrier concentration, defect density, etc.). The results of an optimization process, where these key parameters were fine-tuned, gave rise to a device with fairly improved performance. The ultimate parameters obtained were $V_{oc}=1.416$ V, $J_{sc}=10.29$ mA/cm², fill factor of 74.30%, and power conversion efficiency of a PCE of 10.83%. The results are encouraging and may open up future research in this field.

Chapter 5

Simulation studies of P3HT:IC₆₀BA Organic Solar Cells with additional P3HT Layer

In the last chapter, we performed a suite of simulations investigating and optimizing the performance of a bulk heterojunction solar cell with an active layer of P3HT:IC₆₀BA - importantly, with no electron transport layer in place. The study involved a detailed characterization of the device to determine its performance limits. Furthermore, the influence of several performance-affecting variables—specifically absorber thickness, doping concentration, and defect densities—was thoroughly investigated. The results inform the study of a differentiated cell in Chapter 5. With these understandings in hand, an additional change in device architecture was considered which should lead to better charge extraction and overall efficiency. In this chapter, an additional P3HT layer is added between the active layer and the anode. This layer of P3HT will act as an acceptor for holes and an insulator for electrons, thereby reducing the possibility of recombination and improving the device quality, as measured by open-circuit voltage (Voc), short-circuit current density (Jsc), and fill factor (FF).

It is with these new simulations that the effect of this P3HT layer on the solar cell performance, quantifying the improvements obtained, and elucidating the physical reasons for the effect, is studied.

5.1 Introduction

The performance of a polymer solar cell with P3HT: IC₆₀BA as the active layer is improved when P3HT is used as an extra layer. Based on previous studies, the operational performance and efficiency of the device were improved to 26.5% by introducing an ultrathin P3HT polymeric layer as an interfacial medium between the hole transport and perovskite layer [122]. To effectively separate and collect charges P3HT promotes the mobility of the electrons and holes which adds to improved charge transport qualities. It helps to maximize the open circuit voltage and current density through maximizing energy level alignment at the heterojunction interface [123, 124].

P3HT is capable of enhancing the structural ordering and crystallinity of the active layer, leading to larger crystal domain sizes that reinforce the interpenetrating network necessary for efficient charge transfer [10]. Furthermore, P3HT's solubility in general solvents allows the use of simple solution-processing steps, which enhances the scalability of the layers along with compatibility with a wide range of production methods [125]. All these features contribute to a noticeable increase in the power conversion efficiency of the solar cells. To the best of our knowledge, this is the first simulation study reporting the effect of using P3HT on Polymer solar cell with P3HT:IC₆₀BA as the active layer. We have performed this simulation study for the

structure by investigating the effect of P3HT layer being added. The simulated solar structure is given in Fig 5.1 .

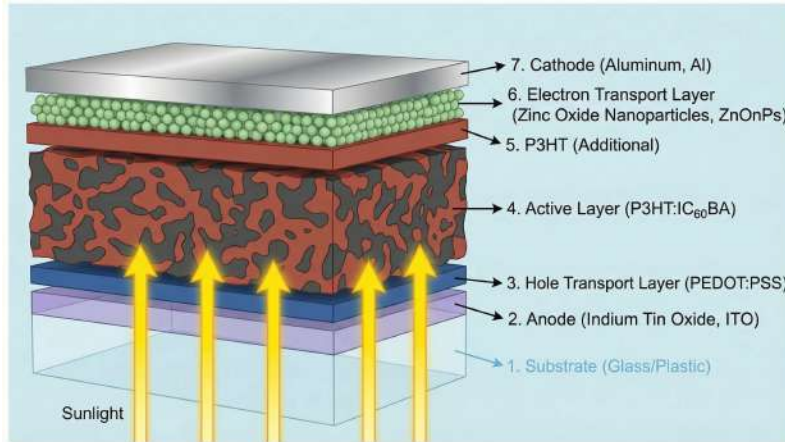


Figure 5.1: Simulated Polymer Solar Cell Structure

5.2 Optimization of the cell structure ITO/PEDOT:PSS/P3HT:IC₆₀BA/P3HT/ ZnONPs/ Al

Optimization of the solar cell structure shown in Fig. 5.1 is performed by varying the parameters of the different layers.

5.2.1 Effect of Active Layer Properties

5.2.1.1 Influence of thickness

The thickness of the active layer is varied between 100 and 300 nm in order to see the characteristics related to solar cell performance. The parameters Voc, Jsc, FF and PCE show a similar trend where all are maximized at a thickness of 300 nm. It makes sense that all these parameters improve at the ideal thickness because it is

where recombination is less and light is absorbed better. Increasing the thickness of the active layer can increase the J_{sc} a little by absorbing more light. However, it is not a linear relationship; after the ideal thickness, additional thickness can lead to a smaller increase due to increased charge recombination and decreased charge transport [126, 127].

V_{oc} is primarily not dictated by the thickness of the active layer. The factors that dictate it are the energy levels of the donor and acceptor. Recombination dynamics and quality of the interface may play an important role in its behaviour [128, 129]. Some experiments showed that increasing the thickness of the active layer improves the power conversion efficiency (PCE) to a certain extent; the relevant graph is present in Fig 5.2.

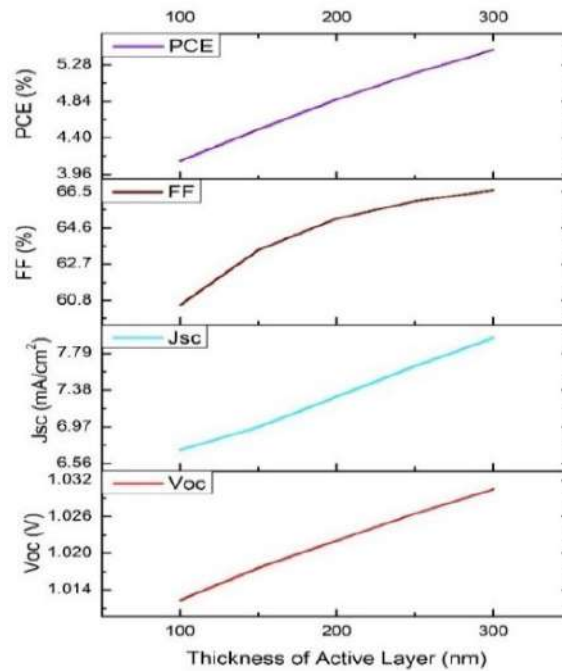


Figure 5.2: Influence of Thickness

5.2.1.2 Influence of Band Gap

The band gap of the active layer is varied from 1.5eV to 1.7eV, and both power conversion efficiency (PCE) and open-circuit voltage (Voc) are varied similarly with band gap. Both PCE and Voc are maximized at 1.7eV. In organic solar cells, Voc is set by the difference between LUMO and HOMO. As the band gap of the active layer increases, the Voc generally increases, suggesting a higher LUMO of the donor or a lower HOMO of the acceptor. This relates to the energy levels of the donor and acceptor, and to how Voc corresponds to the difference between the quasi-Fermi levels of electrons and holes. A larger band gap means a smaller energy loss between E_g and Voc, although in organic solar cells, the Voc is usually less than the band gap because of non-radiative recombination and other losses.

The open-circuit voltage (Voc) increases with the band gap and can thus increase the power-conversion efficiency (PCE), since the wider the band gap, the less of the solar spectrum is incident on the solar cell. Of course, this is primarily in the infrared light for the most part and so the solar current may go down, and in some cases, an increase in Voc may be enough to bring the PCE somewhat up. The relationship of FF with increasing band gap is due to the improved charge transfer [130]. Generally, with a wider band gap, the FF may increase and certainly may remain about constant according to competitive effects due to the recombination processes and exciton lifetimes of the material used [131, 132]. The relevant graph illustrating these dynamics is presented in Fig 5.3.

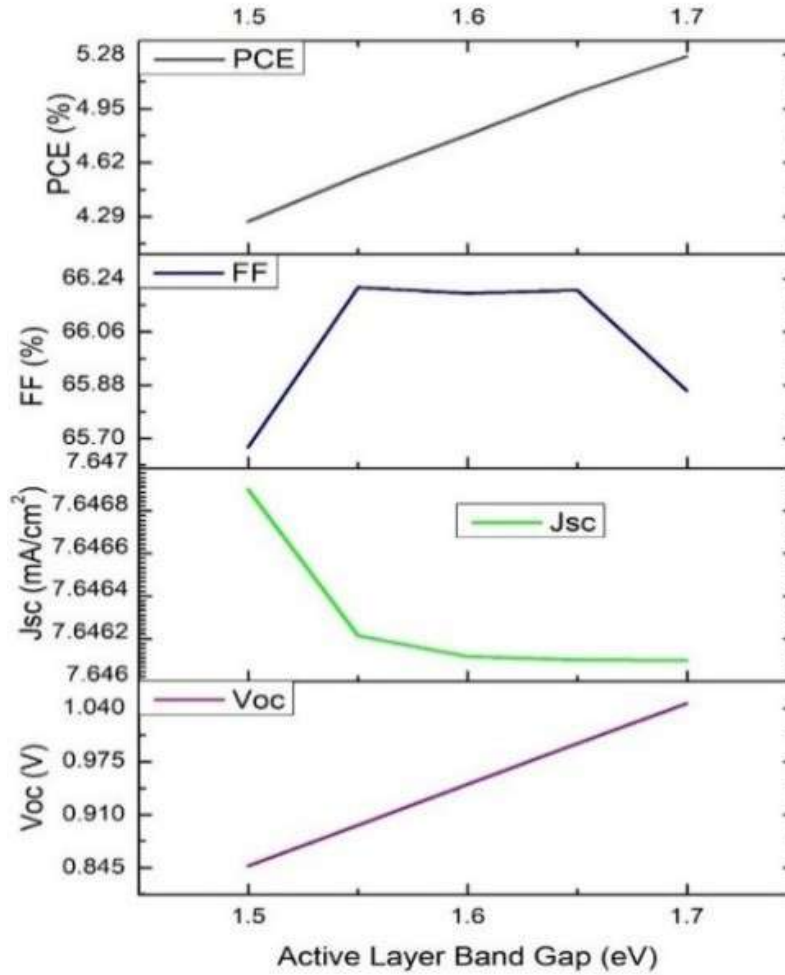


Figure 5.3: Influence of Bandgap

5.2.1.3 Influence of Electric Permittivity

In this section, the effect of electric permittivity ϵ of the active layer on the performance of the polymer solar cell is studied. The electric permittivity of the active layer is varied from 3.0 to 3.4. We see that while the open-circuit voltage V_{oc} increases with ϵ , all the other parameters drop. The electric permittivity of the active layer

is important because it may affect the dielectric properties of the material, which subsequently leads to influence the open-circuit voltage of polymer solar cells (PSCs). This means that a higher electric permittivity of the active layer would lead to an improved Voc due to the dynamics of charges in the solar cell.

A higher electric permittivity is usually observed to improve charge-carrier separation and reduce recombination losses by reducing Coulombic interactions [133]. This goes to improve the built-in electric field for stronger charge extraction, and therefore better response, leading to better Voc. Although just having a higher permittivity would not in itself improve the performance all around, i.e., make better PSC, all contributing parameters have to be well understood. Improving the electric permittivity of the active layer enhances the dissociation of radiatively generated electron-hole pairs and promotes charge-carrier transport, thereby affecting Jsc, FF, and PCE. The elevation of the coulombic attraction between the opposite charges arises sometimes because of high electric permittivity; it is better that they not be so inseparable, because this would more or less automatically constitute a reduction in Jsc [134]. This higher permittivity may also affect charge-carrier mobility, thereby enhancing recombination and likely lowering both FF and PCE. The graph representing this is shown in Fig 5.4.

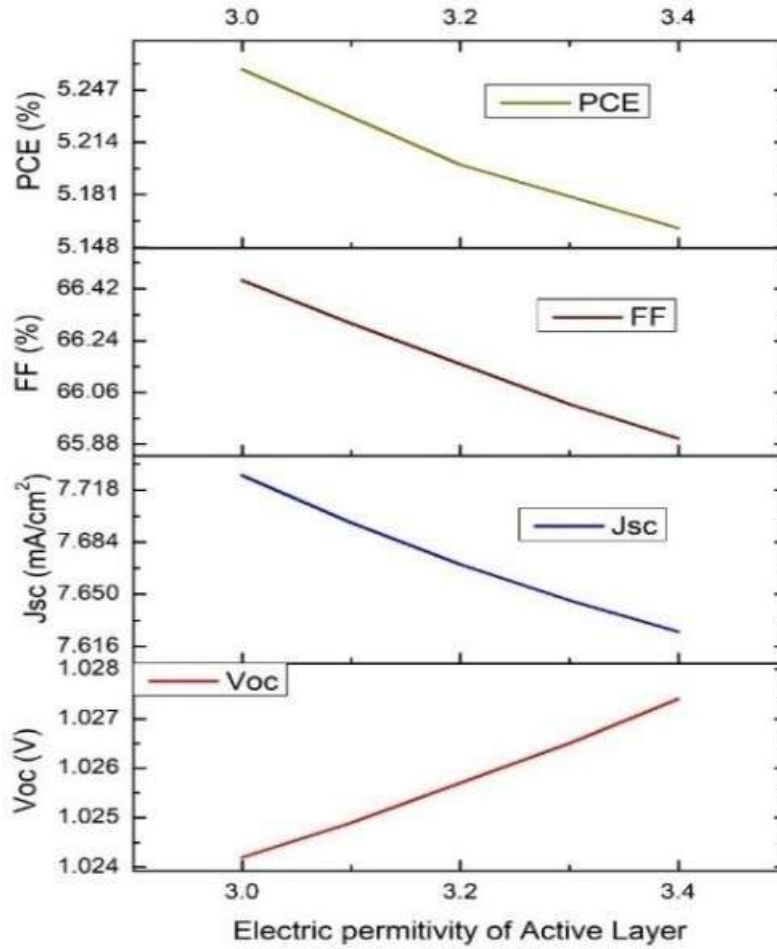


Figure 5.4: Influence of Electric Permittivity

5.2.1.4 Influence of Conduction Band Density of States

In this section, the effect of varying the conduction band density of states (DOS) in the active layer from 1.6×10^{19} to $1.6 \times 10^{21} \text{ cm}^{-3}$ on the metrics of polymer solar cells is considered. The resulting data for charge carriers are presented in Fig. 5.5. It is observed that the short-circuit current (Jsc) and power conversion efficiency (PCE) vary in a similar pattern, whereas the open-circuit voltage (Voc) and fill factor (FF)

vary differently.

The falling V_{oc} , with an increasing conduction-band density of states, can be attributed to increased recombination losses and shifts in the Fermi level. A recombination event is more likely to occur if the density of states is higher, as this provides more energy levels for charge carriers to occupy [135].

The increase in short-circuit current density (J_{sc}) for a higher conduction band density of states (DOS) is due to greater overall charge carrier generation and improved collection efficiency. A larger DOS increases the number of available conduction states, leading to more efficient photon absorption and thus a greater production of electron-hole pairs. This, in turn, leads directly to an increase in J_{sc} [136].

However, the concurrent rise in power conversion efficiency and fall in the fill factor with an increasing conduction band DOS is a result of competing effects: increased charge generation and increased recombination losses. While the absorption and generation of charge carriers increase with DOS, so do the opportunities for recombination, which decreases the FF [135].

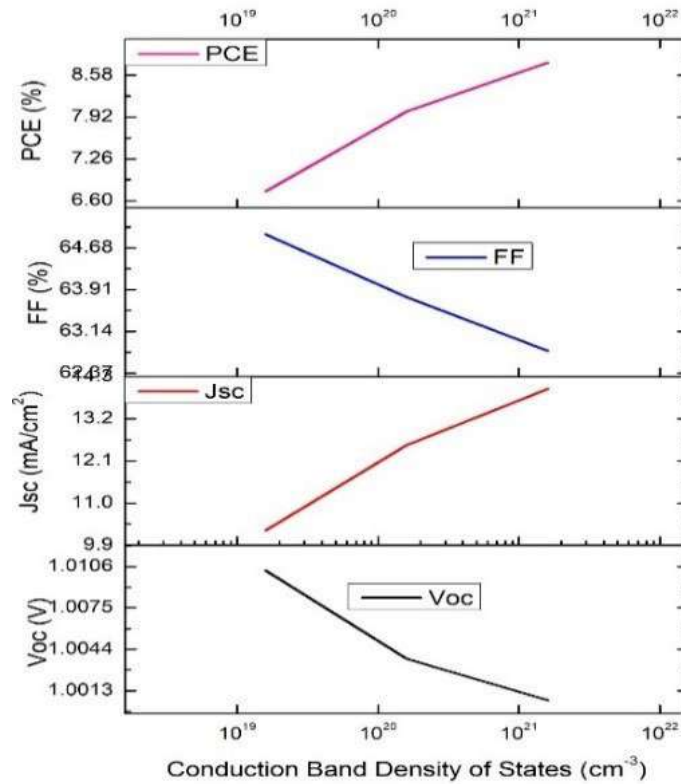


Figure 5.5: Influence of CB density of states

5.2.1.5 Influence of Valence Band Density of States

The valence band density of states in the active layer is changed in the range of 1×10^{16} to $1 \times 10^{20} \text{ cm}^{-3}$, and the influence on the characteristics of the polymer solar cells is investigated. It is shown that while Jsc and FF changed in almost the same fashion, Voc and PCE did not display the same trend. PCE and Voc decline with increasing valence band DOS mainly due to the more place for the recombination and increased trapping of charge carriers. Valence band DOS is very easy to increase so that more energy states to which the charge carriers can recombine non-radiatively

will be made. The more of these processes that happen, the worse the performances (PCE and Voc) will be [137, 138]. The respective graph of is given in Fig 5.6.

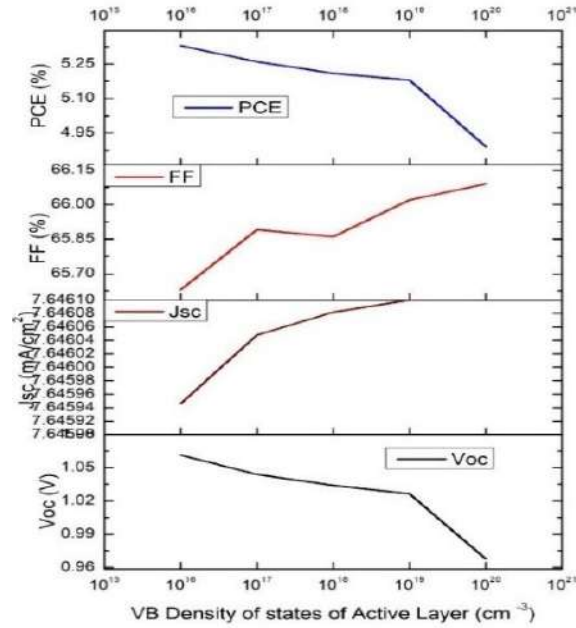


Figure 5.6: Influence of VB density of states

5.2.1.6 Influence of Uniform Donor Density of States

The uniform donor density of states in the active layer is varied from 3.2×10^{14} to $3.2 \times 10^{16} \text{ cm}^{-3}$. Higher Voc, FF, and PCE are found to increase by increasing this density, while Jsc is found to decrease [139]. Higher donor density means a higher intrinsic potential across the junction, enhancing the Voc, via the availability of more charge carriers, reducing the loss associated with charge separation in an ideal solar cell [138]. Higher donor density also means more charge carriers in the active layer, improving its conductivity, resulting in lower series resistance. Lower series resistance may improve the fill factor of the solar cells, also giving a minimal voltage drop on

load [139]. The graph is given in Fig 5.7 [140].

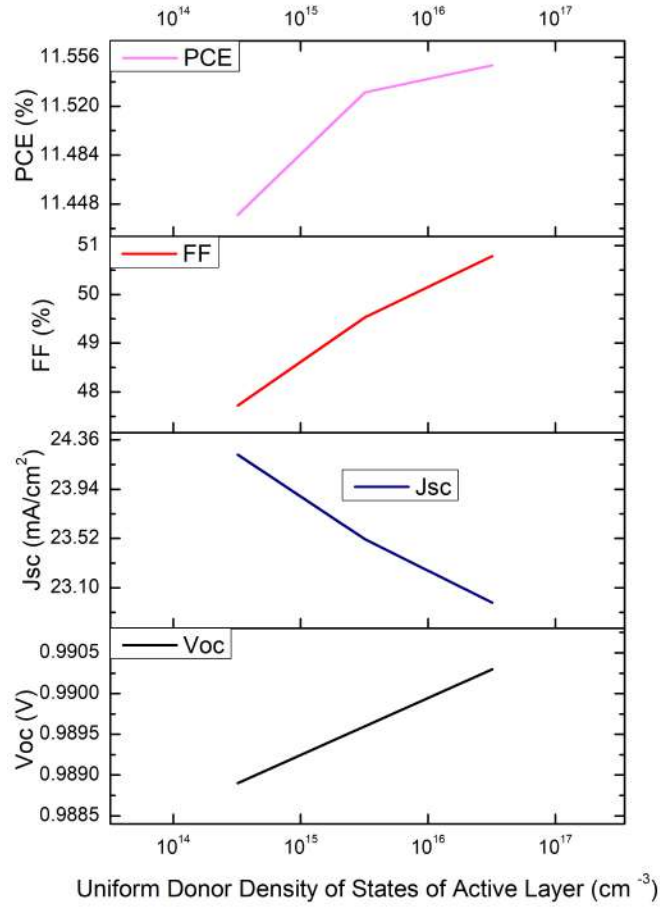


Figure 5.7: Influence of Uniform Donor Density of states

5.2.2 Effect of ETL Properties

5.2.2.1 Influence of Thickness

This study investigates the effects of changing the thickness of the electron transport layer (ETL) made from zinc oxide nanoparticles (ZnONPs), from 40nm to 48nm. The

best performance by changing the thickness was found at 48nm where the values of PCE peaked at 5.22%; Voc of 1.0270 V, and Jsc as 7.699979mA/cm². That at 40nm thickness, FF was discovered to have higher values as indicated by 66.09%.

An interpretation of output parameters shows that an increase in thickness results in a decrease in FF, while Voc, Jsc and the PCE increased. With a thicker ETL, it becomes easy to mitigate the rate of charge recombination in polymer solar cells by slowing down the process by way of increasing the open circuit voltage (Voc). A thicker electron transport layer increases the distance the electron has to travel before reaching the electrode (also known as the transit space of the electron). This reduces the chances for electrons and holes to recombine before they can reach the electrodes.

The thick electron layer makes it easy for photo-generated electrons to be conveniently collected by the electrode since more electrons arrive at the electrode with a minimum of them being trapped or recombined hence, this improvement made in the region of charge space causes more charge carriers to readily reach the electrode, resulting in a higher Jsc [141]. Polymer solar cells have been known to report lesser values for the FF when associated with greater thicknesses of the ETL because of higher series resistance, lower carrier mobility, higher recombination, band, and interface charge transfer resistance [142]. Also, the power conversion efficiency (PCE) of the polymer can duly improve with increasing thickness of ETL, and up to a certain limit, due to better light absorption, better charge extraction and the charge transport ability associated with the applied thickness [143]. The respective graph is as shown in Fig. 5.8.

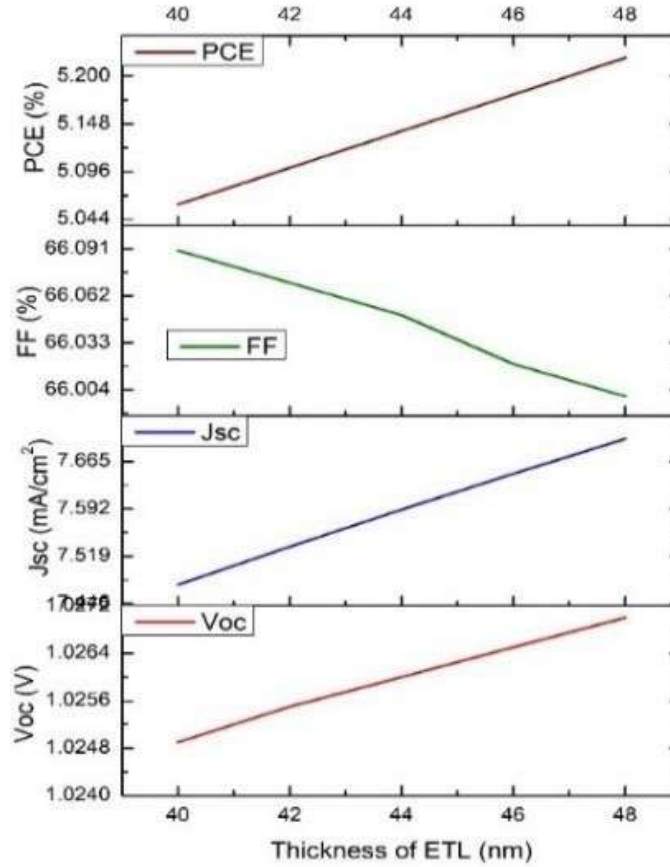


Figure 5.8: Influence of Thickness

5.2.2.2 Influence of Electric Permittivity

The effect of the electric permittivity of the ETL on polymer solar cells is shown in Fig. 5.9. It can be seen that with an increase in electric permittivity from 8.2 to 9.0, Voc decreases while both Jsc and efficiency increase. A value of 9.0 gives the best optimized case. To enable effective charge transfer, the energy levels of the ETL must be well-aligned to those of the polymer in the active layer; a too high permittivity of the ETL may disrupt the alignment at the ETL/polymer interface, leading to a

voltage loss and decreasing the Voc due to ineffective charge transfer.

Imperfections in the ETL may even lower the Voc and block charge transfer, notwithstanding a high permittivity. Continuous charge collection is dependent on the quality of the interaction between charge carriers and their layer. All directions affecting the electrostatic potential, especially exacerbated by high dielectric constant materials [144] lead to lower Voc. Higher electric permittivity allows faster charge transfer at the donor-acceptor interface, thus reducing non-geminate recombination losses. Eliminating these losses, more charge carriers can be collected from the cell, leading to enhanced performance. Improved charge dynamics and reduced recombination events lead to higher values of Jsc and PCE, and this can be a result of a better dielectric environment [145]. However, defects or poor interfaces may still induce some remix losses, thereby lowering the responsiveness of the fill factor.

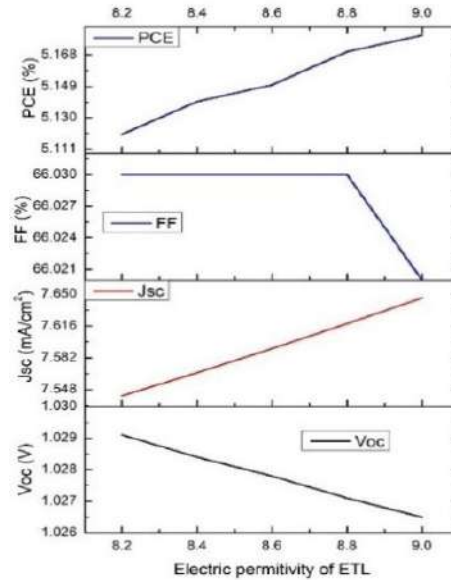


Figure 5.9: Influence of Electric Permittivity

5.2.2.3 Influence of Conduction Band Density of States

This section studies the effect of the conduction band density of states (CBDoS) of the electron transport layer (ETL) on the performance of the polymer solar cell. By varying the CBDoS from 2.2×10^{17} to $2.2 \times 10^{20} \text{ cm}^{-3}$, it was observed that the J_{sc} and PCE followed a similar downward trend, while the FF decreased and then increased. The maximum performance was achieved at a CBDoS of $2.2 \times 10^{17} \text{ cm}^{-3}$, as shown in Fig. 5.10.

The decline of J_{sc} and PCE with increasing CBDoS can be attributed to carrier trapping, higher recombination rates, and potentially other effects such as energy level misalignment [146]. With a greater CBDoS, additional trap states can hinder charge extraction and promote more effective carrier recombination, resulting in a lower V_{oc} .

The behavior of the fill factor (FF) is more complex. The non-linear relationship is due to the intricate interplay between energy level alignment, the presence of trap states, and carrier dynamics. Compensating effects from increased recombination and changes in carrier mobility contribute to this non-linear behavior of the FF.

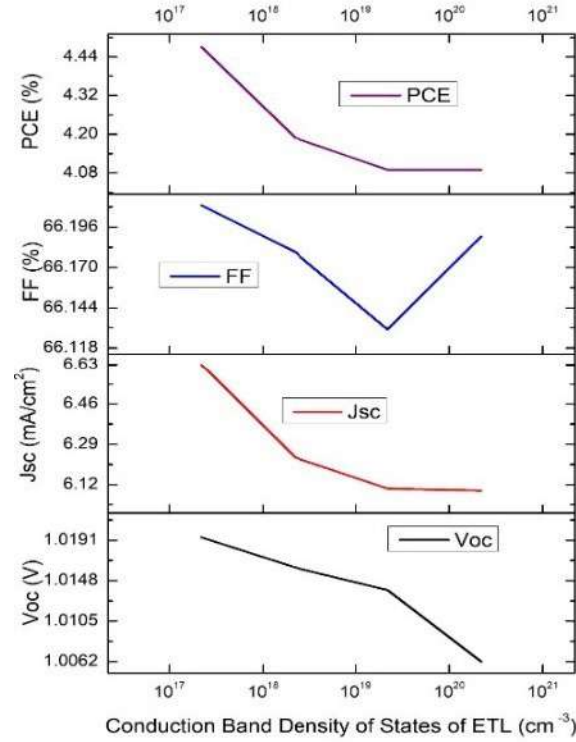


Figure 5.10: Influence of Conduction Band Density of States

5.2.2.4 Influence of Valence Band Density of States

This section considers changes to the valence band density of states of the electron transport layer in polymer solar cells, from 1.8×10^{17} to $1.8 \times 10^{21} \text{ cm}^{-3}$. It was also found that the optimal case, with maximum efficiency of 5.18% is achieved at a density of $1.8 \times 10^{17} \text{ cm}^{-3}$. Parameterized variation of the valence-band density of states in the electron-transport layer yields complex modifications to the output characteristics of polymer solar cells. Result metrics (J_{sc} , V_{oc} , FF and PCE) depend both on optimized density of states and how charge transport is balanced against recombination and interfacial energy levels. The respective graph is given in Fig.

5.11

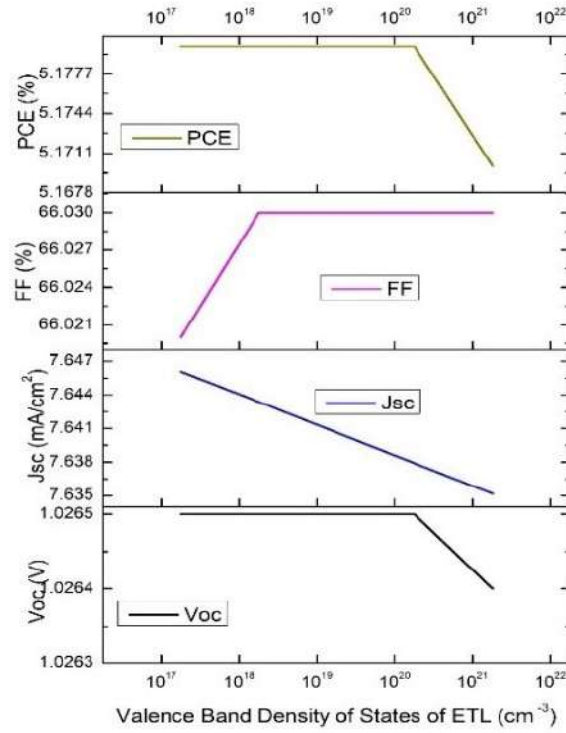


Figure 5.11: Influence of Valence Band Density of States

5.2.2.5 Influence of Donor Density

Donor density of the electron transport layer is varied from 1.1×10^{16} to $1.1 \times 10^{20} \text{ cm}^{-3}$, and the influence on output parameters is shown in Fig. 5.12. The results suggest that the fluctuations in Jsc and PCE are similar, while Voc and FF also vary similarly. Voc increases with donor density, as better charge extraction leads to less recombination. As charge transport and extraction improve, FF also increases with donor density. At high concentrations of donor density, recombination will be high, leading to lower PCE. Jsc varies due to increased recombination, charge carrier traps and reduced

depletion region.

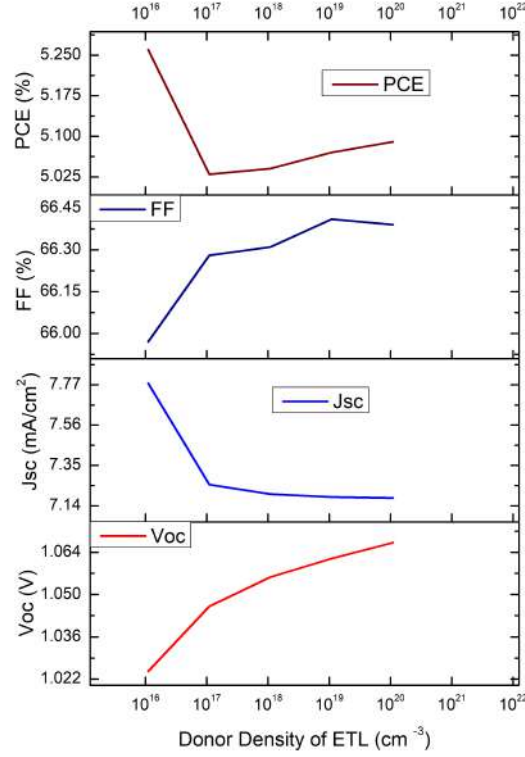


Figure 5.12: Influence of Donor Density

5.2.3 Effect of Additional P3HT Layer Properties

5.2.3.1 Influence of Thickness

In this case, the thickness of the P3HT layer is varied from 16nm to 24 nm, with the optical performance seen at 16 nm. As can be seen from the graph in Fig 5.13, in general, the FF shows a different trend, while Voc, Jsc, and PCE fluctuate similarly.

Thicker P3HT layers create longer and more complex paths for charges to travel. Charge mobility decreases, which is essential for the rapid transfer of electrons and

holes in thin films to the appropriate electrodes. This increase in the distance to cross means a greater opportunity for recombination before reaching the electrode, thereby dropping J_{sc} and PCE. The drop in V_{oc} may be due to less efficient charge extraction in thicker layers or perhaps poorer charge mobility and higher energy losses due to greater rates of recombination. The increase in thickness for P3HT layers used in polymer solar cells (PSCs) results in a corresponding increase in FF due to reduced bimolecular recombination, improved hole collection, reduced series resistance, and enhanced vertical phase separation [147].

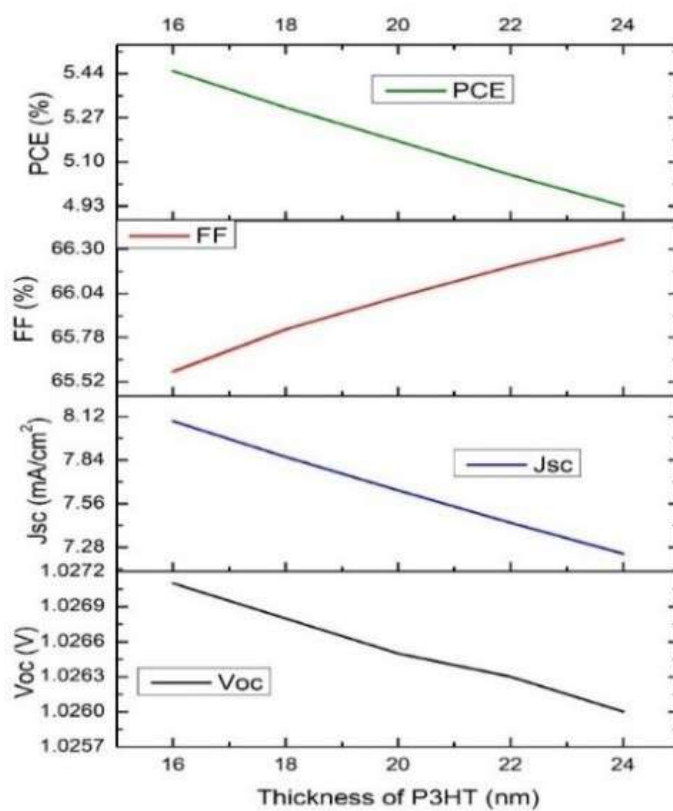


Figure 5.13: Influence of Thickness

5.2.3.2 Influence of Bandgap

The band gap of P3HT varies from 1.7 to 2.1 eV, and for all output parameters, it is observed that they all increase with the band gap as shown in Fig 5.14. There is a dependence of Voc on the LUMO and HOMO levels of the polymer. In general, a larger band gap will correlate with a higher HOMO energy level, which may increase Voc. A high band gap material will be a better absorber of high-energy photons, meaning that Jsc may be increased. Increasing the band gap increases the difference between the energy levels required for charge separation, making it less likely for excitons to recombine, making these improvements in FF and PCE.

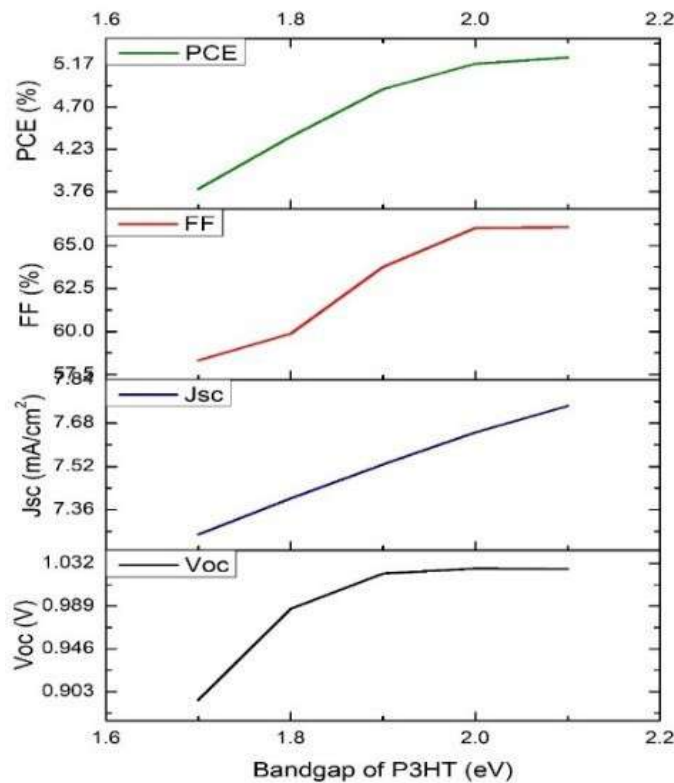


Figure 5.14: Influence of Band gap

5.2.3.3 Influence of Electron Affinity

This section examines the influence of the electron affinity of the electron-donating polymer P3HT on the performance of the cell, with the electron affinity varying between 2.9 and 3.3 eV. The output parameters of the cell exhibited a consistent response to these variations, as illustrated in Fig. 5.15. In general, a higher value of electron affinity will result in a higher LUMO energy level, which, when no current is applied, increases the open circuit voltage (V_{oc}). Higher electron affinities represent a greater energy offset and allowing the electrons to flow quickly away, possibly increasing J_{sc} , followed by reducing recombination loss to give an improvement to the FF and PCE with respect to the pure polymer donor [148].

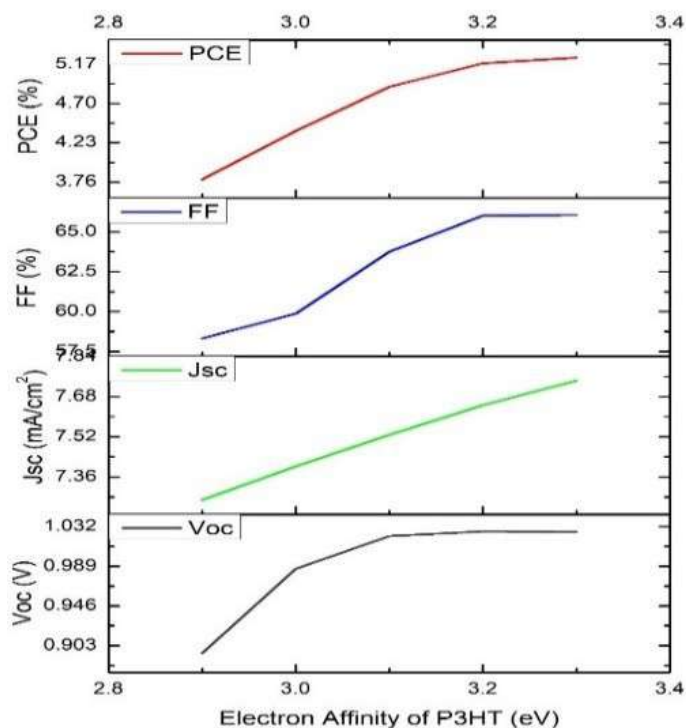


Figure 5.15: Influence of Electron Affinity

5.2.3.4 Influence of Valence Band Density of States

In this section, the effect of changing the valence band density of states of P3HT from 1.8×10^{16} to $1.8 \times 10^{20}, \text{cm}^{-3}$ on the output parameters of the polymer solar cells is discussed. Whereas the J_{sc} is different among the output parameters, V_{oc} , FF, and PCE follow similar trends with respect to varying material parameter values, as shown in Fig 5.16. Improvements in measures like PCE, FF, and V_{oc} arise from a more optimal energy level alignment and improved charge transport. However, as the valence band density increases, the performances begin to degrade due to higher defect densities and/or shorter diffusion lengths. The optimal high value is present at a valence band density of $1.8 \times 10^{17}, \text{cm}^{-3}$. The drop in the J_{sc} for the polymer solar cells, especially for the cells that incorporate P3HT, does indeed arise from increased defect density and shorter charge carrier diffusion lengths associated with a high density of states.

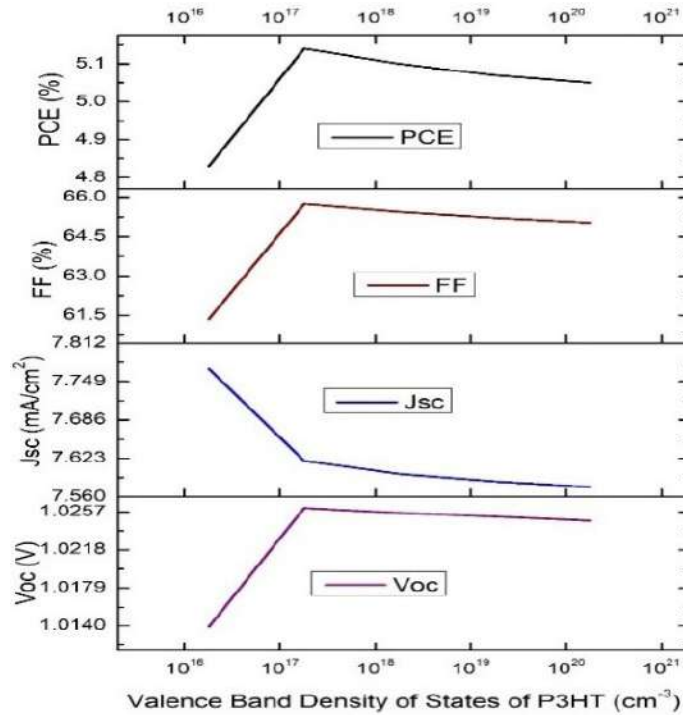


Figure 5.16: Influence of Valence Band Density of States

5.2.4 Effect of PEDOT:PSS [HTL] Properties

5.2.4.1 Influence of Band gap

This section deals with the effect on the output parameters of polymer solar cells of changing the band gap of the hole transport layer PEDOT:PSS from 1.2 to 2.0 eV. All parameters are seen to vary similarly. There is a maximum efficiency at 1.6 eV and the results are shown in Fig. 5.17. Voc is related to the energy level alignment between the donor's highest occupied molecular orbital (HOMO) and the acceptor's lowest unoccupied molecular orbital (LUMO). An 'ideal' bandgap for HTL promotes a better energy alignment at the anode interface, thereby extracting charge while

maximizing the ‘potential difference’ (voltage) across the device: If this bandgap is too low/high, an energy barrier/mismatch occurs, which reduces V_{oc} .

The role of the HTL is to extract holes efficiently from the active layer; an ‘optimal’ bandgap (1.6 eV in this case) allows for both efficient hole transport/collection while preventing electron leakage effectively, thereby reducing charge recombination at this interface, enabling more charge to be collected as current. Band gaps too low or high increase the difficulty of this extraction, leading to a relatively higher degree of recombination and thereby a lower J_{sc} . The fill factor is a measure of the quality of the cell and is extremely sensitive to series and shunt resistance.

The optimal bandgap at (1.6 eV) corresponds respectively to the lowest series resistance (good transport) and therefore the highest shunt resistance, at which there is the least ‘leakage’ for the current. This ideal balance translates to the minimum internal losses and thereby has the highest FF. The peak PCE is realised when all other parameters are simultaneously optimally adjusted. The 1.6 eV bandgap for PEDOT:PSS gives the best mix in voltage, current, and fill factor (i.e., optimized overall efficiency).

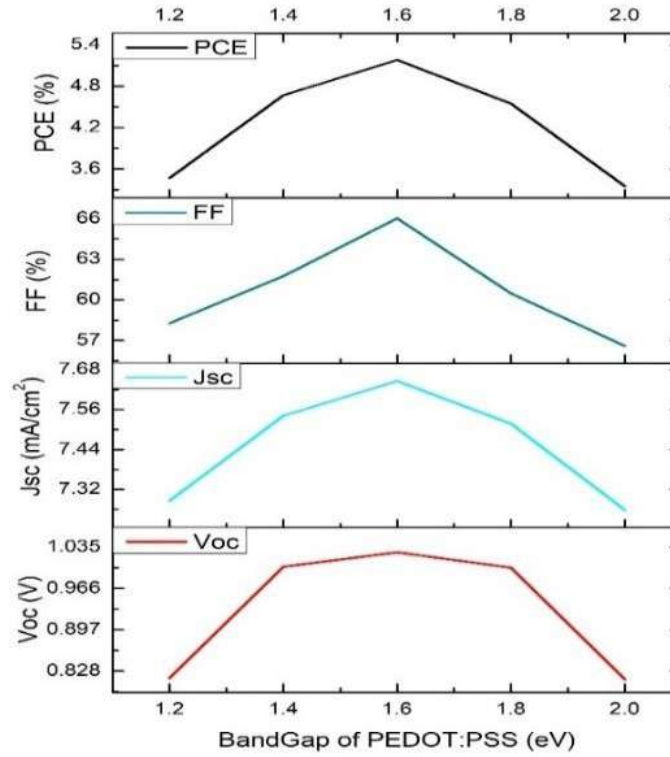


Figure 5.17: Influence of Bandgap

5.2.4.2 Influence of Electron Affinity

The electron affinity of PEDOT:PSS is varied from 3.0 eV to 3.8 eV, with optimal values typically around 3.4 eV, as shown in Fig 5.18. The output parameters of polymer solar cells are modified in a fashion similar to that described for the variation of band gap and electron affinity of PEDOT:PSS. This is due to the interdependency between the two processes of energy level alignment and charge transport, both of which are varied by these parameters.

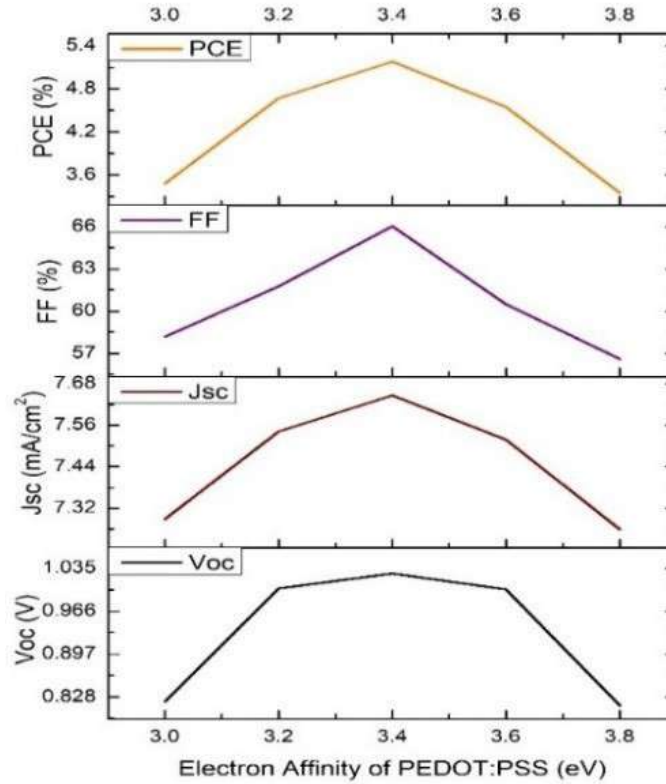


Figure 5.18: Influence of Electron Affinity

5.3 Conclusion

In this modelling study the introduction of an additional P3HT layer greatly improved device performance by enhancing charge transport and enabling optimal energy level alignment at the heterojunction interface, thus increasing the Voc and Jsc. Following optimization, the device achieved a power conversion efficiency (PCE) of 15.26%, a dramatic improvement from the initial 5.18%. The device achieved a final open-circuit voltage (Voc) of 1.113 V, a short-circuit current density (Jsc) of 26.58 mA/cm², and a fill factor (FF) of 51.62%.

Chapter 6

Simulation studies of P3HT:IC₆₀BA Organic Solar Cells using diverse HTL and ETL Layers

6.1 Introduction

Having considered the general architecture of the structured P3HT:IC₆₀BA OSC and the simulation tool, SCAPS-1D, through the previous chapters, we proceeded to the simulation studies in each chapter investigating the consequences of replacing the standard PEDOT:PSS HTL in Chapter 3 with CuI, and in Chapter 4, removing the ETL entirely. Modifications to the active layer through the addition of a layer of P3HT during the course of simulation studies are also reported in Chapter 5. All these studies have shown to the sensitivity of the photovoltaic performance parameters of the device; J_{sc}, V_{oc}, FF and PCE to variations in the composition and architecture of the internal layer.

While all the other chapters focused on targeted investigations of the reference device in ITO/PEDOT:PSS/P3HT:IC₆₀BA/ZnONPs/Al, overall optimization would require us to visit the material space of charge-transport layers. The PCE of an OSC

is dependent on the energetics around the interfaces and how selectively charge is withdrawn across these layers and transported through the HTL and ETL. These are crucial for avoiding additional recombination processes, effective charge extraction, and improving the PCE. In this chapter, we extend the simulation-guided studies on optimizing the OSC by exploring the effect of variations in Hole Transport Layers (HTLs) and Electron Transport Layers (ETLs) on the performance of the device.

The objective is to do a comprehensive optimization study through characterizing the pairing of two materials for HTL and ETL. It is hoped to provide a justification of the physics concerning interface engineering for OSCs, a highly efficient simulated device architecture in the final chapter. The materials selected here are chosen based on their reported electronic properties, band alignment with the P3HT:IC₆₀BA active layer, charge mobility, defect characteristics, etc. The simulation of the device performance across a matrix of HTL and ETL materials will allow us to:

- Quantify the individual and synergistic effect of various HTL and ETL materials on the output parameters J_{sc}, V_{oc}, FF, and PCE.
- Identification of the most promising material combinations that lead to effective “cascades” of energy levels and improved transport for the respective device.
- Providing guidelines for the selection of charge transport layers in P3HT:IC₆₀BA based OSCs, assisting suggesting experimental work in the future. The following sections should demonstrate the two chosen materials for the HTL and ETL, simulation parameters in SCAPS-1D, and performance of the OSC using a systematic comparison this time.

Several ETL and HTL materials are utilized to simulate the structure of the Organic solar cell . The most suitable and efficient BHJOSC design consists of a combination of cuprous oxide (Cu_2O) as the HTL and indium gallium zinc oxide (IGZO) as the ETL. The device is further optimized to analyze the effect of various material parameters on performance.

6.2 Standardization

The software is calibrated by comparing the results produced by an experimental paper [10]. Fig 6.1 illustrates the general structure used for simulations with various ETL and HTL materials. Fig 6.2 presents the J-V curves for both experimental and simulated results.

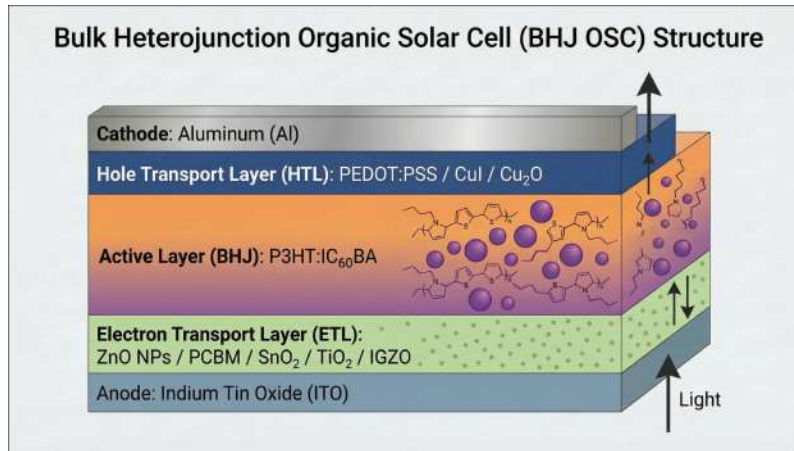


Figure 6.1: Generalised structure for simulations with various ETL and HTL

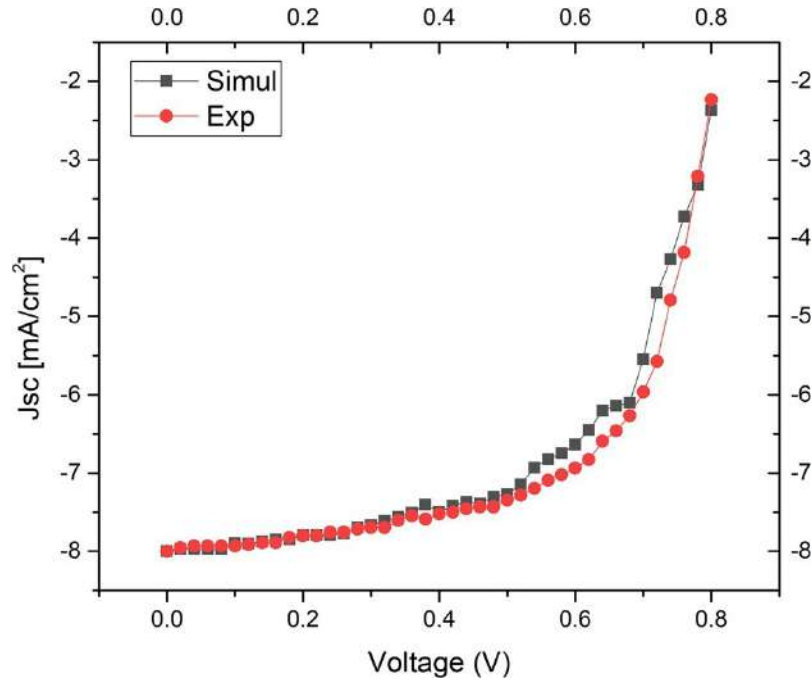


Figure 6.2: Comparison of experimental and simulated JV graphs

The anode and cathode work functions are set at 4.8 and 4.2 eV, respectively. The surface recombination velocity for electrons on the anode is set at 10^7 cm/s, whereas for holes it is fixed at 10^5 cm/s. In the cathode, electrons have a surface recombination velocity of 10^5 cm/s, and holes have a value of 10^7 cm/s. The thermal velocity is set at 10^7 cm/s, and the capture cross-section of both holes and electrons are set at 1×10^{-25} cm² across all layers. The primary output parameters of a solar cell are V_{oc} (open-circuit voltage), J_{sc} (short circuit current), FF (fill factor), and PCE (power conversion efficiency).

The solar cell structure is simulated with different HTL and ETL layers, and the most efficient combination is chosen. The thickness of HTL and ETL is maintained at 27 and 46 nm, respectively. The HTL layers considered are Poly(3,4-ethylene

dioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS), Copper Iodide (CuI), and Cuprous oxide (Cu_2O). The ETL layers considered are Zinc oxide nanoparticles (ZnO NPs), Phenyl-C61-butyric acid methyl ester (PCBM), Tin dioxide (SnO_2), Titanium dioxide (TiO_2), and Indium gallium zinc oxide (IGZO). The device parameters and absorption profiles of these layers were taken from the literature [149, 150, 151, 152]. Figure 6.3 depicts the energy level diagram for the various HTL and ETL layers under consideration.

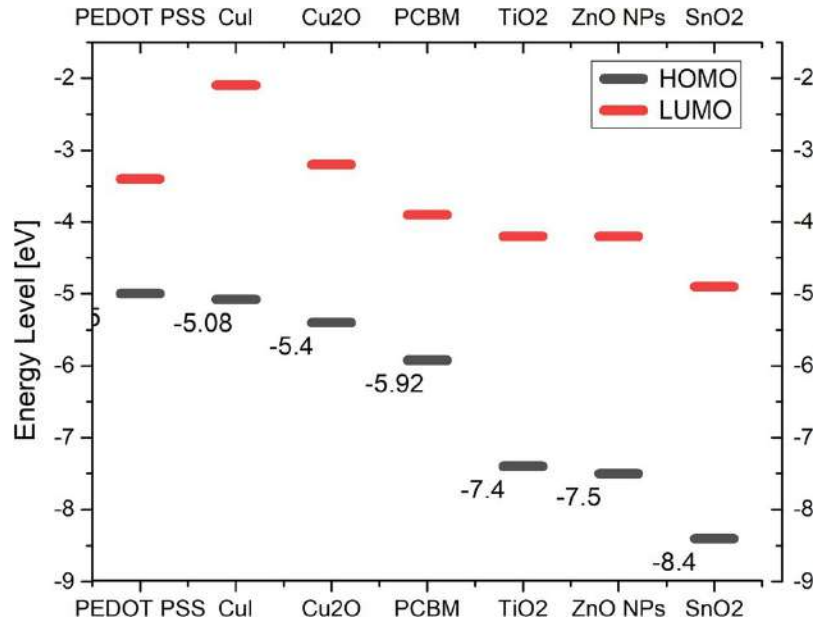


Figure 6.3: Energy level diagram of HTL and ETL layers

Table 6.1 shows the device parameters of HTL layers while Table 6.2 represents the device parameters of ETL layers used in the simulation study

Parameters	PEDOT:PSS	CuI	Cu_2O
Thickness [nm]	27	27	27
E_g [eV]	1.6	2.98	2.17
χ [eV]	3.4	2.1	3.2
ϵ_r	3.0	6.5	7.1
μ_n [cm^2/Vs]	1.0	1.69×10^{-4}	200
μ_p [cm^2/Vs]	4×10^2	1.69×10^{-4}	800
N_C [cm^{-3}]	2.2×10^{15}	10^{22}	2.5×10^{18}
N_V [cm^{-3}]	1.8×10^{19}	10^{22}	1.8×10^{19}
N_t [cm^{-3}]	10^{15}	10^{15}	10^{14}

Table 6.1: Device parameters of HTL layers

Parameters	ZnONPs	PCBM	SnO_2	TiO_2	IGZO
Thickness [nm]	46	46	46	46	46
E_g [eV]	3.2	1.9	3.6	3.2	3.05
χ [eV]	4.6	3.9	4.4	4.1	4.0
ϵ_r	9.0	4.0	9.0	9.0	9.0
μ_n [$cm^2/(V \cdot s)$]	2×10^2	0.5	240	2.0	100
μ_p [$cm^2/(V \cdot s)$]	5.0	1.5×10^{-2}	25	1.0	2.5
N_C [cm^{-3}]	2.2×10^{19}	1.8×10^{20}	4.36×10^{18}	2.2×10^{18}	10^{19}
N_V [cm^{-3}]	1.8×10^{19}	2.2×10^{21}	2.52×10^{19}	10^{19}	10^{19}
N_t [cm^{-3}]	10^{15}	10^{14}	10^{15}	10^{15}	10^{14}

Table 6.2: Device parameters of ETL layers

Table 6.3: Device Performance of OSC with different combinations of HTL and ETL

HTL	ETL	Voc [Volts]	Jsc [mA/cm^2]	FF %	PCE %
PEDOT:PSS	SnO_2	0.9596	9.30	59.98	5.35
	TiO_2	0.8750	9.50	63.48	5.27
	IGZO	0.9646	9.78	60.26	5.68
	ZnO NPs	0.8546	8.20	56.65	3.97
	PCBM	0.9798	12.46	59.58	7.27
CuI	SnO_2	0.8797	7.63	66.20	4.45
	TiO_2	0.6467	7.86	59.14	3.00
	IGZO	0.9104	12.79	62.06	7.22
	ZnO NPs	0.7586	6.47	61.72	3.03
	PCBM	0.6468	7.87	59.13	3.01
Cu_2O	SnO_2	1.3190	10.75	78.34	11.11
	TiO_2	1.3213	10.93	78.47	11.34
	IGZO	1.3221	11.26	78.20	11.64
	ZnONPs	1.1985	12.13	72.75	10.57
	PCBM	1.3261	11	78.41	11.41

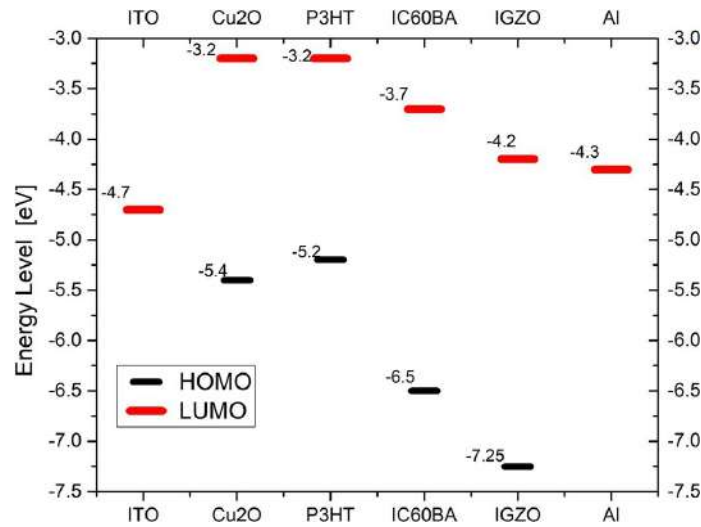


Figure 6.4: Energy level diagram of optimised organic solar cell structure

6.3 Simulation studies of P3HT:IC₆₀BA organic solar cell with different HTL and ETL Layers

Various HTL and ETL materials were employed in different combinations to simulate a bulk heterojunction organic solar cell involving P3HT:IC₆₀BA as the active layer. Each unique organic solar cell device combination was characterized for each respective HTL and ETL combination, with the characterization put together in Table 6.1 and 6.2, and the electrical output parameters are shown in Table 6.3.

Of the configurations, the highest performing organic solar cell was constructed with Cu_2O as the HTL and IGZO as the ETL, with a V_{oc} of 1.3221 V, J_{sc} of 11.26mA/ cm^2 , FF of 78.20%, and PCE of 11.64%. The performance is a function of the alignment of the band structures of the individual materials making up the organic solar cell and other electrical parameters specific to the device. Using Cu_2O as the HTL and TiO_2 as the ETL configuration yielded the highest FF (78.47%); the lowest FF (56.65%) was obtained with PEDOT: PSS as the HTL and ZnONPs as the ETL.

The power conversion efficiency varied from a low of 3% with CuI as the HTL and TiO_2 as the ETL, to a high of 11.64% with Cu_2O as the HTL and IGZO as the ETL. The energy-level diagram of the optimized organic solar cell structure is shown in Fig. 6.4. The highest performing organic solar structure, ITO/ Cu_2O /P3HT:IC₆₀BA/IGZO/Al was further studied to get optimized results.

6.4 Optimization of the cell structure ITO/Cu₂O/P3HT:IC₆₀BA/IGZO/Al

6.4.1 Effect of Active Layer Properties

6.4.1.1 Influence of thickness

As shown in Fig. 6.5, the active layer thickness is increased from 50nm to 300nm, and the effect on cell parameters is investigated. Jsc reaches a maximum value of 11.84 mA/ cm² at a thickness of 300 nm, whereas Voc reaches a maximum value of 1.3240 V at the same thickness. The FF reduces with thickness and peaks at 79.88% at 50 nm, while efficiency increases with thickness and peaks at 12.19% at 300 nm. These data show how the thickness of the active layer impacts light absorption and thus solar cell efficiency.

The difference between the HOMO level of the donor and the LUMO level of the acceptor determines Voc values. To promote exciton dissociation at the interface, a thick layer allows for increased electron and hole trapping in the film as well as considerable photon absorption, which results in a rise in Voc with thickness [153].

In this simulation, Jsc increases with active layer thickness because nearly all electron–hole pairs generated by photons are effectively collected at the electrode before recombination. This indicates a higher production of charge carriers. The fill factor (FF) diminishes in thicker films due to uneven charge extraction rates for holes and electrons [154]. This demonstrates that efficiency will increase because a thicker active layer allows for the formation of a continuous network throughout, producing the highest number of dissociated excitons and delaying charge recombination due to

coulombic attraction at the interface.

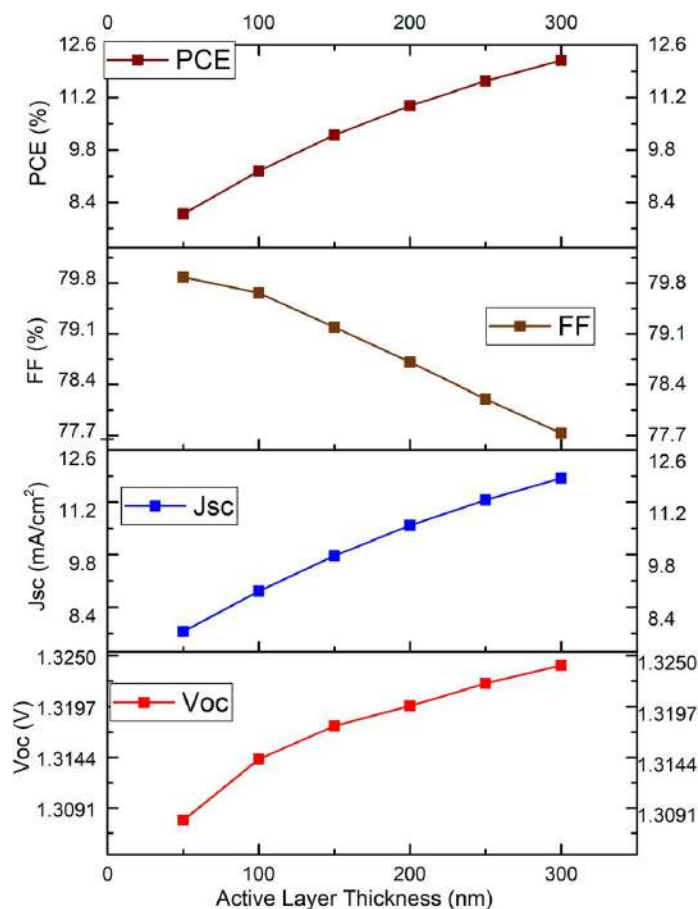


Figure 6.5: Influence of thickness

6.4.1.2 Influence of defect density

The relationship between the active layer defect density and the functioning of organic cells is shown in Fig. 6.6. The V_{oc} decreases with defect density, reaching a maximum value at 10^{15}cm^{-3} . The J_{sc} reaches its peak value of $9.003\text{mA}/\text{cm}^2$ at a defect density of 10^{20}cm^{-3} . While the PCE is seen to accompany defect density and reports a maximum value of 9.31% efficiency at a defect density of 10^{20}cm^{-3} , FF on the other

hand decreases with defect density of the active layer and attains a peak value of 79.66% at 10^{15}cm^{-3} . The consequence of defect concentrations in the absorber layer leads to faster film degradation, reduced stability, and poor device performance due to greater pinhole generation and recombination [155]. An increased density of defects within the absorber layer shortens charge carrier diffusion lengths and promotes carrier recombination, thereby acting as a key determinant of the device's overall efficiency [156].

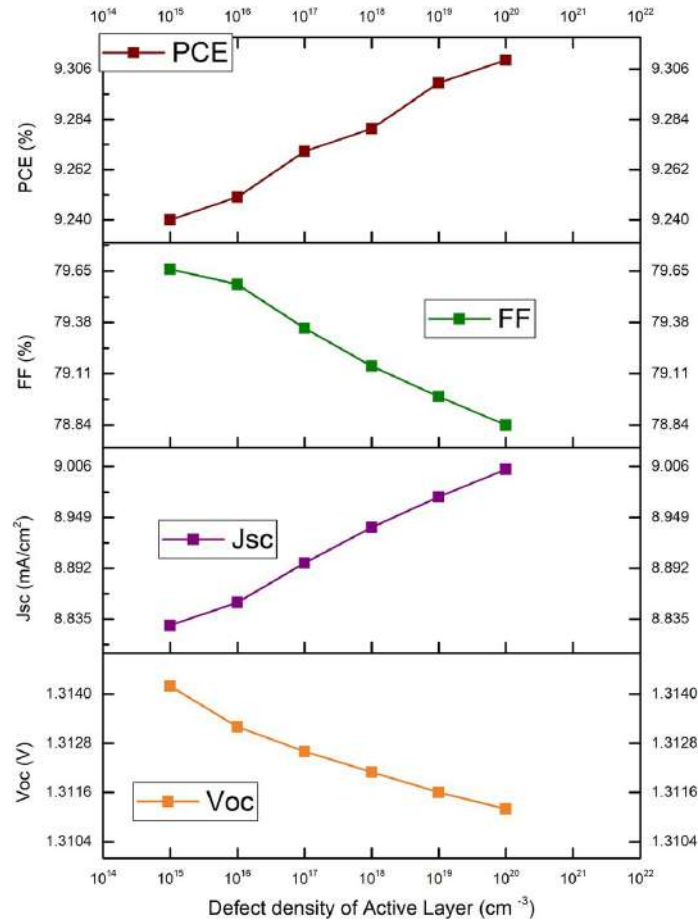


Figure 6.6: Influence of defect density

6.4.2 Effect of HTL Properties

6.4.2.1 Influence of electron affinity

The effect of the HTL electron affinity on cell performance is illustrated in Fig. 6.7. The electron affinity is associated with the lowest unoccupied molecular orbital (LUMO) of the hole transport layer that represents the energy (in eV) required to transfer a free electron from the lowest energy level of that orbital to the vacuum level. In the case of Voc, Jsc, and PCE they have a similar tendency, whereas FF behaves differently. It was found that the optimum Voc and Jsc values are 1.3673 V and 9.527 mA/cm² at 3.9 eV. At 3.5 eV, the optimum FF value is 80%, while at 3.9 eV the optimum PCE value is 10.42%.

Increasing the electron affinity of the HTL increases the cell performance as a consequence of improving the electron blocking property. This reduces recombination thus leading to a higher current (Jsc) and voltage (Voc), together raising overall efficiency (PCE), despite an almost negligible negative effect on fill factor.

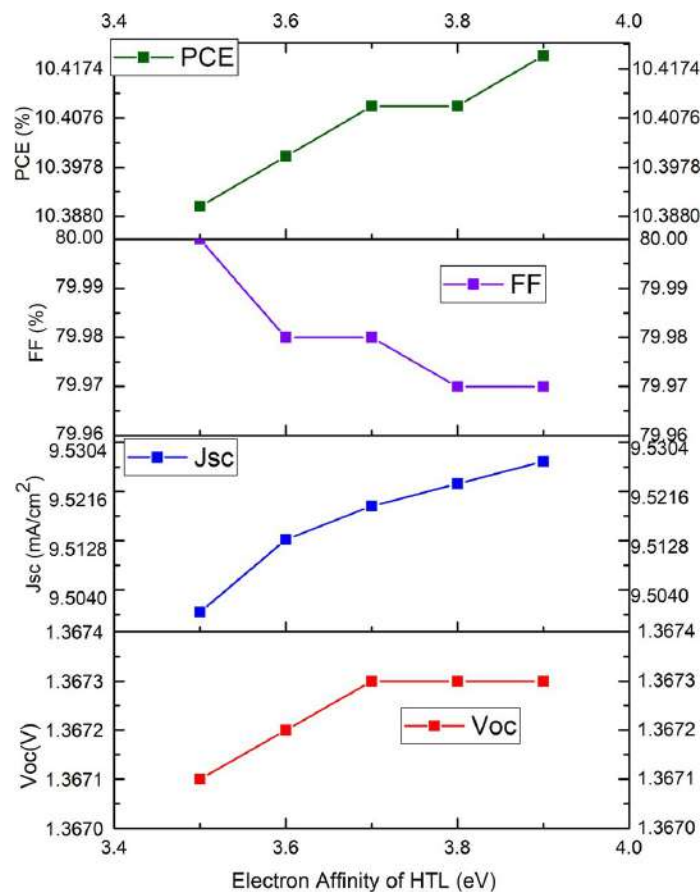


Figure 6.7: Influence of electron affinity

6.4.2.2 Influence of doping density

The significance of the doping density of HTL on cell performance is stated here. When the doping density of HTL was increased from 10^{17} to 10^{21} cm^{-3} , Voc, FF, and PCE fell accordingly. The greatest values of these output parameters found at a doping density of 10^{17} cm^{-3} are 1.3142V, 79.66%, and 9.24% respectively. Jsc increased by 8.833 mA/cm^2 at the optimal doping density of 10^{21} cm^{-3} . Fig 6.8 shows the relevant graphs. The electric field intensity at the HTL/absorber interface is affected by the

HTL doping density, which influences the solar cell's performance. The device gets more conductive as the HTL doping density increases, lowering the series resistance [157, 158].

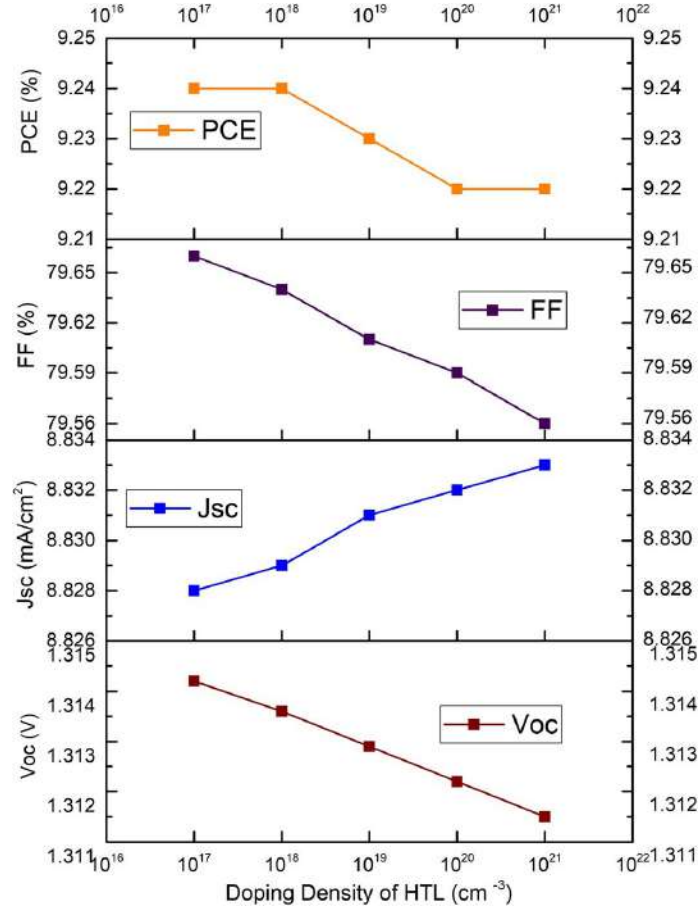


Figure 6.8: Influence of doping density

6.4.3 Effect of Electron affinity of ETL

In this section, the impacts of changing ETL's electron affinity from 3.6 to 4 eV on cell performance were explored. FF has a maximum value of 79.66% at 4 eV, Voc

has an optimal value of 1.3237V at 3.6 eV electron affinity, Jsc has an optimum value of 11.842 mA/cm², and PCE has an optimized value of 12.16% at 3.6 eV. Fig. 6.9 shows the respective graphs. The decline in Voc is due to an increasing energy-level mismatch between the active layer's LUMO (Lowest Unoccupied Molecular Orbital) and the ETL's conduction band, which reduces the maximum achievable voltage. As the ETL's electron affinity increases, the efficiency of electron extraction from the active layer to the ETL decreases, or hole-blocking is compromised, leading to increased charge recombination and lower short-circuit current. The fill factor shows a clear increasing trend due to the reduction in series resistance. These combined losses lead to a sharp overall decrease in the Power Conversion Efficiency (PCE).

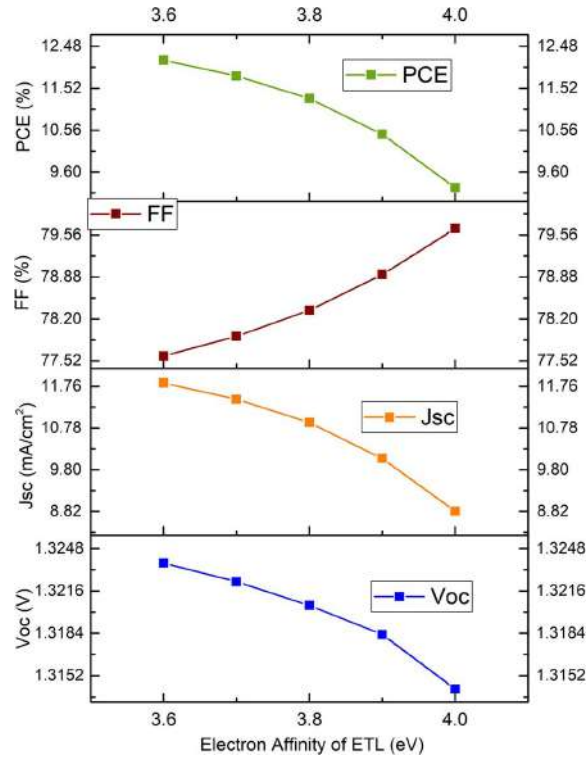


Figure 6.9: Influence of electron affinity

6.4.4 Influence of HTL/active layer interface defect density

Research testifies the fact that increasing the defect density of the HTL/Active Layer Interface from 10^{10} to 10^{14} cm^{-3} , leads to a drop in Voc, Jsc, FF and PCE. Values of 1.3899 V, 11.2571 mA/cm², 78.7% and 12.33% respectively, were obtained for output parameters when defect density becomes 10^{10} cm^{-3} . The respective graph is presented in Fig [6.10](#).

Rate of change of fill factor is lower than the rates at which Jsc and Voc is dropping, leading to a much lower PCE at output [\[149\]](#). At the HTL/active interface, output parameters drop steadily as defect density increases. All defects at the interface are also recombination centers for electrons and holes. This type of recombination, Shockley-Read-Hall (SRH) recombination, is an important loss mechanism reducing the quasi-Fermi level splitting and so the open-circuit voltage. As with the effect on Voc, interfacial defects trap charge carriers (this time holes trying to move to the HTL, and electrons that should be blocked). This greater probability of recombination means fewer charge carriers reach the electrodes and are collected as current, leading to loss of carriers, and thus reduces Jsc. The Fill Factor reflects the “squareness” of the J-V curve, and is sensitive to parasitic resistances and recombination.

A high density of interfacial defects acts as a shunt, allowing current to leak across the junction, and noted in a similar context that increased charge recombination and leakage current degrade the quality of the diodes means lower FF. The simultaneous degradation of voltage, current, and fill factor all contribute to this large loss of overall efficiency. The large drop in PCE is thus mainly driven by the severe drop in Voc.

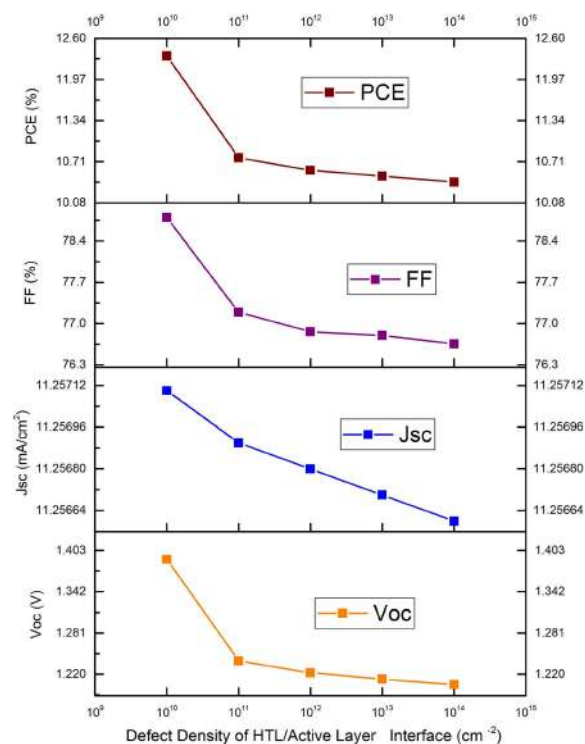


Figure 6.10: Influence of defect density of HTL/Active layer interface

6.4.5 Influence of Active Layer/ETL interface defect density

Varying the defect density of the active layer/ETL interface from 10^{11}cm^{-2} to 10^{15}cm^{-2} lowers V_{oc} , J_{sc} , and PCE while slightly increasing FF. The maximum values of V_{oc} , J_{sc} , and PCE at a defect density of 10^{11}cm^{-2} are 1.3337V, 15.411mA/cm², and 15.47% respectively. The best result for FF was 78.20% at 10^{15}cm^{-2} . The graphs are shown in Fig. 6.11. Increased defect density at interfaces enhances the likelihood of trap recombination, resulting in deterioration of the device. The lower the interface defect density, the better the output [149]. Table 6.4 shows the improved material properties. Table 6.5 compares the performance of the device for the initial and elevated values.

Simulations produce better results than the experimental structure.

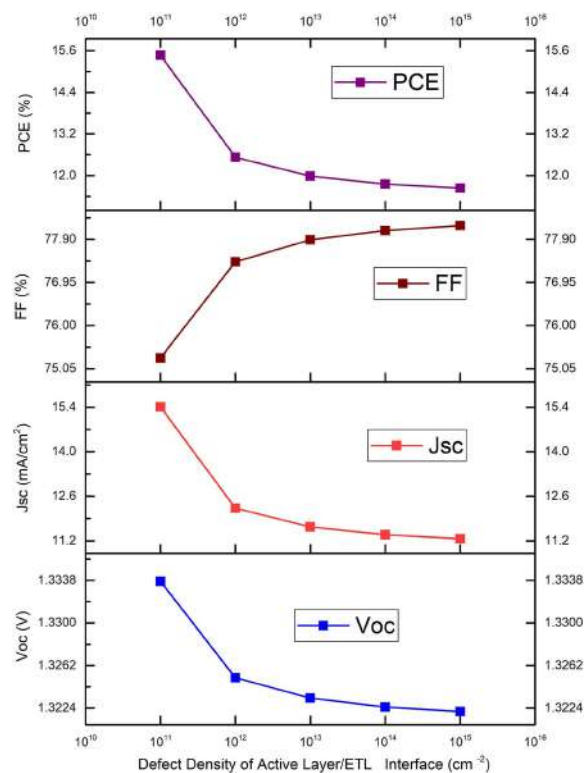


Figure 6.11: Influence of defect density of Active layer/ETL interface

Parameters	HTL	Active Layer	ETL
Doping Density [cm ⁻³]	10 ¹⁷	10 ¹⁶	10 ²⁰
Thickness [nm]	30	300	46

Table 6.4: Optimized material parameters

Parameters	Initial	Optimized
Voc [V]	0.84	1.1873
Jsc [mA/cm ²]	8.36	22.094
FF [%]	59	53.74
PCE [%]	3.9	14.10

Table 6.5: Comparison of organic solar cell with standard and optimized designs

6.5 Conclusion

This work draws attention to the issue of selecting appropriate charge transport layers to develop a highly efficient bulk heterojunction organic solar cell with proper interfacial stability and improved efficiency. It is a prerequisite to choose compatible charge transport layers, which makes it easy for the respective charge carriers to reach their respective electrodes. It is also a prerequisite to have a comparably low energy gap for the active layer for the easy and efficient uptake of the incident photons. A bulk heterojunction organic solar cell with P3HT:IC₆₀BA as the active layer, along with different HTL and ETL layers was investigated. The design, having cuprous oxide [Cu₂O] as the HTL (hole transport layer) and Indium gallium zinc oxide [IGZO] as the ETL (electron transport layer), achieves the highest power conversion efficiency (PCE). The upgraded design is further improved to dwell into the dependence of different material parameters on the output performance of the cell. The optimized parameters of structure are $V_{oc} = 1.1873\text{V}$, $J_{sc} = 22.09\text{ mA/cm}^2$, $FF = 53.74\%$, $PCE = 14.10\%$.

Chapter 7

Conclusion

This chapter summarizes the research work done on simulation studies of a bulk heterojunction organic solar cell with P3H:IC₆₀ as active layer

We have focused on the design of highly efficient bulk heterojunction organic solar cells through device modelling, and the performance of the designed devices are evaluated with varying material properties/operating conditions through simulation studies. Using the SCAPS device simulation tool, bulk heterojunction organic solar cells consisting of polymer donors and fullerene acceptors were modelled. In particular, P3HT:IC₆₀BA organic solar cells based on fullerenes were simulated. Multiple architectures were explored, such as the use of different transport-layer configurations, the incorporation of an additional P3HT layer, and even omitting the electron transport layer. The J–V characteristics of the simulated devices were compared with the experimental data to validate the simulation, indicating a close agreement between the two. The close correspondence between the simulated and experimental outputs validates the use of numerically mimicking parameters used in modelling the device. Optimisation of a P3HT:IC₆₀BA bulk heterojunction organic

solar cell design was undertaken, investigating the influence of different material parameters on cell performance. Maximizing operational efficiency requires optimizing the hole (HTL) and electron (ETL) transport layers for compatibility with the active layer. Extensive material investigation for both HTL and ETL candidates has led to the identification of the most effective combination, resulting in higher performance and enhanced efficiency

7.1 Important Findings

- Fullerene-based bulk heterojunction organic solar cell have shown a better performance when compared to experimental results using CuI as HTL . The results obtained are : $V_{oc} = 1.2170$ V, $J_{sc} = 6.9243$ mA/cm² , $FF = 73.95\%$, and $PCE = 6.23\%$
- Bulk heterojunction organic solar cell has given excellent results by omitting the ETL layer. The output parameters are $V_{oc} = 1.4158$ V; $J_{sc} = 10.294741$ mA/cm² ; $FF = 74.30\%$; $PCE = 10.83\%$
- The additional P3HT layer improved the performance of bulk heterojunction organic solar cell with output parameters $PCE = 15.26\%$, $FF = 51.62\%$, $J_{sc} = 26.577140$ mA/cm² , and $V_{oc} = 1.1126$ V which improved from 5.18% efficiency before optimization
- Influence of diverse HTL and ETL layers on device performance yielded excellent results . The most efficient combination for P3HT :IC₆₀BA solar cell consists of IGZO as ETL and Cu₂O as HTL with high device performance

having Voc of 1.1873V, Jsc of 22.09 mA/ cm^2 , FF of 53.74% and PCE of 14.10%.

- The material parameters of different layers influence the device performance of the organic solar cell
- The refinement of material parameters yields an enhanced operational output of the device, thereby facilitating further advancements in the field of organic photovoltaics

Chapter 8

Recommendations and Scope for Future Work

The realm of organic photovoltaics is of paramount importance for the reasons stated due the demand for energy. The need to commercialise low-cost and green organic solar cells is a requirement in order to satisfy the energy demands around the world. An appreciation of the theory and morphology of the organic solar cell lends itself to a stepping stone of ideas for organic photovoltaic research. This thesis may be followed up in the following ways:

- Optimizing fullerene organic solar cells using new materials like C_{70} and C_{84} by focusing on their properties can provide valuable insights for future study
- The influence of the mobility values of donors and acceptors in bulk heterojunction solar cells can offer a novel approach for optimizing organic materials to enhance their photovoltaic efficiency
- Comparative simulation studies between fullerene and non-fullerene materials as active layer

Bibliography

- [1] Ossila. Polymer-fullerene bulk heterojunction solar cells.
- [2] Jenny A. Nelson. *The physics of solar cells*. World Scientific Publishing Company, 2003.
- [3] Martin A. Green. *Solar cells: operating principles, technology, and system applications*. Prentice-Hall, Inc., Englewood Cliffs, NJ, 1981.
- [4] Steven S. Hegedus and Antonio Luque. Status, trends, challenges and the bright future of solar electricity from photovoltaics. *Handbook of Photovoltaic Science and Engineering*, pages 1–43, 2003.
- [5] H Weber and N Ahmed. A safe, reliable and regenerative electrical energy supply to power the future. In *EPJ Web of Conferences*, volume 246, page 00017. EDP Sciences, 2020.
- [6] Energy Institute. Statistical review of world energy, 2024.
- [7] Rebecca Saive. Entering the solar era: The next 50 years of energy generation. *The Bridge*, 50(5):134–137, 2021.

-
- [8] T. Markvart. Electricity from the sun. In T. Markvart, editor, *Solar Electricity*, pages 1–4. John Wiley, Chichester, England, 2000.
- [9] Ossila. Icba (indene-c60 bisadduct) fullerene.
- [10] Binrui Xu, Gopalan Sai-Anand, Anantha-Iyengar Gopalan, Qiquan Qiao, and Shin-Won Kang. Improving photovoltaic properties of p3ht: Ic60ba through the incorporation of small molecules. *Polymers*, 10(2):121, 2018.
- [11] Australian Bureau of Meteorology, Space Weather Services. The sun and solar activity—the solar constant, 2024.
- [12] U.S. Department of Energy. Solar photovoltaic cell basics, 2020.
- [13] International Energy Agency (IEA). CO₂ Emissions – Global Energy Review 2025. Report, International Energy Agency (IEA), 2025.
- [14] Will Atkinson. Volatility vs. affordability: Globally, renewables’ cost advantage grew last year. <https://rmi.org/affordability-not-volatility-renewables-cost-advantage-grows/>, 2025.
- [15] National Renewable Energy Laboratory (NREL). Solar photovoltaic technology basics, 2025.
- [16] Hamed H. Pourasl, Reza Vatankhah Barenji, and Vahid M. Khojastehnezhad. Solar energy status in the world: A comprehensive review. *Energy Reports*, 10:3474–3493, 2023.

-
- [17] E. Graham, N. Fulghum, et al. Solar power continues to surge in 2024. *Ember*, 2024.
- [18] Patrik Dahl. Environmental impacts of large-scale solar power construction in finland in accordance with the eu solar energy strategy, 2025. Add publisher or URL if available.
- [19] A. E. Becquerel. Mémoire sur la production des courants électriques dans le milieu électrolytique. *Comptes Rendus de l'Académie des Sciences*, 9:561, 1839.
- [20] Willoughby Smith. Effect of light on selenium. *Nature*, 7:303–304, 1873.
- [21] Charles Fritts. On a new form of selenium photocell. *American Journal of Science*, 26:196–204, 1883.
- [22] Albert Einstein. On a heuristic viewpoint concerning the production and transformation of light. *Annalen der Physik*, 17:132–148, 1905.
- [23] Russell S. Ohl. Light-sensitive electric device, June 1946. US Patent 2,402,662.
- [24] Daryl M. Chapin, Calvin S. Fuller, and Gerald L. Pearson. A new silicon p-n junction photocell for converting solar radiation into electrical power. *Journal of Applied Physics*, 25(5):676–677, 1954.
- [25] NASA. Vanguard i: The first solar-powered satellite. NASA Historical Archives, 1958. Add URL if desired.

-
- [26] M.A. Green, K. Emery, D.L. King, A. Iqbal, and V.L. Donley. Silicon solar cell efficiencies: Record progress. *Progress in Photovoltaics: Research and Applications*, 3:123–127, 1985.
- [27] Akihiro Kojima, Kenjiro Teshima, Yasuo Shirai, and Tsutomu Miyasaka. Organometal halide perovskites as visible-light sensitizers for photovoltaic cells. *Journal of the American Chemical Society*, 131(17):6050–6051, 2009.
- [28] Fraunhofer Institute for Solar Energy Systems ISE. Six-junction solar cells achieve a record efficiency of 47.1%. Press Release, 2023.
- [29] Hsiang-Yu Chen, Jianhui Hou, Su-Cheng Zhang, Su-Hung Chen, and Yang Yang. Polymer solar cells with enhanced open-circuit voltage and efficiency. *Nature Photonics*, 3(11):649–653, 2009.
- [30] Yongye Liang, Dan Feng, Yun-Xiang Wu, Hsin-Fei Tsai, Gang Li, and Luping Yu. Development of new semiconducting polymers for high-performance solar cells. *Journal of the American Chemical Society*, 131(1):56–57, 2009.
- [31] Jason T. Bloking, Tommaso Giovenzana, Andrew T. Higgs, Andrew J. Poncec, Eric T. Hoke, Koen Vandewal, Sangwon Ko, Zhenan Bao, Alan Sellinger, and Michael D. McGehee. Comparing the device physics and morphology of polymer solar cells employing fullerenes and non-fullerene acceptors. *Advanced Energy Materials*, 4(12):1301426, 2014.
- [32] Martin A. Green. *Third Generation Photovoltaics: Advanced Solar Energy Conversion*. Springer, Berlin, 2000.

-
- [33] Lewis M. Fraas. *Low-Cost Solar Electric Power*. Springer, New York, 2014.
- [34] Adolf Goetzberger, Christopher Hebling, and Hans-Werner Schock. Photovoltaic materials, history, status and outlook. *Materials Science and Engineering: R: Reports*, 40(1):1–46, 2003.
- [35] Martin A. Green, Ewan D. Dunlop, Jochen Hohl-Ebinger, Masahiro Yoshita, Nikos Kopidakis, Karsten Bothe, David Hinken, Michael Rauer, and Xiaojing Hao. Solar cell efficiency tables (version 60). *Progress in Photovoltaics: Research and Applications*, 30(7):687–701, 2022.
- [36] Martin A. Green, Ewan D. Dunlop, Jochen Hohl-Ebinger, Masahiro Yoshita, Nikos Kopidakis, Karsten Bothe, David Hinken, Michael Rauer, and Xiaojing Hao. Solar cell efficiency tables (version 61). *Progress in Photovoltaics: Research and Applications*, 31(7):651–663, 2023.
- [37] Martin A. Green. *Third-Generation Photovoltaics: Advanced Solar Energy Conversion*. Springer Series in Photonics. Springer, 2009.
- [38] Brian O'Regan and Michael Grätzel. A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO_2 films. *Nature*, 353(6346):737–740, 1991.
- [39] Hui-Seon Kim, Chang-Ryul Lee, Jeong-Hyeok Im, Ki-Beom Lee, Thomas Moehl, Arianna Marchioro, Soo-Jin Moon, Robin Humphry-Baker, Jun-Ho Yum, Jacques E Moser, et al. Lead iodide perovskite sensitized all-solid-state submicron thin film mesoscopic solar cell with efficiency exceeding 9%. *Scientific Reports*, 2(1):591, 2012.

-
- [40] Arthur J. Nozik. Quantum dot solar cells. *Physica E: Low-dimensional Systems and Nanostructures*, 14(1-2):115–120, 2002.
- [41] Tomas Leijtens, Giles E. Eperon, Sandeep Pathak, Antonio Abate, Michael Grätzel, and Henry J. Snaith. Stability of metal halide perovskite solar cells. *Advanced Energy Materials*, 7(16):1700503, 2017.
- [42] Ossila. Equivalent circuit of solar cell, 2025.
- [43] MathWorks. Solar cell. Documentation, 2025.
- [44] Libretexts. 4. recombination with defect levels, 2021.
- [45] Libretexts. 3. auger recombination. Web page, 2021.
- [46] Libretexts. 2. radiative (band-to-band) recombination. Web page, 2021.
- [47] Wolfgang Tress. Organic solar cells. In *Organic Solar Cells: Theory, Experiment, and Device Simulation*, pages 67–214. Springer, 2014.
- [48] P. A. Basore. Basic equations. Web page, 2004.
- [49] H. Kallmann and M. Pope. Photovoltaic effect in organic crystals. *The Journal of Chemical Physics*, 30(2):585–586, 1959.
- [50] C. W. Tang. Two-layer organic photovoltaic cell. *Applied Physics Letters*, 48(2):183–185, 1986.
- [51] Gang Yu, Jun Gao, Jan C. Hummelen, Fred Wudl, and Alan J. Heeger. Polymer photovoltaic cells: enhanced efficiencies via a network of internal donor-acceptor heterojunctions. *Science*, 270(5243):1789–1791, 1995.

-
- [52] Sean E. Shaheen, Christoph J. Brabec, N. Serdar Sariciftci, Franz Padinger, Thomas Fromherz, and Jan C. Hummelen. 2.5% efficient organic plastic solar cells. *Applied Physics Letters*, 78(6):841–843, 2001.
- [53] Christoph J. Brabec, Niyazi Serdar Sariciftci, and Jan C. Hummelen. Plastic solar cells. *Advanced Functional Materials*, 11(1):15–26, 2001.
- [54] Wanli Ma, Cuiying Yang, Xiong Gong, Kwanghee Lee, and Alan J. Heeger. Thermally stable, efficient polymer solar cells with nanoscale control of the interpenetrating network morphology. *Advanced Functional Materials*, 15(10):1617–1622, 2005.
- [55] Gang Li, Vishal Shrotriya, Jinsong Huang, Yan Yao, Tom Moriarty, Keith Emery, and Yang Yang. High-efficiency solution processable polymer photovoltaic cells by self-organization of polymer blends. *Nature Materials*, 4(11):864–868, 2005.
- [56] Yuze Lin, Jianhui Wang, Shaoqing Zhang, Jianhui Hou, and Xiaowei Zhan. A star-shaped molecule for high-efficiency non-fullerene organic solar cells. *Journal of the American Chemical Society*, 137(11):3730–3733, 2015.
- [57] Jun Yuan, Yunqiang Zhang, Liuyang Zhou, Guichuan Zhang, Hin-Lap Yip, Tsz-Ki Lau, Xinhui Lu, Can Zhu, Hongjian Peng, Paul A. Johnson, et al. Single-junction organic solar cell with over 15% efficiency using fused-ring acceptor with electron-deficient core. *Joule*, 3(4):1140–1151, 2019.
- [58] Martin Pope and Charles E. Swenberg. *Electronic processes in organic crystals and polymers*. Oxford University Press, 1999.

-
- [59] Antonio Facchetti. Semiconductors for organic transistors. *Materials Today*, 10(3):28–37, 2007.
- [60] Henning Sirringhaus. Device physics of solution-processed organic field-effect transistors. *Advanced Materials*, 17(20):2411–2425, 2005.
- [61] Pankaj Kumar. *Organic solar cells: device physics, processing, degradation, and prevention*. CRC Press, 2016.
- [62] Niyazi S. Sariciftci, L. Smilowitz, Alan J. Heeger, and Fred Wudl. Photoinduced electron transfer from a conducting polymer to buckminsterfullerene. *Science*, 258(5087):1474–1476, 1992.
- [63] Jingbi You, Letian Dou, Ken Yoshimura, Takehito Kato, Kenichiro Ohya, Tom Moriarty, Keith Emery, Chun-Chao Chen, Jing Gao, Gang Li, et al. A polymer tandem solar cell with 10.6% power conversion efficiency. *Nature Communications*, 4(1):1446, 2013.
- [64] Sih-Hao Liao, Yi-Lun Li, Tzu-Hao Jen, Yu-Shan Cheng, and Show-An Chen. Multiple functionalities of polyfluorene grafted with metal ion-intercalated crown ether as an electron transport layer for bulk-heterojunction polymer solar cells: optical interference, hole blocking, interfacial dipole, and electron conduction. *Journal of the American Chemical Society*, 134(35):14271–14274, 2012.
- [65] Dipan Kundu, Brian D. Adams, Victor Duffort, Shahrzad Hosseini Vajargah, and Linda F. Nazar. A high-capacity and long-life aqueous rechargeable zinc

- battery using a metal oxide intercalation cathode. *Nature Energy*, 1(10):16119, 2016.
- [66] Frederik C. Krebs. Fabrication and processing of polymer solar cells: A review of printing and coating techniques. *Solar Energy Materials and Solar Cells*, 93(4):394–412, April 2009.
- [67] Mikkel Jørgensen, Kion Norrman, Suren A. Gevorgyan, Thomas Tromholt, Birgitta Andreasen, and Frederik C. Krebs. Stability of polymer solar cells. *Advanced Materials*, 24(5):580–612, 2012.
- [68] Zhigang He, Chun-hong Tan, Zong-xiang Xu, and Chang-jian Lin. A solution-processed hole-transport layer for polymer light-emitting diodes. *Nature Photonics*, 5(3):178–182, mar 2011.
- [69] Yongsheng Chen, Xiangjian Wan, and Guankui Long. High performance photovoltaic applications using solution-processed small molecules. *Accounts of Chemical Research*, 46(11):2645–2655, 2013.
- [70] Harold W. Kroto, James R. Heath, Sean C. O’Brien, Robert F. Curl, and Richard E. Smalley. C60: Buckminsterfullerene. *Nature*, 318(6042):162–163, 1985.
- [71] Ramasamy Ganesamoorthy, Govindasamy Sathiyar, and Pachagounder Sakthivel. Fullerene based acceptors for efficient bulk heterojunction organic solar cell applications. *Solar Energy Materials and Solar Cells*, 161:102–148, 2017.

- [72] Zhicai He, Chengmei Zhong, Shijian Su, Miao Xu, Hongbin Wu, and Yong Cao. Enhanced power-conversion efficiency in polymer solar cells using an inverted device structure. *Nature Photonics*, 6(9):591–595, 2012.
- [73] Jan C. Hummelen, Brian W. Knight, F. LePeq, Fred Wudl, Jie Yao, and Charles L. Wilkins. Preparation and characterization of fulleroid and methanofullerene derivatives. *The Journal of Organic Chemistry*, 60(3):532–538, 1995.
- [74] Yongye Liang, Zheng Xu, Jiangbin Xia, Szu-Ting Tsai, Yue Wu, Gang Li, Claire Ray, Luping Yu, et al. For the bright future—bulk heterojunction polymer solar cells with power conversion efficiency of 7.4%. *Advanced Materials*, 22(20):E135, 2010.
- [75] Geneviève Sauvé and Roshan Fernando. Beyond fullerenes: designing alternative molecular electron acceptors for solution-processable bulk heterojunction organic photovoltaics. *The journal of physical chemistry letters*, 6(18):3770–3780, 2015.
- [76] Tiantian Cao, Ning Chen, Guangxin Liu, Yingbo Wan, José Darío Perea, Yijun Xia, Zhaowei Wang, Bo Song, Ning Li, Xiaohong Li, et al. Towards a full understanding of regioisomer effects of indene-c60 bisadduct acceptors in bulk heterojunction polymer solar cells. *Journal of Materials Chemistry A*, 5(21):10206–10219, 2017.
- [77] Fuwen Zhao, Xiangyue Meng, Yongqiang Feng, Zhiwen Jin, Qing Zhou, Hui Li, Li Jiang, Jizheng Wang, Yongfang Li, and Chunru Wang. Single crystalline

- indene-c60 bisadduct: isolation and application in polymer solar cells. *Journal of Materials Chemistry A*, 3(29):14991–14995, 2015.
- [78] Youjun He, Hsiang-Yu Chen, Jianhui Hou, and Yongfang Li. Indene-c60 bisadduct: a new acceptor for high-performance polymer solar cells. *Journal of the American Chemical Society*, 132(4):1377–1382, 2010.
- [79] Bing Guo, Wanbin Li, Xia Guo, Guangda Li, Xiangyi Meng, Wei Ma, Shanshan Chen, Maojie Zhang, and Kuan Sun. Fine-tuned active layer morphology for bulk heterojunction organic solar cells with indene-c60 bisadduct as a third component. *ACS Applied Materials & Interfaces*, 15(50):58693–58699, 2023.
- [80] Marc Burgelman, Peter Nollet, and Stefaan Degraeve. Modelling polycrystalline semiconductor solar cells. *Thin Solid Films*, 361:527–532, 2000.
- [81] Anthony C. Ozurumba, Nnamdi V. Ogueke, Chinyere A. Madu, Eli Danladi, Chisom P. Mbachu, Abubakar S. Yusuf, Philibus M. Gyuk, and Ismail Hos-sain. Scaps-1d simulated organometallic halide perovskites: A comparison of performance under sub-saharan temperature condition. *Heliyon*, 10(8), 2024.
- [82] Mahmoud Abdelfatah, Adel M. El Sayed, Walid Ismail, Stephan Ulrich, Volker Sittinger, and Abdelhamid El-Shaer. Scaps simulation of novel inorganic zrs2/cuo heterojunction solar cells. *Scientific Reports*, 13(1):4553, 2023.
- [83] Rahutosh Ranjan, Nikhil Anand, Manish Nath Tripathi, Neelabh Srivastava, Arvind Kumar Sharma, Masamichi Yoshimura, Li Chang, and Rajanish N. Tiwari. Scaps study on the effect of various hole transport layer on highly

- efficient 31.86% eco-friendly czts based solar cell. *Scientific Reports*, 13(1):18411, 2023.
- [84] Koen Decock, Samira Khelifi, and Marc Burgelman. Modelling multivalent defects in thin film solar cells. *Thin Solid Films*, 519(21):7481–7484, 2011.
- [85] ASTM Standard. G173-03 - standard tables for reference solar spectral irradiances: Direct normal and hemispherical on 37 tilted surface. *Ann. Book of ASTM Standards 2003*, 14:1–20, 2012.
- [86] Vívian Helene Diniz Araújo, Ana Flávia Nogueira, Juliana Cristina Tristão, and Leandro José Dos Santos. Fullerene-c60 and pcbm as interlayers in regular and inverted lead-free pscs using $\text{ch}_3\text{nh}_3\text{sn}_3$: an analysis of device performance and defect density dependence by scaps-1d. *RSC Advances*, 14(16):10930–10941, 2024.
- [87] Md Mojahidur Rahman, Md Hasan Ali, Md Dulal Haque, and Abu Zafor Md Touhidul Islam. Numerical modeling and extensive analysis of an extremely efficient rbgei₃-based perovskite solar cell by incorporating a variety of etl and htl materials to enhance pv performance. *Energy Advances*, 3(9):2377–2398, 2024.
- [88] P. Singh and S. N. Singh. Effect of interface defects on high efficiency a-si:h/ μ c-si:h tandem solar cells. *Solar Energy Materials and Solar Cells*, 95(1):266–272, 2011.

- [89] Takashi Minemoto and Masashi Murata. Device modeling of perovskite solar cells based on structural similarity with thin film inorganic semiconductor solar cells. *Journal of Applied Physics*, 116(5), 2014.
- [90] Muhammad Noman, Abdul Haseeb Hassan Khan, and Shayan Tariq Jan. Interface engineering and defect passivation for enhanced hole extraction, ion migration, and optimal charge dynamics in both lead-based and lead-free perovskite solar cells. *Scientific Reports*, 14(1):5449, 2024.
- [91] Julien Singer, Pierre Brand, and Philippe Thony. Method of simulation of an optoelectronic device, June 2015. US Patent App. 14/411,662.
- [92] Andrés Julián Aristizábal Cardona, Carlos Arturo Páez Chica, and Daniel Hernán Ospina Barragán. Pv generator characterization. In *Building-Integrated Photovoltaic Systems (BIPVS) Performance and Modeling Under Outdoor Conditions*, pages 65–78. Springer, 2018.
- [93] Jennifer T. Heath. Capacitance-voltage and drive-level-capacitance profiling. *Capacitance Spectroscopy of Semiconductors*, pages 85–109, 2018.
- [94] Weikai Fan, Wenquan Zhou, Le Chen, Chen Hu, Yiming Fu, Lin Peng, Xiaojing Liu, Jiang Wu, Ping He, and Jun Jin. Scaps-1d simulation and first-principles calculation of cssncl_3 hole transport layer-free perovskite solar cells based on gradient doping. *Materials Today Communications*, 40:109750, 2024.

-
- [95] M. Burgelman, P. Nollet, and S. Degrave. *SCAPS Manual: A Solar Cell Capacitance Simulator*. Department of Electronics and Information Systems (ELIS), 2024.
- [96] KS Nithya and KS Sudheer. Numerical modelling of non-fullerene organic solar cell with high dielectric constant itic-oe acceptor. *Journal of Physics Communications*, 4(2):025012, 2020.
- [97] KS Nithya and KS Sudheer. Device modelling of non-fullerene organic solar cell with inorganic cui hole transport layer using scaps 1-d. *Optik*, 217:164790, 2020.
- [98] S. Rasool, V. V. Doán, H. K. Lee, S. K. Lee, J. Lee, S. Moon, W. So, C. E. Song, and W. S. Shin. Enhanced photostability in polymer solar cells achieved with modified electron transport layer. *Thin Solid Films*, 669:42, 2018.
- [99] Syed Zulqarnain Haider, Hafeez Anwar, Sehrish Manzoor, Ahmad G Ismail, and Mingqing Wang. A theoretical study for high-performance inverted pin architecture perovskite solar cells with cuprous iodide as hole transport material. *Current Applied Physics*, 20(9):1080–1089, 2020.
- [100] S. Samaki, F. T. Nya, G. M. D. Kenfack, and A. Laref. Materials and interfaces properties optimization for high-efficient and more stable rbgei3 perovskite solar cells: optoelectrical modelling. *Scientific Reports*, 13(1), 2023.
- [101] Jeromy J Rech, Liang Yan, Zhengxing Peng, Shuixing Dai, Xiaowei Zhan, Harald Ade, and Wei You. Utilizing difluorinated thiophene units to improve

- the performance of polymer solar cells. *Macromolecules*, 52(17):6523–6532, 2019.
- [102] Chaoyuth Sae-Kung, Brendan F Wright, Tracey M Clarke, Gordon G Wallace, and Attila J Mozer. Effects of interfacial layers on the open circuit voltage of polymer/fullerene bulk heterojunction devices studied by charge extraction techniques. *ACS Applied Materials & Interfaces*, 11(23):21030–21041, 2019.
- [103] S. Zhang, M. Stolterfoht, A. Armin, Q. Lin, F. Zu, J. Sobuś, H. Jin, N. Koch, P. Meredith, P. L. Burn, and D. Neher. Interface engineering of solution-processed hybrid organohalide perovskite solar cells. *ACS Applied Materials Interfaces*, 10(25):21681, 2018.
- [104] M. Lanzi, E. Salatelli, L. Giorgini, A. Mucci, F. Pierini, and F. P. D. Nicola. Water-soluble polythiophenes as efficient charge-transport layers for the improvement of photovoltaic performance in bulk heterojunction polymeric solar cells. *European Polymer Journal*, 97:378, 2017.
- [105] Jos’e G S’anchez, Victor S Balderrama, Salvador I Garduno, Edith Osorio, Aurelien Viterisi, Magali Estrada, Josep Ferr’e-Borrull, Josep Pallares, and Lluís F Marsal. Impact of inkjet printed zno electron transport layer on the characteristics of polymer solar cells. *RSC advances*, 8(24):13094–13102, 2018.
- [106] Han Pan, Xiaojuan Zhao, Xiu Gong, Hao Li, Najib Haji Ladi, Xiao Li Zhang, Wenchao Huang, Shahzada Ahmad, Liming Ding, Yan Shen, et al. Advances in design engineering and merits of electron transporting layers in perovskite solar cells. *Materials Horizons*, 7(9):2276–2291, 2020.

-
- [107] Xinxin Huang, Huangzhong Yu, Shengwei Shi, and Chengwen Huang. Improving the performance of inverted polymer solar cells by the efficiently doping and modification of electron transport layer-zno. *Organic Electronics*, 65:311–320, 2019.
- [108] Soyeon Kim, Jaehoon Jeong, Quoc Viet Hoang, Joo Won Han, Adi Prasetyo, Muhammad Jahandar, Yong Hyun Kim, Shinuk Cho, and Dong Chan Lim. The role of cation and anion dopant incorporated into a zno electron transporting layer for polymer bulk heterojunction solar cells. *RSC advances*, 9(65):37714–37723, 2019.
- [109] Wenna Zhang, Fanchen Bu, Wenfei Shen, Xiaohua Qi, Na Yang, Mengyao Chen, Di Yang, Yao Wang, Miaorong Zhang, Haoyang Jiang, et al. Strongly enhanced efficiency of polymer solar cells through unzipped swnt hybridization in the hole transport layer. *RSC advances*, 10(42):24847–24854, 2020.
- [110] SH Tsai, ST Ho, HJ Jhuo, CR Ho, SA Chen, and Jr-Hau He. Toward high efficiency of inverted organic solar cells: Concurrent improvement in optical and electrical properties of electron transport layers. *Applied Physics Letters*, 102(25), 2013.
- [111] Sheng Bi, Zhongliang Ouyang, Shoieb Shaik, and Dawen Li. Effect of donor-acceptor vertical composition profile on performance of organic bulk heterojunction solar cells. *Scientific reports*, 8(1):9574, 2018.
- [112] Maryam Hashemi, Mehran Minbashi, Seyed Mohammad Bagher Ghorashi, and Arash Ghobadi. A modeling study on utilizing low temperature sprayed in2s3

- as the buffer layer of cubasn (s, se) solar cells. *Scientific reports*, 11(1):20038, 2021.
- [113] Nidhi Sharma, Saral K Gupta, and Chandra Mohan Singh Negi. Influence of active layer thickness on photovoltaic performance of ptb7: Pc70bm bulk heterojunction solar cell. *Superlattices and Microstructures*, 135:106278, 2019.
- [114] Faisal Saeed, Muhammad Haseeb Khan, Haider Ali Tauqeer, Asfand Haroon, Asad Idrees, Syed Mzhar Shehrazai, Lukas Prokop, Vojtech Blazek, Stanislav Misak, and Nasim Ullah. Numerical investigation of photo-generated carrier recombination dynamics on the device characteristics for the perovskite/carbon nitride absorber-layer solar cell. *Nanomaterials*, 12(22):4012, 2022.
- [115] Poonam Subudhi and Deepak Punetha. Pivotal avenue for hybrid electron transport layer-based perovskite solar cells with improved efficiency. *Scientific Reports*, 13(1):19485, 2023.
- [116] M Khalid Hossain, Md Aminul Islam, M Shihab Uddin, Prabhu Paramasivam, Junainah Abd Hamid, Razan A Alshgari, VK Mishra, and Rajesh Haldhar. Design and simulation of cspb. 625zn. 375ibr2-based perovskite solar cells with different charge transport layers for efficiency enhancement. *Scientific Reports*, 14(1):30142, 2024.
- [117] Martin Stolterfoht, Pietro Caprioglio, Christian M Wolff, José A Márquez, Joleik Nordmann, Shanshan Zhang, Daniel Rothhardt, Ulrich Hörmann, Yohai Amir, Alex Redinger, et al. The impact of energy alignment and interfacial

- recombination on the internal and external open-circuit voltage of perovskite solar cells. *Energy & environmental science*, 12(9):2778–2788, 2019.
- [118] S. Ijaz, M. A. Monnaf, M. R. Islam, and M. A. H. M. Shajib. A numerical approach to optimize the performance of htl-free carbon electrode-based perovskite solar cells using organic etls. *Heliyon*, 10(6):e27798, 2024.
- [119] Md Tashfiq Bin Kashem and Sadia Anjum Esha. A Comprehensive Computational Photovoltaic Study of Lead-free Inorganic NaSnCl₃-based Perovskite Solar Cell: Effect of Charge Transport Layers and Material Parameters, 2025.
- [120] A. Ali, H. Noor, A. Aiman, A. Karim, M. Rashid, M. Mumtaz, M. Ahsan, and A. Illahi. Design and numerical simulation of lead-free inorganic perovskites (Cs₂BiAgI₆) solar cell through computation method. *Heliyon*, 11(3):e42361, 2025.
- [121] Mathias Nyman, Christian Ahläng, Staffan Dahlström, Manasi Pranav, Johannes Benduhn, Syeda Qudsia, Jan-Henrik Smått, Donato Spoltore, and Ronald Österbacka. Highly conductive charge transport layers impair charge extraction selectivity in thin-film solar cells. *Advanced Energy and Sustainability Research*, 4(10):2300030, 2023.
- [122] S. Hosseini, M. Bahramgour, N. Delibaş, and A. Niaie. Investigation of a perovskite solar cell and various parameters impact on its layers and the effect of interface modification by using p3ht as an ultrathin polymeric layer through scaps-1d simulation. *Sakarya University Journal of Science*, 25(5):1168–1179, 2021.

-
- [123] A. Maillard and A. Rochefort. Quantum transport properties of graphene nanoribbons with embedded heptagon-pentagon defects. *Organic Electronics*, 15(9):2091–2098, 2014.
- [124] H. Lee. Optimization of energy level alignment for efficient organic photovoltaics. *Vacuum Magazine*, 2(2):12–16, 2015.
- [125] Sarah Holliday, , et al. High-efficiency and air-stable P3HT solar cells with a new non-fullerene acceptor. *Nature Communications*, 7:11585, 2016.
- [126] Olga D. Iakobson, Oxana L. Gribkova, Alexey R. Tameev, and Jean-Michel Nunzi. Polymeric semiconductors for hybrid organic-inorganic solar cells: A comparative study of water-soluble polythiophenes. *Scientific Reports*, 11(1):3697, 2021.
- [127] N. Sharma, S.K. Gupta, and C.M. Singh Negi. Enhanced performance of organic solar cells by using zinc oxide and graphene quantum dots as electron transport layer. *Superlattices and Microstructures*, 135:106278, 2019.
- [128] Kangkang Weng, Linglong Ye, Lei Zhu, Jinqiu Xu, Jiajia Zhou, Xiang Feng, Guanghao Lu, Songting Tan, Feng Liu, and Yanming Sun. Optimized active layer morphology toward efficient and polymer batch insensitive organic solar cells. *Nature Communications*, 11(1):2855, 2020.
- [129] Yingying Zhang, Xiong Li, Tingting Dai, Denghui Xu, Jianfeng Xi, and Xiaobai Chen. Charge transport and extraction of ptb7:pc71bm organic solar cells: effect of film thickness and thermal-annealing. *RSC Adv.*, 9:24895–24903, 2019.

-
- [130] Abhinav Gusain, Roberto M. Faria, and Paulo B. Miranda. Controlling the morphology and performance of bulk heterojunctions in solar cells: Lessons from interfacial forces. *Frontiers in Chemistry*, 7:61, 2 2019.
- [131] M. Zhang, X. Xu, L. Yu, and Q. Peng. High-performance ternary organic solar cells with controllable morphology via sequential layer-by-layer deposition. *Journal of Power Sources*, 506:229961, 2021.
- [132] D. Spoltore, W. D. Oosterbaan, S. Khelifi, J. N. Clifford, A. Viterisi, E. Palomares, M. Burgelman, et al. A combined experimental and modeling study of the factors limiting the performance of polymer:fullerene solar cells processed from chlorobenzene and 1,2- dichlorobenzene. *Advanced Energy Materials*, 3(2):227–236, 2013.
- [133] Muhammad Ramzan Khan and Bogusław Jarzabek. Recent advances in polymer-based materials for high-performance perovskite solar cells. *Polymers*, 15(18):3674, 2023.
- [134] Carsten Deibel. Photocurrent in organic solar cells – part 1. Blog post, Notes on Disordered Matter, July 2009.
- [135] B. Qi and J. Wang. Fill factor in organic solar cells. *Journal of Materials Chemistry*, 22(46):24315–24325, 2012.
- [136] M. Wright and A. Uddin. Organic-inorganic hybrid perovskites: A solution for cost-effective solar cells. *Solar Energy Materials and Solar Cells*, 107:87–117, 2012.

-
- [137] S. Galindo, M. Ahmadpour, L. G. Gerling, A. Marsal, C. Voz, R. Alcubilla, and J. Puigdollers. Analysis of the origin of open circuit voltage in organic solar cells with different device architectures. *Organic Electronics*, 15(11):3034–3041, 2014.
- [138] Z. Liu and Y. Lin. Recent advances in polymer-based interfacial materials for efficient and stable organic solar cells. *Polymer Testing*, 131:108387, 2024.
- [139] A. Kumar and S. Sharma. Performance analysis of organic solar cells with different anode buffer layers. *IOSR Journal of Electrical and Electronics Engineering (IOSR-JEEE)*, 14(4):49–54, 2019.
- [140] Jianjun Yang, Xiansheng Wang, Xiaobao Yu, Jiaxuan Liu, Zhi Zhang, Jian Zhong, and Junsheng Yu. Improved short-circuit current and fill factor in pm6:y6 organic solar cells through d18-cl doping. *Nanomaterials*, 13(21):2899, 2023.
- [141] Sreeram Valsalakumar, Shubhranshu Bhandari, Anurag Roy, Tapas K. Mallick, Justin Hinshelwood, and Senthilarasu Sundaram. Machine learning driven performance for hole transport layer free carbon-based perovskite solar cells. *npj Computational Materials*, 10(1):212, 2024.
- [142] T.M. Mukametkali, B.R. Ilyassov, A.K. Aimukhanov, T.M. Serikov, A.S. Baltabekov, L.S. Aldasheva, and A.K. Zeinidenov. Optical and electronic properties of monolayer mos under different strains: Dft study. *Physica B: Condensed Matter*, 660:414784, 2023.

-
- [143] S. Sakib, M.Y. Mohd Noor, M.R. Salim, A.S. Abdullah, A.I. Azmi, M.H. Ibrahim, and M.H. Ibrahim. Effect of different composition ratio on structural and morphological properties of tio nanoparticles synthesized by sol-gel method. *Materials Today: Proceedings*, 76:176–182, 2023.
- [144] Carl Poelking, Johannes Benduhn, Donato Spoltore, Martin Schwarze, Steffen Roland, Fortunato Piersimoni, Dieter Neher, Karl Leo, Koen Vandewal, and Denis Andrienko. Open-circuit voltage of organic solar cells: interfacial roughness makes the difference. *Communications Physics*, 5(1):307, 2022.
- [145] Muhammad Raheel Khan and Bożena Jarzabek. The effect of 3d printing process parameters on the mechanical properties of pla polymer and polymer composites: A review. *Polymers*, 15(18):3674, 2023.
- [146] Syed Abdul Moiz, Mohammed Saleh Alzahrani, and Ahmed N. M. Alahmadi. Influence of process parameters on mechanical properties of natural fiber-reinforced polymer composites: A review. *Polymers*, 14(17):3610, 2022.
- [147] Davide Bartesaghi, Irene del Carmen Pérez, Juliane Kniepert, Steffen Roland, Mathieu Turbiez, Dieter Neher, and L Jan Anton Koster. Competition between recombination and extraction of free charges determines the fill factor of organic solar cells. *Nature Communications*, 6:7083, 2015.
- [148] J. C. Nolasco, R. Cabré, J. Ferré-Borrull, L. F. Marsal, M. Estrada, and J. Pallarès. Design of two-dimensional silicon photonic crystals by means of a guided-mode resonant filter structure. *Journal of Applied Physics*, 107(2):023108, 2010.

-
- [149] K.S. Nithya and K.S. Sudheer. Device modelling and optimization studies on novel itic-oe based non-fullerene organic solar cell with diverse hole and electron transport layers. *Optical Materials*, 123:111912, 2022.
- [150] Noya Loew, Shigehiro Komatsu, Hiroshi Akita, Katsuya Funayama, Tetsuharu Yuge, Takashi Fujiwara, and Manabu Ihara. Tio₂ as electron-extraction-layer in reverse type p3ht/icba organic solar cells. *ECS Transactions*, 58(45):77–88, 2014.
- [151] M.S.I. Sadek, M.S. Aktar, M.F.K. Shimul, and M.A. Islam. Performance analysis of lead-free all-inorganic cs₂sn_i6 based perovskite solar cell with diverse hole and electron transport layers. *Journal of Engineering Studies and Research*, 1(1):25–30, 2021.
- [152] Nguyen Dinh Lam. Modelling and numerical analysis of zno/cuo/cu₂o heterojunction solar cell using scaps. *Engineering Research Express*, 2(2):025033, 2020.
- [153] W. Abdelaziz, A. Zekry, A. Shaker, and M. Abouelatta. Numerical study of organic graded bulk heterojunction solar cell using scaps simulation. *Solar Energy*, 211:375–382, 2020.
- [154] Hung-Yang Lin, Guan-Jhih Li, Chih-Ping Chen, and Show-An Chen. A new donor–acceptor-structured poly(triphenylamine-alt-benzothiadiazole) derivative for bulk heterojunction solar cells. *Solar Energy Materials and Solar Cells*, 122:264–272, 2014.

-
- [155] Shambhavi Rai, BK Pandey, and DK Dwivedi. Modeling of highly efficient and low cost $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}-\text{xCl})_3$ based perovskite solar cell by numerical simulation. *Optical Materials*, 100:109631, 2020.
- [156] Lhoussayne Et-taya, Touria Ouslimane, and Abdellah Benami. Numerical analysis of earth-abundant $\text{Cu}_2\text{ZnSn}(\text{SxSe1-x})_4$ solar cells based on spectroscopic ellipsometry results by using scaps-1d. *Solar Energy*, 201:827–835, 2020.
- [157] W. Abdelaziz, A. Shaker, M. Abouelatta, and A. Zekry. Possible efficiency boosting of non-fullerene acceptor solar cell using device simulation. *Optical Materials*, 91:239–245, 2019.
- [158] Zhicai He, Feng Liu, Cheng Wang, Jihua Chen, Lilin He, Dennis Nordlund, Hongbin Wu, Thomas P. Russell, and Yong Cao. Simultaneous spin-coating and solvent annealing: manipulating the active layer morphology to a power conversion efficiency of 9.6 *Materials Horizons*, 2:592–597, 2015.

List of Publications

- C.D.Ramabadran and K.S.Sudheer A Study on the influence of different HTL and ETL layers on the performance of a P3HT:IC₆₀BA bulk heterojunction organic solar cell, Indian Journal of Physics (2024) <https://doi.org/10.1007/s12648-024-03278-1>
- Ramabadran C D, Dhanya N P , Sudheer K S Paper titled “IMPROVED EFFICIENCY OF BULK HETEROJUNCTION POLYMER SOLAR CELLS BY OMITTING ELECTRON TRANSPORT LAYER : A SCAPS 1D SIMULATION STUDY” Vietnam Journal of Science and Technology, 63 (5) (2025) 985-1002, doi:10.15625/2525-2518/20703
- C.D.Ramabadran and K.S.Sudheer Paper titled “INFLUENCE OF AN ADDITIONAL P3HT LAYER ON THE PERFORMANCE OF P3HT: IC₆₀BA POLYMER SOLAR CELL”, East European Journal of Physics (2025) DOI:10.26565/2312-4334-2025-3-50
- Study of influence of inorganic CuI hole transport layer on bulk heterojunction P3HT:IC₆₀BA polymer solar cells using SCAPS 1-D :C. D. Ramabadran and K. S. Sudheer AIP Conf. Proc. 2901, 040001 (2023) <https://doi.org/10.1063/5.0178644>

List of Presentations

- Presented the paper “ Study of influence of inorganic CuI hole transport layer on bulk heterojunction P3HT:IC60BA polymer solar cells using SCAPS 1-D “as in oral session (Online) at Third International Conference on Advances in Physical Sciences and Materials (ICAPSM 2022) held from 18 – 19 August 2022 at Coimbatore
- Presented a paper at the International Seminar on Emerging Technologies for Sustainable Development held at Government Victoria College, Palakkad on 20-21st December 2023
- Presented the paper” Comparative Simulation Study on the Performance of Polymer solar cell with two different Electron Transport Layers” as an oral presentation at Multidisciplinary National Conference and Research Expo 2024 Christ College[Autonomous],Irinjalakuda,Kerala on 14- 15th February , 2024
- Presented a paper at the International Conference on Innovative Functional Materials [ICIFM -2025] at Christ College [Autonomous], Irinjalakuda on February 17-18, 2025
- Presented a paper at the Second International Conference on Innovative Functional Materials [ICIFM -2026] at Christ College [Autonomous], Irinjalakuda on January 13-14, 2026

INFLUENCE OF AN ADDITIONAL P3HT LAYER ON THE PERFORMANCE OF P3HT: IC₆₀BA POLYMER SOLAR CELL

 Chittur Devarajan Ramabadran^{a,b§},  K. Sebastian Sudheer^{a*}

^aDepartment of Physics, Opto-electronic Device Simulation Research Lab, Christ College [Autonomous], Irinjalakuda, Thrissur, 680125, University of Calicut, Kerala, India

^bDepartment of Physics, Government College, Chittur, Palakkad, 678104, University of Calicut, Kerala, India

Corresponding Author e-mail: [§]ramabadrandc@christcollegeijk.edu.in; *e-mail: sudheersebastian@christcollegeijk.edu.in

Received May 9, 2025; revised July 11, 2025; accepted July 17, 2025

A simulation study utilizing SCAPS 1-D software was conducted to explore the effects of an additional P3HT (Poly 3-hexylthiophene) layer on the performance of bulk heterojunction polymer solar cells, specifically with the active layer P3HT: IC₆₀BA. The investigated cell structure is ITO/PEDOT:PSS/P3HT/P3HT:IC₆₀BA/ZnO NPs/Al. Following the standardization of the software, we determined the optimal parameters of the solar cell structure by analyzing various factors influencing cell performance across different layers. Subsequently, after optimizing the structure, the power conversion efficiency (PCE) improved significantly, rising from 5.18% without additional layer to 15.26% with additional layer.

Keywords: Bulk heterojunction polymer solar cell; Fullerene; SCAPS 1-D; Active Layer; ETL; HTL

PACS: 84.60.Jt; 81.05.Lg; 07.05.Tp

1. INTRODUCTION

A polymer solar cell (PSC) is an organic solar cell that utilizes polymers as the primary component for converting sunlight into electrical energy. The operation of PSCs relies on photoinduced charge transfer between a donor (the polymer) and an acceptor, typically a fullerene derivative. When exposed to sunlight, excitons are generated within the polymer, which then separate into electrons and holes at the donor-acceptor interface. These charge carriers are subsequently collected at the electrodes, producing an electric current. Key characteristics of PSCs include their ease of fabrication, adaptability, and low production costs. Additionally, they can be made partially transparent. However, a significant limitation is their relatively low power conversion efficiency (PCE), comparable to that of traditional silicon solar cells. While PSCs hold environmental advantages, they often exhibit lower stability and increased susceptibility to environmental changes.

Polymer solar cells (PSCs) can be broadly categorized into four main types: tandem PSCs, bulk heterojunction PSCs (the most prevalent and favored type), single-layer PSCs, and bilayer PSCs. These solar cells find applications in wearable technology, portable electronics, integrated photovoltaics, and energy harvesting for the Internet of Things. One of the key materials used in bulk heterojunction (BHJ) polymer solar cells is poly(3-hexylthiophene) (P3HT), which is notable for its high hole mobility, environmental stability, and extended absorption in the red region. Consequently, [6,6]-phenyl-C61-butyric acid methyl ester (PCBM), a soluble C60 derivative, has emerged as one of the most widely utilized organic semiconductors for acting as an electron acceptor [1-3]. P3HT:PCBM solar cells have demonstrated an open circuit voltage (Voc) of approximately 0.6 V [4], attributed to the low LUMO energy levels of PCBM, which is around -3.8 eV. In 2009, a novel C60 derivative named indene-C60 bisadduct (IC₆₀BA) was introduced, and due to its higher LUMO energy levels of approximately -3.74 eV, IC₆₀BA has increasingly replaced PCBM as the preferred acceptor material in these solar cells [14]. This substitution has resulted in improved efficiencies and a higher Voc of about 0.84 V [5-9].

Bulk heterojunction organic solar cells (OSCs), in which the donor and acceptor materials are combined to form the active layer, were first proposed by Yu et al. in 1995. Since the introduction of this bulk heterojunction (BHJ) structure, the power conversion efficiency (PCE) of BHJ OSCs has significantly improved, rendering it a promising technology. Heeger and colleagues [10] were the first to utilize the BHJ structure for polymer-fullerene blends. Subsequent research revealed that increasing the diameters of the donor and acceptor domains through various additive techniques leads to enhanced performance [11,12]. In an effort to improve results and address the limitations associated with the poly(3-hexylthiophene) and [6,6]-phenyl-C61-butyric acid methyl ester (P3HT:PCBM) blend, which exhibits a relatively low open-circuit voltage (Voc) of 0.6 V, Li et al. demonstrated the use of indene-C60 bisadduct (IC₆₀BA) as an electron acceptor in BHJ PSCs [13]. This P3HT:IC₆₀BA combination resulted in higher PCE values and an increased Voc of approximately 0.84 V [14,15].

In this study, the solar cell comprised of P3HT:IC₆₀BA (poly(3-hexylthiophene): (indene-C60 bisadduct) was simulated using SCAPS 1-D Software [16]. The simulation software was standardized by comparing the simulated output with experimental data [17]. In the numerical simulation of the polymer solar cell (PSC), the hole transport layer (HTL) is represented by PEDOT:PSS, the active layer by the fullerene blend P3HT: IC₆₀BA, and the electron transport layer (ETL) by ZnO nanoparticles (ZnONPs). Aluminum (Al) and indium tin oxide (ITO) serve as the electrode materials in

this structure. The performance of a polymer solar cell with P3HT: IC₆₀BA as the active layer is improved when P3HT is added as an extra layer. Based on previous investigations, the operational performance and efficiency of the device were augmented to 26.5% through the incorporation of an ultrathin P3HT polymeric layer serving as an interfacial medium between the hole transport material and the perovskite layer [18]. In order to effectively separate and collect charges, P3HT helps to increase the mobility of electrons and holes, which adds to improved charge transport qualities. It assists in maximizing the open-circuit voltage and current density by maximizing energy level alignment at the heterojunction interface [19,20]. P3HT can improve the active layer's structural ordering and crystallinity, resulting in bigger crystal domain sizes that strengthen the interpenetrating network necessary for efficient charge transfer [17]. Furthermore, P3HT's solubility in common solvents makes it possible to use simple solution-processing processes, which increases the layers' scalability and compatibility with a range of production methods [21]. All things considered, these characteristics help the solar cells' power conversion efficiency to rise noticeably. To the best of our knowledge, this is the first simulation study reporting the impact of using P3HT on Polymer solar cell with P3HT:IC₆₀BA as active layer. In this simulation, we specifically investigated the impact of adding P3HT layer to the structure above.

2. DEVICE SIMULATION METHODOLOGY

The one-dimensional Solar Cell Capacitance Simulator (SCAPS) version 3.3.08 was employed as the numerical simulation tool in this investigation. This software is capable of solving both optical and electrical models throughout the entire configuration. We utilized a 1.5 AM spectrum for illumination. Previous simulations have focused on various types of solar cells, including perovskite, fullerene, non-fullerene bulk heterojunction (BHJ), and tandem solar cells (Abdelaziz et al., 2019, 2020; Gupta and Dixit, 2018; Bahrami et al., 2019). The application allows users to modify parameters and select the desired output across its various panels. Additionally, the software numerically solves Burgelman et al.'s generic semiconductor equations to obtain a steady-state operating point solution [16]

Equations foundational to semiconductors, including the Poisson equation, continuity equations, drift equations, and diffusion current equations, are used to efficiently evaluate the electrical performance of electronic devices when stimulated. The stimulation can take the form of heat energy, photons, or voltages. This indicates that we can compute the electrical performance of the devices and analyze their I-V characteristics using these equations. Consequently, these equations provide a theoretical basis for interpreting and assessing the measured performance of the devices. Furthermore, they can be employed to reduce experimental labor costs and to evaluate and predict the performance of new device structures, potentially leading to significant savings in both time and money.

They can be applied to streamline mathematical analysis and determine the ideal device structure parameters. The local microscopic behaviour of the material is described by these differential equations. The local electric field change resulting from the volumetric charge density at a particular location within a material is described by the Poisson equation [Equation 1]. The conservation of electrons and holes at a specific place in the material is expressed by the continuity equations [Equation 2]. In this instance, an electric field (drift current) or a concentration gradient (diffusion current) is driving the electron and hole currents [Equations 4-5]. [22]

$$\frac{\partial \epsilon_0 \epsilon}{\partial x} \frac{\partial \Psi}{\partial x} = -q (p - n + N_D - N_A + \frac{\rho_{def}}{q}), \quad (1)$$

$$-\frac{\partial J_n}{\partial x} - Un + G = \frac{\partial n}{\partial t}, \quad (2)$$

$$-\frac{\partial J_p}{\partial x} - Up + G = \frac{\partial p}{\partial t}, \quad (3)$$

$$J_n = -\frac{\mu_n n}{q} \frac{\partial E_{Fn}}{\partial x}, \quad (4)$$

$$J_p = +\frac{\mu_p p}{q} \frac{\partial E_{Fp}}{\partial x}. \quad (5)$$

In the abovementioned equations, the charge is denoted by q , and the electrostatic potential is denoted by Ψ . The symbols p , n , stand for free holes and electrons, respectively. Ionised donor-like and ionised acceptor-like doping concentrations are denoted by N_D and N_A . Here, ϵ signifies absolute permittivity, while ϵ_0 denotes permittivity in free space. The terms J_n and J_p signify electron and hole current densities, respectively, whereas Un denotes electron recombination rate. G is the generation rate, while Up is the rate of hole recombination. In contrast to what has been described so far, E_{Fn} and E_{Fp} stand for the electron and hole quasi-Fermi levels, whereas n and p indicate the electron and hole mobility, respectively

The glass substrate/ITO/PEDOT: PSS/P3HT:IC₆₀BA/ZnONPS/Al simulated bulk heterojunction structure is shown in Figure 1. The simulation's material properties, such as electron affinity (χ), bandgap energy (E_g), and relative permittivity (ϵ_r), are displayed in Table 1. Hole and electron mobilities are μ_p and μ_n , respectively. Donor and acceptor densities are denoted by N_D and N_A , respectively. The effective densities of states in the valence and conduction bands are denoted by N_v and N_c , respectively, while the defect density is represented by N_t . 4.8 and 4.2 eV are the settings for the anode and cathode work functions, respectively. The capture cross section for holes and electrons is set to $1 \times 10^{-25} \text{ cm}^2$ across all layers, and the thermal velocity is set to 10^7 cm/s .

The four primary output parameters of a solar cell are power conversion efficiency (PCE), fill factor (FF), short circuit current (Jsc), and open circuit voltage (Voc). The necessary material parameters for simulation and absorption spectra have been compiled from the literature [16,17, 21–35]. A comparison analysis of the simulated and experimental data for SCAPS 1-D calibration is presented in Table 2. Additionally, Figure 2 illustrates the simulated polymer solar cell structure with additional P3HT layer.

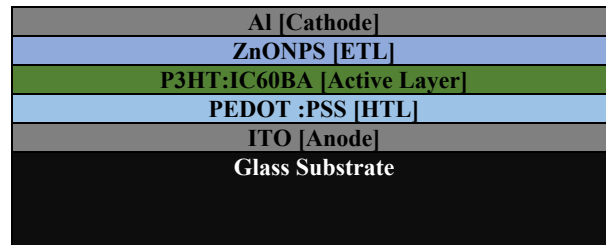


Figure 1. Simulated Bulk Heterojunction Polymer Solar Cell Structure for Standardization of SCAPS 1D

Table 1. Simulation parameters of Bulk Heterojunction Polymer Solar Cell

Parameters	HTL	Active Layer	ETL
Thickness [nm]	27	250	46
E _g (eV)	1.6	1.68	3.2
χ (eV)	3.4	3.880	4.6
ε _r	3.0	3.3	9.0
μ _n cm ² /vs	10	10 ⁷	10 ⁷
μ _p cm ² /vs	400	10 ⁷	10 ⁷
N _A (cm ⁻³)	10 ¹⁵	0	0
N _D (cm ⁻³)	0	3.2×10 ¹⁸	1.1×10 ¹⁸
N _C (cm ⁻³)	2.2×10 ¹⁵	1.6×10 ²⁰	2.2×10 ¹⁹
N _V (cm ⁻³)	1.8×10 ¹⁸	10 ¹⁹	1.8×10 ¹⁹
N _t (cm ⁻³)	10 ¹⁵	10 ¹⁶	10 ¹⁵

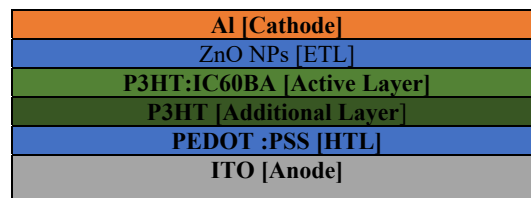


Figure 2. Simulated Polymer Solar Cell Structure

Table 2. Comparison of simulated parameters with experimental results

Parameters	Experimental	Simulated
Voc (V)	0.84	0.85
Jsc(mA/cm ²)	8.36	8.21
FF(%)	59	57
PCE(%)	4.1	3.97

3. RESULTS AND DISCUSSION

We will analyze the solar cell structure shown in Figure 2 in greater detail. Each layer of the structure is examined to determine its performance under various parameters for optimal results. To achieve these optimal outcomes, new device structures are developed and their performance is thoroughly investigated. The following section provides an explanation of the simulation studies that were conducted.

3.1 Effect of Active Layer thickness

The thickness of the active layer is varied between 100 and 300 nm to assess the characteristics related to solar cell performance. The parameters Voc, Jsc, FF, and PCE demonstrate similar trends, with all reaching their optimal values at a thickness of 300 nm. These improvements at the ideal thickness can be attributed to reduced recombination and enhanced light absorption. Increasing the active layer thickness may enhance Jsc by allowing more light to be absorbed. However, this relationship is nonlinear; beyond an optimal thickness, it is affected by increased charge recombination and decreased charge transport efficiency, leading to diminishing returns [36, 37]. While Voc is not directly influenced by the thickness of the active layer, it is primarily determined by the energy levels of the donor and acceptor materials.

Nevertheless, recombination dynamics and the quality of the interface may play significant roles in its behavior [38, 39]. Certain experiments have demonstrated that increasing the thickness of the active layer can improve the power conversion efficiency (PCE) to a certain extent. The relevant graph is presented in Figure 3.

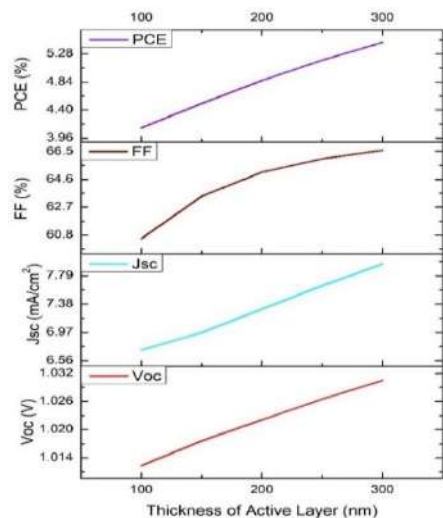


Figure 3. Influence of Active Layer Thickness

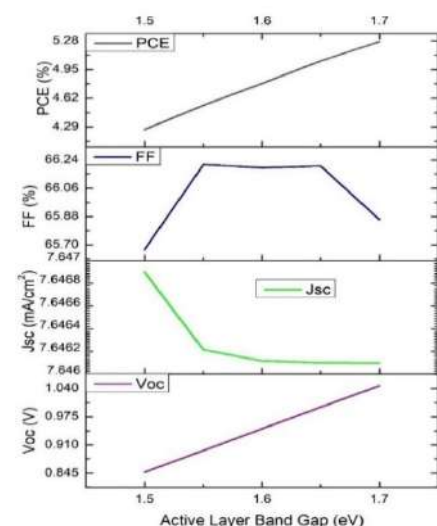


Figure 4. Influence of Active Layer Bandgap

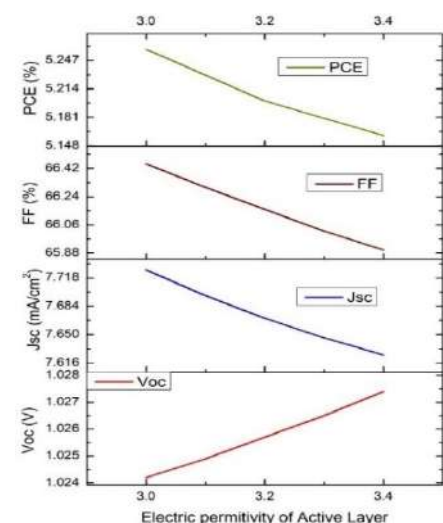


Figure 5. Influence of Electric Permittivity of Active Layer

3.2 Influence of Active Layer Band Gap

The active layer band gap is varied from 1.5 eV to 1.7 eV, and both power conversion efficiency (PCE) and open-circuit voltage (Voc) exhibit similar variations with bandgap. Both PCE and Voc reach their maximum at 1.7 eV. In organic solar cells, Voc is determined by the difference between the lowest unoccupied molecular orbital (LUMO) of the acceptor material and the highest occupied molecular orbital (HOMO) of the donor material. As the bandgap of the active layer increases, the Voc tends to increase as well, reflecting either a higher LUMO level of the donor or a lower HOMO level of the acceptor. This relationship is influenced by the energy levels of the donor and acceptor materials, which impact Voc in connection with the difference between the quasi-Fermi levels of the electrons and holes. A larger bandgap reduces the energy loss between the bandgap energy (E_g) and Voc. However, in organic solar cells, the Voc is generally lower than the bandgap energy due to non-radiative recombination and other losses. As the bandgap increases, Voc approaches E_g , resulting in an overall increase in Voc. Furthermore, the power conversion efficiency of a solar cell is calculated by multiplying its fill factor (FF), open-circuit voltage (Voc), and short-circuit current (Jsc).

As mentioned earlier, an increase in the bandgap can lead to a higher open-circuit voltage (Voc), which may subsequently enhance the power conversion efficiency (PCE). However, a wider bandgap often results in reduced solar spectrum absorption by the solar cell, particularly in the infrared region, which can lower the short-circuit current (Jsc). Nevertheless, in some cases, the increase in Voc associated with a wider bandgap can compensate for the decrease in Jsc, resulting in an overall increase in PCE. Additionally, the fill factor (FF) may improve with an increasing bandgap due to enhanced charge carrier transfer [40], although this trend is not always consistent. With further increases in bandgap, FF may remain stable or even decline, possibly due to the effects of recombination processes and exciton lifetimes [41, 42]. The relationship between FF and bandgap is complex and depends on the specific materials and cell architecture used in the organic solar cell. The relevant graph illustrating these dynamics is presented in Figure 4.

3.3 Effect of Electric Permittivity of Active Layer

In this section, we have analyzed the impact of the active layer's electric permittivity [ϵ] on the performance of the polymer solar cell. The electric permittivity is varied from 3.0 to 3.4. The findings indicate that while the open-circuit voltage (Voc) increases with ϵ , all other parameters are found to decrease. The electric permittivity of the active layer influences the dielectric properties of the material, which in turn affects the open-circuit voltage of polymer solar cells (PSCs). Consequently, a higher electric permittivity in the active layer may lead to an increase in Voc due to its effect on the dynamics of charge carriers within the solar cell.

A higher electric permittivity enhances charge carrier separation and reduces recombination losses by decreasing the Coulombic interaction between charge carriers [43]. This improvement can result in a stronger built-in electric field and more efficient charge extraction, both of which contribute to an increased open-circuit voltage (Voc). However, optimizing the performance of polymer solar cells (PSCs) requires a comprehensive understanding

of all contributing parameters, as a higher permittivity can indeed enhance Voc. The electric permittivity of the active layer also influences the short-circuit current (Jsc), fill factor (FF), and power conversion efficiency (PCE) of PSCs by affecting the dissociation of photogenerated electron-hole pairs and the movement of charge carriers within the device. Elevated Coulombic attraction between opposing charges can occur with high electric permittivity, potentially hindering their separation and reducing Jsc [44]. Additionally, a larger permittivity may negatively impact charge carrier mobility, leading to increased recombination rates and declines in both FF and PCE. The relevant graph illustrating these relationships is presented in Figure 5.

3.4: Effect of Conduction Band Density of States of Active Layer

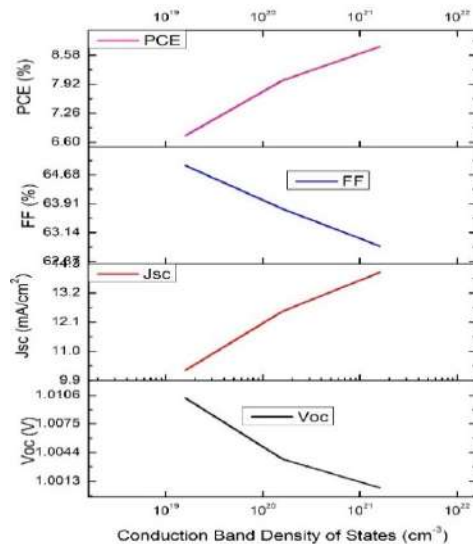


Figure 6. Influence of CB density of states of Active Layer

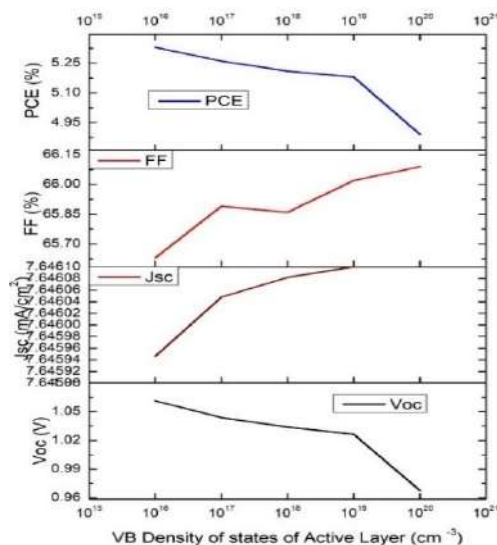


Figure 7. Influence of VB density of states of Active Layer

seem beneficial, it generates more energy states that facilitate non-radiative recombination processes and hinder charge transport, which adversely affects both PCE and Voc [47-48]. Figure 7 shows the respective graph.

3.6. Effect of Shallow Uniform Density of States of Active Layer

The shallow uniform density of states in the active layer experiences a significant shift from 3.2×10^{14} to $3.2 \times 10^{16} \text{ cm}^{-3}$. As this density increases, parameters such as open-circuit voltage (Voc), fill factor (FF), and power conversion efficiency (PCE) tend to improve, while the short-circuit current (Jsc) shows a decline. An elevated donor density can enhance the intrinsic potential across the junction by increasing the availability of charge carriers. This rise in potential positively impacts Voc, which is crucial for the efficient operation of solar cells, as it reduces losses during charge separation. Furthermore, an increase in donor density leads to enhanced conductivity within the active layer, which

In this section, we examine how varying the conduction band density of states in the active layer from 1.6×10^{19} to $1.6 \times 10^{21} \text{ cm}^{-3}$ affects the performance metrics of polymer solar cells. Figure 6 illustrates the corresponding data. The graph indicates that while short-circuit current (Jsc) and power conversion efficiency (PCE) fluctuate similarly, the open-circuit voltage (Voc) and fill factor (FF) do not exhibit the same trend. The decline in Voc with an increase in the conduction band density of states in the active layer is primarily attributed to enhanced recombination losses and shifts in the Fermi level.

Recombination events are more likely to occur when the density of states is higher, as this provides more energy levels for charge carriers to occupy [45]. The increase in short-circuit current density (Jsc) associated with a higher conduction band density of states (DOS) in the active layer can be attributed to increased charge carrier generation and improved collection efficiency. A greater DOS enhances the availability of conduction states, leading to more efficient photon absorption and, consequently, a higher production of electron-hole pairs. This directly correlates with an increase in Jsc [46]. However, the rise in power conversion efficiency (PCE) and the decline in fill factor (FF) of polymer solar cells with an increasing conduction band DOS result from a combination of enhanced charge generation and increased recombination losses. While a higher DOS contributes to improved charge carrier absorption and generation, it also results in more opportunities for recombination, which negatively impacts the FF [45].

3.5 Effect of Valence Band Density of States of Active Layer

The valence band density of states in the active layer is varied from 1×10^{16} to $1 \times 10^{20} \text{ cm}^{-3}$, and its impact on the characteristics of polymer solar cells is evaluated. The analysis reveals that while short-circuit current density (Jsc) and fill factor (FF) change in a similar manner, open-circuit voltage (Voc) and power conversion efficiency (PCE) do not exhibit the same trend. The decline in PCE and Voc with an increase in valence band density of states (DOS) is primarily due to enhanced recombination rates and increased charge carrier trapping. Although a higher valence band DOS might initially

in turn lowers series resistance. This reduction in series resistance facilitates a higher fill factor by minimizing voltage drops during operation [49]. The graphical representation is given in Figure 8. However, a noticeable increase in shallow donor density can also result in a higher defect density within the active layer. This heightened defect density may give rise to additional recombination centers that foster nonradiative recombination of charge carriers. Consequently, this adverse effect results in fewer charge carriers contributing to the photocurrent, thereby reducing the short-circuit current [50].

3.7. Effect of Thickness of ETL

This study examines the effect of varying the thickness of the electron transport layer (ETL) made from zinc oxide nanoparticles (ZnONPs), changing it from 40 nm to 48 nm. The optimal performance was identified at a thickness of 48 nm, where the power conversion efficiency (PCE) peaked at 5.22%, the open circuit voltage (Voc) reached 1.0270 V, and the short-circuit current density (Jsc) was measured at 7.699979 mA/cm². At 40 nm thickness, the fill factor (FF) was observed to achieve a higher value of 66.09%. The analysis of output parameters indicates that as the thickness increases, the fill factor tends to decrease, while Voc, Jsc, and PCE show an upward trend. Thicker ETLs can enhance the open circuit voltage (Voc) in polymer solar cells by slowing down charge recombination processes. By expanding the electron transport pathway, a thicker ETL minimizes the likelihood of electron-hole recombination before charge carriers reach the electrodes. This increased thickness facilitates a more effective collection of photo-generated electrons due to the greater transit space afforded to the electrons. Consequently, as more charge carriers efficiently reach the electrodes with minimal losses from trapping or recombination, this improvement in charge transport can lead to higher Jsc [51]. However, the decline in the fill factor in polymer solar cells at greater ETL thicknesses is primarily attributed to increased series resistance, reduced charge carrier mobility, elevated recombination rates, and interfacial charge transfer resistance [52]. The power conversion efficiency (PCE) of polymer solar cells can improve with increased thickness of the ETL, up to a certain optimal point, largely due to better light absorption, enhanced charge extraction, and superior charge transport capabilities [53]. The respective graph is shown in Figure 9.

3.8 Effect of Electric Permittivity of ETL

This section provides a comprehensive analysis of how the electric permittivity of the electron transport layer (ETL) affects polymer solar cells, as illustrated in Figure 10. The findings reveal that changing the electric permittivity from 8.2 to 9.0 results in a decrease in the open-circuit voltage (Voc), while both the short-circuit current density (Jsc) and overall efficiency experience an increase. Specifically, a permittivity value of 9.0 yields the best optimized performance. For effective charge transfer, the energy levels of the ETL and the active layer must be closely aligned. Higher permittivity in the ETL may disrupt this alignment at the ETL/polymer interface, leading to voltage loss and a reduction in Voc due to ineffective charge transfer. Quality issues within the ETL can further diminish Voc and impede charge transfer, even if the permittivity is elevated. Since successful charge collection relies on the interactions between charge carriers and their respective layers, any adverse effects on the electrostatic potential exacerbated by materials with high dielectric constants [54] can contribute to a lower Voc. Conversely, increased electric permittivity facilitates faster charge transfer at the donor-acceptor interface, significantly reducing non-geminate recombination losses. By minimizing these losses, more charge carriers become available for collection, enhancing the overall performance of the solar cell.

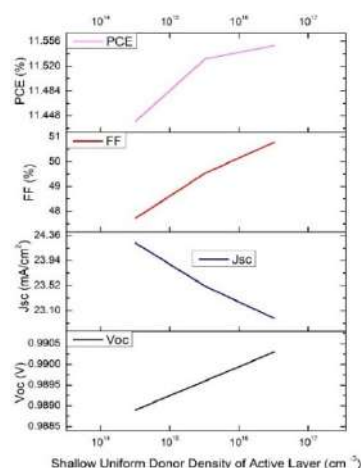


Figure 8. Influence of Shallow Uniform Donor Density of states of Active Layer

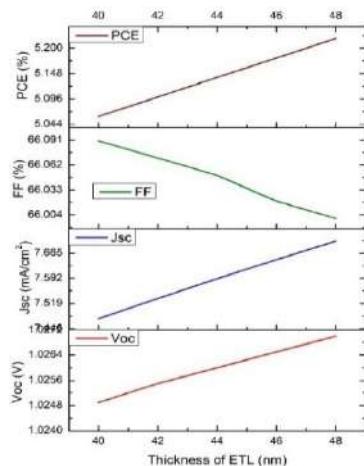


Figure 9. Influence of ETL Thickness

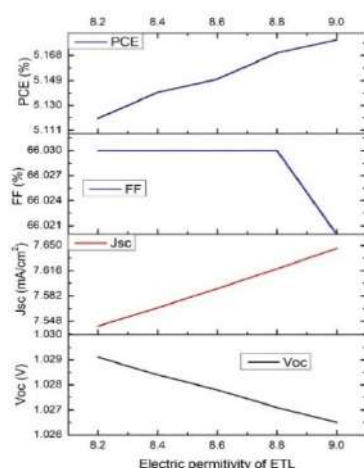


Figure 10. Effect of Electric Permittivity of ETL

Ultimately, improved charge dynamics and reduced recombination lead to higher values of Jsc and power conversion efficiency (PCE), which can be attributed to a favourable dielectric environment [55]. However, flaws or suboptimal interfaces can still generate recombination losses, which may diminish the fill factor's responsiveness to changes in permittivity.

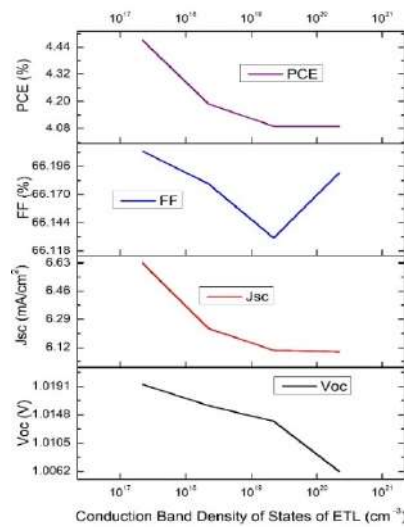


Figure 11. Influence of Conduction Band Density of States of ETL

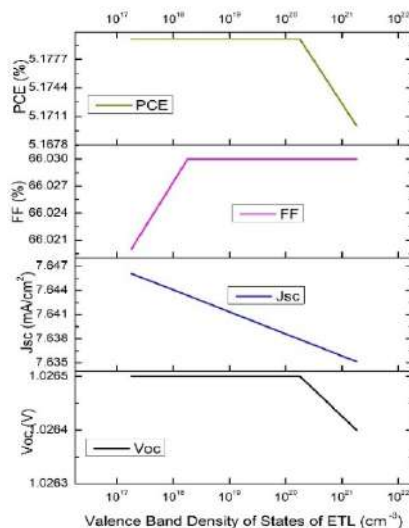


Figure 12. Influence of Valence Band Density of States of ETL

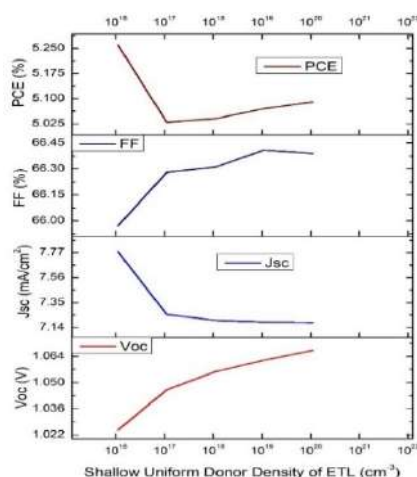


Figure 13. Influence of Shallow Donor Density of ETL

3.9 Effect of Conduction Band Density of States of ETL

This section investigates the influence of the conduction band density of states (CBDoS) of a polymer cell on the performance of its electron transport layer (ETL). For this analysis, the CBDoS is varied from 2.2×10^{17} to $2.2 \times 10^{20} \text{ cm}^{-3}$. It was observed that the short-circuit current density (Jsc) and power conversion efficiency (PCE) exhibited similar trends, while the fill factor (FF) initially decreased before increasing at higher CBDoS values. The performance was found to be optimized at $2.2 \times 10^{17} \text{ cm}^{-3}$, as illustrated in Figure 11. The decline in Jsc and PCE with increasing CBDoS is primarily attributed to increased carrier trapping, higher recombination rates, and potential misalignment of energy levels [56]. As the CBDoS rises, the presence of additional trap states hinders charge extraction and facilitates enhanced carrier recombination, leading to a reduction in open-circuit voltage (Voc). The behaviour of the fill factor (FF) is influenced by the complex interactions among energy level alignment, trap states, and carrier dynamics. This results in a non-linear relationship, where the competing effects of increased recombination and improved carrier mobility contribute to the observed patterns in FF.

3.10 Effect of Valence Band Density of States of ETL

This section focuses on the changes in the valence band density of states of the electron transport layer in polymer solar cells, varying from 1.8×10^{17} to $1.8 \times 10^{21} \text{ cm}^{-3}$. Figure 12 provides a graphical representation of these changes.

It was found that the optimal performance, characterized by a maximum efficiency of 5.18%, occurs at a density of $1.8 \times 10^{17} \text{ cm}^{-3}$. Variations in the valence band density of states within the electron transport layer can lead to complex changes in the output characteristics of polymer solar cells. The effects on short-circuit current density (Jsc), open-circuit voltage (Voc), fill factor (FF), and power conversion efficiency (PCE) are determined by optimizing the density of states, alongside the ratio of charge transport to recombination and the alignment of interfacial energy levels. The respective graph is given in Figure 12.

3.11 Effect of Shallow Donor Density of ETL

This section analyzes the fluctuations in the shallow donor density of the electron transport layer, which ranges from 1.1×10^{16} to $1.1 \times 10^{20} \text{ cm}^{-3}$. Figure 13 provides the corresponding graph illustrating these fluctuations. The findings indicate that while the variations in short-circuit current density (Jsc) and power conversion efficiency (PCE) are comparable, the changes in open-circuit voltage (Voc) and fill factor (FF) exhibit a similar trend. Voc typically increases with a higher shallow donor density, as this leads to improved charge extraction and reduced recombination losses. Additionally, as charge transport and extraction enhance, the fill factor (FF) also rises with the shallow donor density. However, excessive concentrations of shallow donors can lead to significant recombination, ultimately resulting in a decrease in PCE.

3.12 Effect of Thickness of Additional P3HT Layer

In this case, the thickness of the P3HT layer is varied from 16 nm to 24 nm, with the optimal performance observed at 16 nm.

This is illustrated in Figure 14, which provides a graphical representation of the findings. The investigation reveals that while the fill factor (FF) shows a different trend, the open-circuit voltage (Voc), short-circuit current density (Jsc), and power conversion efficiency (PCE) fluctuate similarly. As the P3HT layer increases in thickness, the pathways for charge transport become longer and more convoluted.

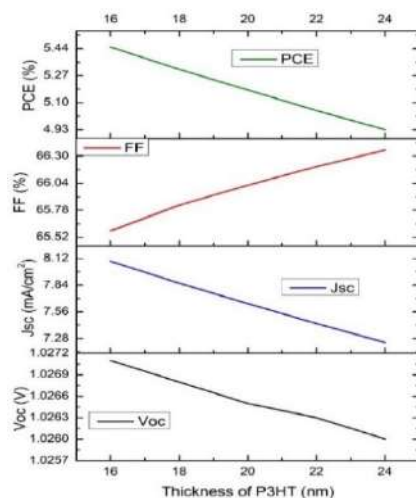


Figure 14. Influence of P3HT Thickness

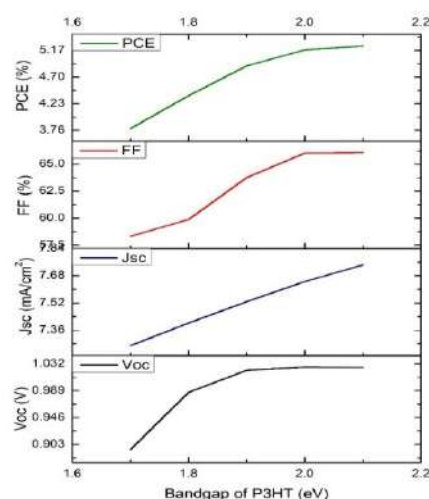


Figure 15. Effect of Band gap of P3HT

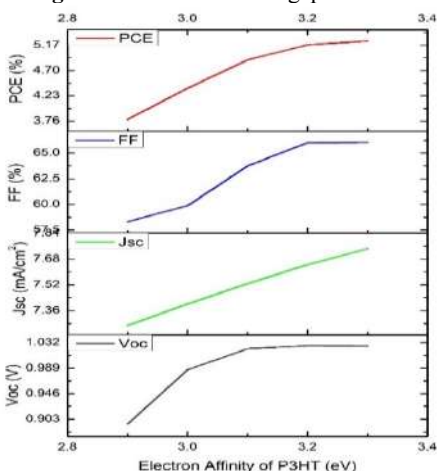


Figure 16. Influence of Electron Affinity of P3HT

declines due to higher defect densities and shorter diffusion lengths, ultimately reducing the overall efficiency of the solar cell. The maximum efficiency is observed at a valence band density of $1.8 \times 10^{17} \text{ cm}^{-3}$. The decrease in Jsc for polymer solar

Consequently, charge carrier mobility diminishes, which is crucial for the effective transfer of electrons and holes to their respective electrodes. This increase in the distance that charge carriers must traverse can lead to a higher likelihood of recombination before reaching the electrodes, potentially resulting in reduced Jsc and PCE. Moreover, the drop in Voc may be attributed to lower charge extraction efficiency in thicker layers and higher energy losses associated with elevated recombination rates. Key factors contributing to the decline in FF in polymer solar cells (PSCs) as the P3HT layer thickness increases include enhanced bimolecular recombination, inefficient charge transport, and imbalanced charge carrier mobilities [57].

3.13. Effect of Bandgap of Additional P3HT Layer

The bandgap of P3HT varies from 1.7 eV to 2.1 eV, with observations indicating that all output parameters increase alongside the bandgap. This trend is illustrated in Figure 15. There is a direct correlation between the open-circuit voltage (Voc) and the energy levels of the polymer's lowest unoccupied molecular orbital (LUMO) and highest occupied molecular orbital (HOMO). Generally, a larger bandgap ensures a higher HOMO energy level, which can enhance Voc. Additionally, higher bandgap materials are more effective at absorbing high-energy photons, potentially leading to increased short-circuit current density (Jsc). Increasing the bandgap creates a larger energy difference necessary for charge separation, which reduces the likelihood of exciton recombination. This mechanism ultimately contributes to an improvement in both fill factor (FF) and power conversion efficiency (PCE).

3.14 Effect of Electron Affinity of Additional P3HT Layer

This section investigates the influence of P3HT's electron affinity on the performance of polymer solar cells. The electron affinity is varied from 2.9 eV to 3.3 eV, revealing that all output parameters of the cell respond similarly to these changes. Figure 16 illustrates this correlation. In general, a higher electron affinity results in an elevated LUMO energy level, which can lead to an increase in the open-circuit voltage (Voc) when no current is flowing. Larger electron affinities create greater energy offsets, facilitating the efficient movement of electrons and potentially enhancing the short-circuit current density (Jsc). Furthermore, a polymer donor with a higher electron affinity can reduce recombination losses, leading to improved fill factor (FF) and power conversion efficiency (PCE) [58].

3.15 Effect of Valence Band Density of States of Additional P3HT Layer

The present study investigates the effect of varying the valence band density of states of P3HT from 1.8×10^{16} to $1.8 \times 10^{20} \text{ cm}^{-3}$ on the output parameters of polymer solar cells. While the short-circuit current density (Jsc) shows a differential response among the output parameters, the open-circuit voltage (Voc), fill factor (FF), and power conversion efficiency (PCE) exhibit similar trends. This relationship is illustrated in Figure 17.

Improvements in metrics such as PCE, FF, and Voc can be attributed to better energy level alignment and enhanced charge transport. However, as the valence band density increases, performance declines due to higher defect densities and shorter diffusion lengths, ultimately reducing the overall efficiency of the solar cell. The maximum efficiency is observed at a valence band density of $1.8 \times 10^{17} \text{ cm}^{-3}$. The decrease in Jsc for polymer solar

cells, particularly those utilizing P3HT, is primarily caused by increased defect density and shorter charge carrier diffusion lengths associated with high density states.

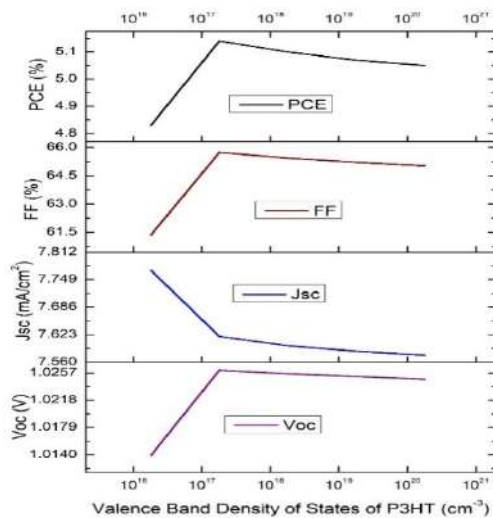


Figure 17. Influence of Valence Band Density of States of P3HT

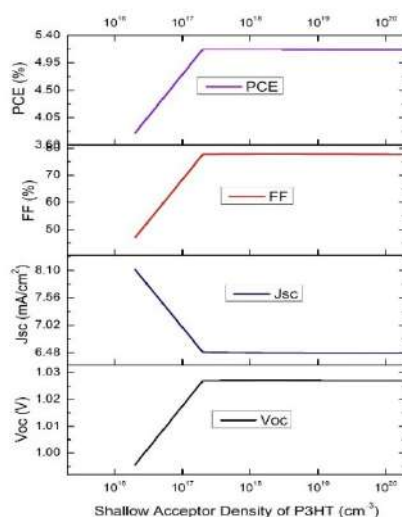


Figure 18. Influence of Shallow Acceptor Density of States of P3HT

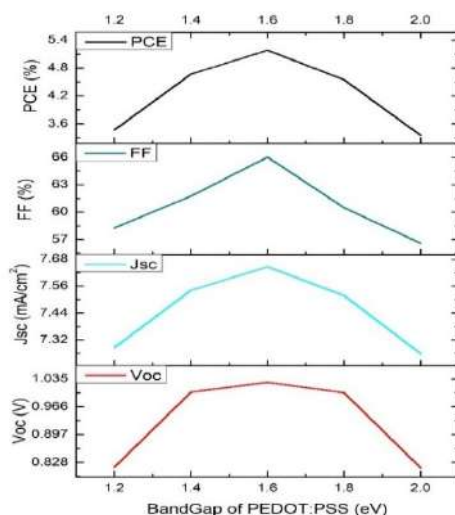


Figure 19. Influence of Bandgap of PEDOT : PSS

3.16 Effect of Shallow Acceptor Density of States of Additional P3HT Layer

In our subsequent endeavor, we adjusted the shallow acceptor density of states in the P3HT layer, conducting a study that encompassed values ranging from 2×10^{16} to $2 \times 10^{20} \text{ cm}^{-3}$. The optimal performance was observed at a density of $2 \times 10^{17} \text{ cm}^{-3}$, as illustrated in Figure 18, which displays the graphical variations. The fluctuations in performance were similar to those seen with the layer's valence band density of states. This similarity arises because both factors significantly influence the solar cell's charge transport, carrier density, trap formation, and recombination dynamics. The sensitivity of performance to these concentrations highlights the delicate balance required to enhance charge mobility while minimizing the adverse effects of defects.

3.17 Effect of Bandgap of PEDOT: PSS [HTL]

This study investigates the impact of changing the bandgap of the hole transport layer, PEDOT:PSS, from 1.2 eV to 2.0 eV on the output parameters of polymer solar cells. All parameters were observed to fluctuate similarly, with maximum efficiency achieved at a bandgap of 1.6 eV, as illustrated in Figure 19. Increasing the PEDOT:PSS bandgap can enhance the open-circuit voltage (Voc) in polymer solar cells. However, a higher bandgap limits sunlight absorption, resulting in lower charge carrier production. Due to the significantly high bandgap of PEDOT:PSS, sunlight absorption is reduced, which subsequently decreases the short-circuit current density (Jsc). Additionally, the hydrophilic nature and instability of the hole transport layer (HTL) may lead to increased series resistance, further affecting the fill factor (FF). The power conversion efficiency (PCE) increases initially but subsequently declines as the hole transport bandgap is raised.

3.18 Effect Electron Affinity of PEDOT:PSS [HTL]

PEDOT:PSS demonstrates a shift in electron affinity ranging from 3.0 eV to 3.8 eV, with optimized values achieved at 3.4 eV, as illustrated in Figure 20. The output parameters of polymer solar cells exhibit similar changes as the bandgap and electron affinity of PEDOT:PSS are increased. This similarity arises from the interplay between energy level alignment and charge transport dynamics, both of which are simultaneously influenced by these two characteristics.

CONCLUSION

In this modeling work, a P3HT layer was integrated into a polymer solar cell structure using SCAPS 1-D software, with P3HT:IC₆₀BA serving as the active layer. The study revealed that the structure exhibited significantly better performance with the inclusion of P3HT. The P3HT layer enhances charge transit and maximizes Voc and Jsc by aligning energy levels at the heterojunction surfaces. After optimization, the following output parameters were achieved: power conversion efficiency (PCE) = 15.26%, fill factor

(FF) = 51.62%, short-circuit current density (J_{sc}) = 26.577140 mA/cm², and open-circuit voltage (V_{oc}) = 1.1126 V. Before optimization, we recorded an efficiency of only 5.18%. These results are quite promising and will contribute to the advancement of photovoltaic research in the future.

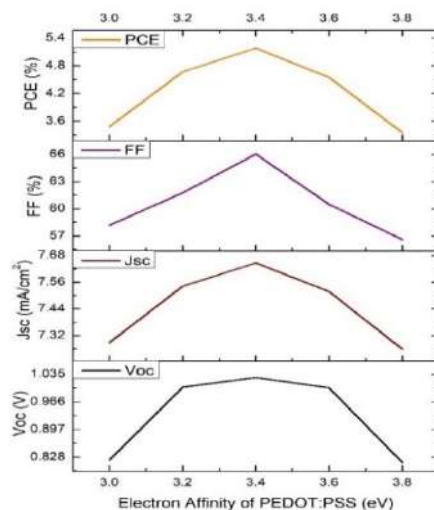


Figure 20. Influence of Electron Affinity of PEDOT: PSS

Statements and Declarations:

Data availability statement: The data that support the findings of this study are available on request from the corresponding author.

Acknowledgements. For providing SCAPS 1-D software for the simulation investigation, the authors would like to thank Pr.M. Burgelman of the University of Gent in Belgium.

CRedit authorship contribution statement.: Ramabadran C.D.: Conceptualization, Data curation, Methodology, Software, Investigation, Validation, Formal analysis, Writing – original draft. Sudheer K.S.: Supervision, Conceptualization, Software, Writing – review & editing

Conflict of interest. The authors declare that they have no conflict of interest.

ORCID

Chittur Devarajan Ramabadran, <https://orcid.org/0009-0003-2255-3087>; K. Sebastian Sudheer, <https://orcid.org/0000-0002-9019-4405>

REFERENCES

- [1] Y. He, G. Zhao, B. Peng, and Y. Li, "High-Performance Polymer Solar Cells with P3HT:PCBM Bulk Heterojunction: A Combined Experimental and Simulation Study," *Advanced Functional Materials*, **20**(19), 3383–3389 (2010). <https://doi.org/10.1002/adfm.201000973>
- [2] C. Bendenia, H. Merad-Dib, S. Bendenia, and B. Hadri, "Numerical simulation of CIGS solar cells with SCAPS-1D: Optimization of the absorber band gap grading and thickness," *Optik*, **174**, 167–172 (2018). <https://doi.org/10.1016/j.ijleo.2018.08.058>
- [3] A. Hazra, I. Mal, D. Samajdar, and T. Das, "Device simulation of lead-free CH₃NH₃SnI₃ perovskite solar cells with high efficiency," *Optik*, **168**, 747–753 (2018). <https://doi.org/10.1016/j.ijleo.2018.04.117>
- [4] Y. Sun, C. Cui, H. Wang, and Y. Li, "Fabrication of Flexible, Free-Standing, Ultralight Carbon Nanotube Aerogel Films for Supercapacitor Electrodes," *Advanced Energy Materials*, **1**(6), 1058–1061(2011). <https://doi.org/10.1002/aenm.201100322>
- [5] Y. He, H.-Y. Chen, J. Hou, and Y. Li, "Indene-C60 Bisadduct: A New Acceptor for High-Performance Polymer Solar Cells," *Journal of the American Chemical Society (JACS)*, **132**(4), 1377–1382 (2010). <https://doi.org/10.1021/ja908602j>
- [6] G. Zhao, Y. He, and Y. Li, "6.5% Efficiency of Polymer Solar Cells Based on poly(3-hexylthiophene) and Indene-C60 Bisadduct by Device Optimization," *Advanced Materials*, **22**(39), 4355–4358 (2010). <https://doi.org/10.1002/adma.201001339>
- [7] Z. Tan, D. Qian, W. Zhang, L. Li, Y. Ding, Q. Xu, F. Wang, and Y. Li, "Efficient and stable polymer solar cells with a solution-processable small molecule as anode buffer layer," *Journal of Materials Chemistry A*, **1**(3), 657–664 (2013). <https://doi.org/10.1039/C2TA00575F>
- [8] S. Gartner, M. Christmann, S. Sankaran, H. Röhm, E.-M. Prinz, F. Pentz, A. Pütz, *et al.*, "Design Rules for High-Efficiency Polymer Solar Cells with Low Energy Loss and High Fill Factor," *Advanced materials*, **26**(38), 6653–6657 (2014). <https://doi.org/10.1002/adma.201402932>
- [9] E.F. Oliveira, L.C. Silva, and F.C. Lavarda, "Electronic structure and charge transport properties of star-shaped molecules with 1,3,5-triazine core and thiophene arms for photovoltaic applications," *Structural Chemistry*, **28**, 1133–1140 (2017). <https://doi.org/10.1007/s11224-017-0926-y>
- [10] G. Yu, J. Gao, J.C. Hummelen, F. Wudl, and A.J. Heeger, "Polymer Photovoltaic Cells: Enhanced Efficiencies via a Network of Internal Donor-Acceptor Heterojunctions," *Science*, **270**, 1789–1791 (1995). <https://doi.org/10.1126/science.270.5243.1789>
- [11] J. Peet, J.Y. Kim, N.E. Coates, W.L. Ma, D. Moses, A.J. Heeger, and G.C. Bazan, "Efficiency enhancement in low-bandgap polymer solar cells by processing with alkane dithiols," *Nature materials*, **6**(7), 497–500 (2007). <https://doi.org/10.1038/nmat1928>
- [12] J.K. Lee, W.L. Ma, C.J. Brabec, J. Yuen, J.S. Moon, J.Y. Kim, K. Lee, *et al.*, "Processing additives for improved efficiency from bulk heterojunction solar cells," *Journal of the American Chemical Society*, **130**(11), 3619–3623 (2008). <https://doi.org/10.1021/ja710079w>

- [13] N. Loew, S. Komatsu, H. Akita, K. Funayama, T. Yuge, T. Fujiwara, and M. Ihara, "Development of a novel glucose sensor using engineered glucose dehydrogenase," *ECS Transactions*, **58**(45), 77- 82 (2014). <https://doi.org/10.1149/05845.0077ecst>
- [14] P. Matavulj, M.K. Islam, and C.S. Živanović, "Numerical simulation of organic solar cells: Impact of interface layers on device performance," in: *Proceedings of the International Conference on Numerical Simulation of Optoelectronic Devices (NUSOD)*, (IEEE, Copenhagen, Denmark, 2017), pp. 133–134. <https://doi.org/10.1109/NUSOD.2017.8010026>
- [15] S.-A. Gopalan, A.-I. Gopalan, A. Vinu, K.-P. Lee, and S.-W. Kang, "A new strategy for designing high-efficiency cascaded solar cells: Optical and electrical modeling using SCAPS-1D," *Solar Energy Materials and Solar Cells*, **174**, 112-123 (2018). <https://doi.org/10.1016/j.solmat.2017.08.033>
- [16] M. Burgelman, P. Nollet, and S. Degraeve, "Modelling polycrystalline semiconductor solar cells," *Thin Solid Films*, **361-362**, 527-532 (2000). [https://doi.org/10.1016/S0040-6090\(99\)00825-1](https://doi.org/10.1016/S0040-6090(99)00825-1)
- [17] B. Xu, G. Sai-Anand, A.-I. Gopalan, Q. Qiao, and S.-W. Kang, "Improving Photovoltaic Properties of P3HT:IC₆₀BA through the Incorporation of Small Molecules," *Polymers*, **10**(2), 121 (2018). <https://doi.org/10.3390/polym10020121>
- [18] S. Hosseini, M. Bahramgour, N. Delibaş, and A. Niaie, "Investigation of a Perovskite Solar Cell and Various Parameters Impact on Its Layers and the Effect of Interface Modification by Using P3HT as an Ultrathin Polymeric Layer Through SCAPS-1D Simulation," *Sakarya University Journal of Science*, **25**(5), 1168-1179 (2021). <https://doi.org/10.16984/aufenbilder.947735>
- [19] A. Maillard and A. Rochefort, "Quantum transport properties of graphene nanoribbons with embedded heptagon-pentagon defects," *Organic Electronics*, **15**(9), 2091-2098 (2014). <https://doi.org/10.1016/j.orgel.2014.05.028>
- [20] H. Lee, (2015). "Optimization of energy level alignment for efficient organic photovoltaics," *Vacuum Magazine*, **2**(2), 12-16 (2015). <https://doi.org/10.5757/VACMAG.2.2.12> (in Korean)
- [21] S. Holliday, *et al.*, "High-efficiency and air-stable P3HT solar cells with a new non-fullerene acceptor," *Nature Communications*, **7**, 11585 (2016). <https://doi.org/10.1038/ncomms11585>
- [22] A. Kumar, and R. Singh, "Numerical Modelling Analysis for Carrier Concentration Level Optimization of CdTe Heterojunction Thin Film-Based Solar Cell with Different Non-Toxic Metal Chalcogenide Buffer Layers Replacements: Using SCAPS-1D Software," *Crystals*, **11**(12), 1454 (2021). <https://doi.org/10.3390/cryst11121454>
- [23] K. Nithya, and K. Sudheer, "Numerical modelling of non-fullerene organic solar cell with high dielectric constant ITIC-OE acceptor," *Journal of Physics Communications*, **4**(2), 025012 (2020). <https://doi.org/10.1088/2399-6528/ab772a>
- [24] S.W. Heo, E.J. Lee, K.W. Seong, and D.K. Moon, "Enhanced stability in polymer solar cells by controlling the electrode work function via modification of indium tin oxide," *Solar energy materials and solar cells*, **115**, 123–128, (2013). <https://doi.org/10.1016/j.solmat.2013.03.023>
- [25] S. Loser, C.J. Bruns, H. Miyauchi, R.P. Ortiz, A. Facchetti, S.I. Stupp, and T.J. Marks, "A Naphthodithiophene-Diketopyrrolopyrrole Donor Molecule for Efficient Solution-Processed Solar Cells," *Journal of the American Chemical Society*, **133**(21), 8142– 8145 (2011). <https://doi.org/10.1021/ja202791n>
- [26] G. Li, Y. Yao, H. Yang, V. Shrotriya, G. Yang, and Y. Yang, "Solvent annealing' effect in polymer solar cells based on poly(3-hexylthiophene) and methanofullerenes," *Advanced Functional Materials*, **17**(10), 1636–1644 (2007). <https://doi.org/10.1002/adfm.200600624>
- [27] Z. He, F. Liu, C. Wang, J. Chen, L. He, D. Nordlund, H. Wu, T.P. Russell, and Y. Cao, "Simultaneous Enhancement of Open-Circuit Voltage, Short-Circuit Current Density, and Fill Factor in Polymer Solar Cells," *Materials Horizons*, **2**(6), 592–597(2015). <https://doi.org/10.1039/C5MH00164A>
- [28] X. Zhu, K. Lu, B. Xia, J. Fang, Y. Zhao, T. Zhao, Z. Wei, *et al.*, "Improving the Performances of Random Copolymer Based Organic Photovoltaics by Incorporating Fluorine Substituents," *Polymers*, **8**(1), 4 (2015). <https://doi.org/10.3390/polym8010004>
- [29] L. Lu, *et al.*, "High-performance ternary blend polymer solar cells involving both energy transfer and hole relay processes," *Nat. Commun.* **6**, 7327 (2015). <https://doi.org/10.1038/ncomms8327>
- [30] M.M. Stylianakis, D. Konios, C. Petridis, G. Kakavelakis, E. Stratakis, and E. Kymakis, "Efficient and stable hybrid perovskite-graphene solar cells via interfacial passivation with 2D–2D heterostructures," *2D Materials*, **4**(4), 042005 (2017). <https://doi.org/10.1088/2053-1583/aa87b6>
- [31] B.N. Ezealigo, A.C. Nwanya, A. Simo, R.U. Osuji, R. Bucher, M. Maaza, and F.I. Ezema, "Optical and electrochemical capacitive properties of copper (I) iodide thin film deposited by SILAR method," *Arabian Journal of Chemistry*, **12**(8), 5380–5391 (2019). <https://doi.org/10.1016/j.arabjc.2017.01.008>
- [32] J. Liu, Y. Zhang, C. Liu, M. Peng, A. Yu, J. Kou, W. Liu, *et al.*, "High-Performance UV Photodetector Based on a Heterojunction of a ZnO Nanowire Array and a Few-Layer MoS₂ Film," *Nanoscale research letters*, **11**, 1–7 (2016). <https://doi.org/10.1186/s11671-016-1291-2>
- [33] Z. El Jouad, M. Morsli, G. Louarn, L. Cattin, M. Addou, and J.C. Bernède, "Improving the efficiency of subphthalocyanine based planar organic solar cells through the use of MoO₃/CuI double anode buffer layer," *Solar Energy Materials and Solar Cells*, **141**, 429– 435 (2015). <https://doi.org/10.1016/j.solmat.2015.06.017>
- [34] M.I. Hossain, F.H. Alharbi, and N. Tabet, "Copper oxide as inorganic hole Transport material for lead halide perovskite based solar cells," *Solar Energy*, **120**, 370–380 (2015). <https://doi.org/10.1016/j.solener.2015.07.037>
- [35] B.M. Omer, A. Khogali, and A. Pivrikas, "Combined effects of carriers charge mobility and electrodes work function on the performance of organic photovoltaic devices," in: *37th IEEE Photovoltaic Specialists Conference*, pp. 000734–000743 (IEEE, 2011). <https://doi.org/10.1109/LED.2011.2117953>
- [36] O.D. Iakobson, O.L. Gribkova, A.R. Tameev, and J.-M. Nunzi, "Polymeric semiconductors for hybrid organic-inorganic solar cells: A comparative study of water-soluble polythiophenes," *Scientific Reports*, **11**, 3697 (2021). <https://doi.org/10.1038/s41598-021-84452-x>
- [37] N. Sharma, S.K. Gupta, and C.M. Singh Negi, "Enhanced performance of organic solar cells by using zinc oxide and graphene quantum dots as electron transport layer," *Superlattices and Microstructures*, **135**, 106278 (2019). <https://doi.org/10.1016/j.spmi.2019.106278>
- [38] K. Weng, L. Ye, L. Zhu, J. Xu, J. Zhou, X. Feng, G. Lu, *et al.*, "Molecular design of benzodithiophene-based organic photovoltaic materials to achieve both high VOC and JSC," *Nature Communications*, **11**, 2855 (2020). <https://doi.org/10.1038/s41467-020-16621-x>

- [39] Y. Zhang, X. Li, T. Dai, D. Xu, J. Xi, and X. Chen, "Charge transport and extraction of PTB7:PC71BM organic solar cells: effect of film thickness and thermal-annealing," *RSC Advances*, **9**(31), 17857-17864 (2019). <https://doi.org/10.1039/c9ra02877c>
- [40] A. Gusain, R.M. Faria, and P.B. Miranda, "Controlling the morphology and performance of bulk heterojunctions in solar cells: Lessons from interfacial forces," *Frontiers in Chemistry*, **7**, 61 (2019). <https://doi.org/10.3389/fchem.2019.00061>
- [41] M. Zhang, X. Xu, L. Yu, and Q. Peng, "High-performance ternary organic solar cells with controllable morphology via sequential layer-by-layer deposition," *Journal of Power Sources*, **506**, 229961 (2021). <https://doi.org/10.1016/j.jpowsour.2021.229961>
- [42] D. Spoltore, W.D. Oosterbaan, S. Khelifi, J.N. Clifford, A. Viterisi, E. Palomares, M. Burgelman, *et al.*, "A combined experimental and modeling study of the factors limiting the performance of polymer:fullerene solar cells processed from chlorobenzene and 1,2-dichlorobenzene," *Advanced Energy Materials*, **3**(2), 227-236 (2013). <https://doi.org/10.1002/aenm.201200674>
- [43] M.R. Khan, and B. Jarzabek, "Recent Advances in Polymer-Based Materials for High-Performance Perovskite Solar Cells," *Polymers*, **15**(18), 3674 (2023). <https://doi.org/10.3390/polym15183674>
- [44] C. Deibel, Photocurrent in organic solar cells – Part 1, Blog post, Notes on Disordered Matter (2009). <https://blog.disorderedmatter.eu/2009/07/20/photocurrent-in-organic-solar-cells-part-1>
- [45] B. Qi, and J. Wang, "Fill factor in organic solar cells," *Journal of Materials Chemistry*, **22**(46), 24315- 24325 (2012). <https://doi.org/10.1039/c2jm33719c>
- [46] M. Wright, and A. Uddin, "Organic-inorganic hybrid perovskites: A solution for cost-effective solar cells," *Solar Energy Materials, and Solar Cells*, **107**, 87–117 (2012). <https://doi.org/10.1016/j.solmat.2012.07.006>
- [47] S. Galindo, M. Ahmadpour, L.G. Gerling, A. Marsal, C. Voz, R. Alcubilla, and J. Puigdollers, "Analysis of the origin of open circuit voltage in organic solar cells with different device architectures," *Organic Electronics*, **15**(11), 3034-3041 (2014). <https://doi.org/10.1016/j.orgel.2014.07.011>
- [48] Z. Liu, and Y. Lin, "Recent advances in polymer-based interfacial materials for efficient and stable organic solar cells," *Polymer Testing*, **131**, 108387 (2024). <https://doi.org/10.1016/j.polymertesting.2024.108387>
- [49] A. Kumar, and S. Sharma, "Performance analysis of organic solar cells with different anode buffer layers," *IOSR Journal of Electrical and Electronics Engineering (IOSR-JEEE)*, **14**(4), 49-54 (2019). <https://www.iosrjournals.org/iosr-jeee/Papers/Vol14%20Issue%204/Series-1/G1404014954.pdf>
- [50] J. Yang, X. Wang, X. Yu, J. Liu, Z. Zhang, J. Zhong, and J. Yu, "Recent advances in organic solar cells with non-fullerene acceptors: From morphology control to device optimization," *Nanomaterials*, **13**(21), 2899 (2023). <https://doi.org/10.3390/nano13212899>
- [51] S. Valsalakumar, S. Bhandari, A. Roy, T.K. Mallick, J. Hinshelwood, and S. Sundaram, "Machine learning-based optimization of hole transport layer-free carbon-based perovskite solar cells," *Npj Computational Materials*, **10**(1), 94 (2024). <https://doi.org/10.1038/s41524-024-01383-7>
- [52] T.M. Mukametkali, B.R. Ilyassov, A.K. Aimukhanov, T.M. Serikov, A.S. Baltabekov, L.S. Aldasheva, and A.K. Zeinidenov, "Optical and electronic properties of monolayer MoS₂ under different strains: DFT study," *Physica B: Condensed Matter*, **660**, 414784 (2023). <https://doi.org/10.1016/j.physb.2023.414784>
- [53] S. Sakib, M.Y. Mohd Noor, M.R. Salim, A.S. Abdullah, A.I. Azmi, M.H. Ibrahim and M.H. Ibrahim, "Effect of different composition ratio on structural and morphological properties of TiO₂ nanoparticles synthesized by sol-gel method," *Mater. Today: Proc.* **76**, 176–182 (2023). <https://doi.org/10.1016/j.matpr.2022.11.4564>
- [54] C. Poelking, J. Benduhn, D. Spoltore, M. Schwarze, S. Roland, F. Piersimoni, D. Neher, *et al.*, "Interfacial electrostatics control open-circuit voltage in organic solar cells," *Communications Physics*, **5**(1), 332 (2022). <https://doi.org/10.1038/s42005-022-01084-x>
- [55] M.R. Khan, and B. Jarzabek, "The effect of 3D printing process parameters on the mechanical properties of PLA polymer and polymer composites: A review," *Polymers*, **15**(18), 3674 (2023). <https://doi.org/10.3390/polym15183674>
- [56] S.A. Moiz, M.S. Alzahrani, and A.N.M. Alahmadi, "Influence of process parameters on mechanical properties of natural fiber-reinforced polymer composites: A review," *Polymers*, **14**(17), 3610 (2022). <https://doi.org/10.3390/polym14173610>
- [57] D. Bartesaghi, I. del C. Pérez, J. Kniepert, S. Roland, M. Turbiez, D. Neher, and L.J.A. Koster, "Competition between charge extraction and recombination determines the fill factor in organic solar cells," *Nature Communications*, **6**, 7083 (2015). <https://doi.org/10.1038/ncomms8083>
- [58] J.C. Nolasco, R. Cabré, J. Ferré-Borrull, L.F. Marsal, M. Estrada, and J. Pallarès, "Design of two-dimensional silicon photonic crystals by means of a guided-mode resonant filter structure," *Journal of Applied Physics*, **107**(2), 023108 (2010). <https://doi.org/10.1063/1.3296294>

ВІПЛИВ ДОДАТКОВОГО ШАРУ РЗНТ НА ПРОДУКТИВНІСТЬ ПОЛІМЕРНОГО СОНЯЧНОГО ЕЛЕМЕНТА РЗНТ: ІС₆₀ВА

Чіттур Девараджан Рамабадран^{a,b}, К. Себастьян Судхір^a

^aКафедра фізики, дослідницька лабораторія моделювання оптоелектронних пристроїв, Коледж Христа [Автономний],
Ірінджалакуда, Тріссур, 680125, Університет Калікута, Керала, Індія

^bКафедра фізики, Урядовий коледж, Чіттур, Палаккад, 678104, Університет Калікута, Керала, Індія

Було проведено моделювання з використанням програмного забезпечення SCAPS 1-D для вивчення впливу додаткового шару РЗНТ (полі-3-гексилтіофен) на продуктивність сонячних елементів з об'ємним гетеропереходом, зокрема з активним шаром РЗНТ: ІС₆₀ВА. Досліджувана структура елемента - ІТО/PEDOT:PSS/РЗНТ/РЗНТ:ІС₆₀ВА/ZnO NPs/Al. Після стандартизації програмного забезпечення ми визначили оптимальні параметри структури сонячного елемента, проаналізувавши різні фактори, що впливають на продуктивність елемента в різних шарах. Згодом, після оптимізації структури, ефективність перетворення енергії (PCE) значно покращилася, збільшившись з 5,18% без додаткового шару до 15,26% з додатковим шаром.

Ключові слова: сонячний елемент з об'ємного гетероперехідного полімеру; фулерен; SCAPS 1-D; активний шар; ETL; HTL

Improved efficiency of bulk heterojunction polymer solar cells by omitting electron transport layer: a SCAPS 1D simulation study

Ramabadran C. D.^{1,3}, Dhanya N. P.^{2,*}, Sudheer K. S.^{1,*}

¹Department of Physics, Opto-electronic Device Simulation Research Lab, Christ College [Autonomous], Irinjalakuda, Thrissur, 680125, Kerala, India

²Department of Physics, KKTM Govt. College, Pullut, Thrissur, 680663, University of Calicut, Kerala, India

³Department of Physics, Government College, Chittur, Palakkad, 678104, University of Calicut, Kerala, India

*Emails: 1.ramabadrancd@christcollegeijk.edu.in, 2.dhanyaajan@gmail.com,
3.sudheersebastian@christcollegeijk.edu.in

Received: 1 May 2024; Accepted for publication: 16 September 2024

Abstract. This paper examines the function of bulk heterojunction polymer solar cells (BHJPSCs) in the realm of photovoltaics. Specifically, the performance of a BHJPSC setup eliminating the electron transport layer (ETL) is investigated. With the aid of SCAPS 1D software, simulation studies are done to evaluate the effectiveness of this innovative approach. Initially, the program is standardized by replicating a key experimental study that used P3HT:IC₆₀BA as the active layer, producing results consistent with actual observations. The solar cell structure ITO/PEDOT:PSS/P3HT:IC₆₀BA/ZnO NPs/Al was studied where an efficiency of 3.97 % is achieved through standard procedures. Interestingly, by omitting the ETL in the simulations leads to a substantial efficiency of 10.83 % after optimization. This increased efficiency demonstrates an economically feasible solar cell structure. Improved photovoltaic performance may arise from additional research and optimisation of BHJPSC designs without ETL.

Keywords: bulk heterojunction polymer solar cell, SCAPS 1-D, HTL, Fullerene bisadduct IC₆₀ BA.

Classification numbers: 3.4.1, 4.10.4

1. INTRODUCTION

Non-conventional energy sources are essential when energy demands rise and conventional fossil fuels become less common. Bulk heterojunction polymer solar cells (BHJ PSCs) have

recently received increased attention as a possible renewable energy source due to their simple fabrication technique, low cost, light weight nature, and adaptability [1 - 3]. The active layer of these solar cell devices typically consists of two organic semiconductor materials: an electron acceptor (n-type fullerene derivatives) and an electron donor (p-type conjugated polymer) [4]. The unique features of "small molecule organic or polymer solar cells", QD sensitised solar cells (QD-SSCs) (Ghoreishi *et al.*, 2014) have further led to their appeal [5 - 9].

Poly(3-hexylthiophene) (P3HT) is an important material for electron donation in bulk heterojunction (BHJ) polymer solar cells because of its good hole mobility, stable with the environment and absorption in the extended red light. One of the most popular electron acceptors is [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) [10-12], which has low LUMO energy levels and provides an open circuit voltage of just over 0.6 V [13]. Of late, there has been a push to use the indene-C60 bis adduct (IC₆₀BA), which has higher LUMO energy levels and provides for the open circuit voltage of about 0.84 V, along with more efficiency [14 - 18].

Bulk heterojunction polymer solar cells (BHJPSCs) are beneficial to photovoltaic technology because they are inexpensive and quick to manufacture. The bulk heterojunction (BHJ) structure allows for rapid exciton dissociation by utilizing a bicontinuous network of donor and acceptor materials. Bendenia *et al.* demonstrated that an all polymer blend resulted in excellent photogeneration [19]. The BHJ structure was first described by Heeger and co-workers in 1995 for polymer fullerenes blends [20], and this is where we find that additives can ultimately increase the domain sizes of the donor and acceptor materials and improve performance [21, 22].

To enhance the performance of bulk heterojunction (BHJ) polymer solar cells (PSCs), Li *et al.* studied indene-C60 bis adduct (IC₆₀BA) as an electron acceptor to address the issue of low open circuit voltage (Voc) of the P3HT:PCBM blend [23]. In an experiment with P3HT:IC₆₀BA, they achieved high power conversion efficiencies (PCE) and a Voc value of 0.84 V [24 - 25]. The authors modelled the solar cells using SCAPS 1-D software and validated the model against experiment [26 - 27]. In this case, the hole transport layer was PEDOT:PSS and the active layer was P3HT:IC₆₀BA.

In polymer solar cells, electron transport layers (ETL) are used for collecting the electrons but could limit the performance of the device because of issues with the material such as TiO₂ degrading from UV light or trapping the electrons because of defects in the layer. Studies have shown that removing the ETL can create an increase in efficiency and lower the cost of production. In some cases, the performance of the ETL materials can be greatly affected by lowering the thickness of the layer, so the material becomes a scalability issue for mass production. An optimization of a standard structure replaced without ETL [ZnO nanoparticles] leads to a most significant enhancement of the efficiency from structures with ETL proving evidence to high efficiency polymer solar cells.

2. DEVICE SIMULATION METHODOLOGY

This study used the one-dimensional Solar Cell Capacitance Simulator-SCAPS version (3.3.08). Engineers and researchers benefited from its unique features and enhancements. SCAPS-1D 3.3.08 characteristics are listed below. It solves the optical and electrical models for the full arrangement. SCAPS-1D users can describe their solar cell structure by specifying layers. Each layer can have varied dielectric constant, electron affinity, thickness, and bandgap. Users can enter own material specifications; however, the software has a library of solar cell materials. SCAPS-1D calculates carrier transport via drift-diffusion. This model accounts for

electric field drift and carrier concentration gradient diffusion. The SCAPS-1D device simulation tools enable to model recombination mechanisms by fully specifying defect states. The bulk defect states can be specified with an energy distribution, density of states, and electron and hole capture cross-sections within the bulk of each semiconductor layer. The interface defect states can also be specified at the heterojunctions, with properties unique to each of those interface defect states. The bulk defect states and interface defect states can be parameterized to obtain Shockley-Read-Hall (SRH) recombination rates, which will yield a better representation of your device's electrical behaviour. The software simulates solar cell layer light absorption and electron-hole pair production. SCAPS-1D can simulate varied light intensities and spectrum distributions (AM1.5G, monochromatic, etc.).

The software simulates the I-V characteristics of solar cells under varying conditions, generating outputs such as Voc [Open circuit voltage], Jsc [Short circuit current], FF[Fill factor], and PCE[Power conversion efficiency]. Multi-junction solar cells, which can absorb more sunlight by having a stack of multiple layers with bandgaps, can also be simulated. An intuitive GUI helps shorten the time to setup simulations, adjust parameters, and view results using SCAPS-1D. Result files can be saved in many different types of formats for later analyses, and graphs can be edited and saved for the reporting component. SCAPS-1D allows users to perform sensitivity analysis to see how variations in parameters can affect solar cell performance. For organic solar cells software is used for exploring various donor-acceptor material combinations and their impact on device efficiency and it was found that the ratio of donor to acceptor materials critically affects charge transport and recombination rates. Optimal blend ratios were determined for enhanced efficiency. In our study, we have used a 1.5 AM spectrum for illumination. Previous simulations have been performed on fullerene and non-fullerene BHJ, perovskite, and tandem solar cells (Abdelaziz *et al.*, 2019, 2020; Gupta and Dixit, 2018; Bahrami *et al.*, 2019). In their study in 2019, Abdelaziz *et al.* focused on the simulation and optimization of lead-free perovskite solar cells using SCAPS-1D software. They investigated the performance of a solar cell structure composed of FTO/TiO₂/CH₃NH₃SnI₃/Spiro-OMeTAD/Au. Their primary objective was to enhance the cell efficiency by optimizing the thickness of the absorber layer and the defect density. The simulation results highlighted that optimizing these parameters significantly improved the power conversion efficiency (PCE) of the solar cells.

SCAPS-1D possesses multiple control panels for parameter variation and thin film solar cell simulation. Each of the control panels serves to simplify the management and tuning of a simulation by targeting a single simulation step. Project files (multiple simulations) can be created, saved, and retrieved in the main control panel. The simulation run control panel is where the initiation of simulation, pause, and simulation control takes place. The device construction panel is where layers of the solar cell can be added, deleted, and defined. Depending on the material property panel the user can select the bandgap, electron affinity, and dielectric constants for each material layer. The user has the ability to select carrier mobility, recombination and diffusion coefficients. In the defects and recombination panel, energy levels (and densities), and capture cross-sections for the electron and hole can be selected to specify bulk defect state in the material. The user can select to define the SRH, radiative, and Auger recombination parameters. In the lighting and optical properties panel users can select the type and intensity of lighting source and confirm their designs against AM1.5G physical sun spectra or to define a light source spectral distribution specifically. Each layer's reflectance and absorption coefficients affect light absorption and the device's functionality. The electrical simulation allows the user to select forward and reverse biases. The temperature settings panel allow the temperature settings to be defined for the simulated operating temperatures.

The output and results panel display graphical outputs such as I-V curves, C-V curves, and QE spectra. Users can customize the graphs, zoom in on specific regions, and compare multiple simulations. The data export tool helps in exporting simulation results to various formats (e.g., CSV) for further analysis. These control panels are designed to provide a comprehensive and user-friendly interface for setting up and managing solar cell simulations. By organizing the software into these specific panels, the SCAPS-1D ensures that users can easily find and adjust the parameters they need for their specific simulation requirements.

SCAPS-1D numerically solves the coupled Poisson and continuity equations iteratively to yield a steady-state operating point in equilibrium for the general semiconductor equations. Before and after deployment of the stimulus, the general semiconductor equations, [e.g. Poisson's equations, continuity equations, drift and diffusion current equations], are very quick in determining the electrical performance of the electronic device. Ultimately the electrical performance is derived from potential, photons, or thermal energy as sourced from the stimulus. The device electrical performance and I-V characteristics are formulated through these equations. Therefore, this provides the theoretical method for investigation and understanding device performance. The Poisson equations express the electrostatic potential with semiconductor charge distributions [Equation 1]. The continuity equations express charge conservation with holes and electrons [Equations 2-3]. The drift-diffusion equations express the electron and hole current densities [Equations 4-5]. The finite difference method (FDM) discretizes the semiconductor equations on a one-dimensional grid. The equations are solved at discrete grid nodes across the one-dimensional grid sections of the device. Initial guesses of the electrostatic potential and carrier concentrations are obtained. The guess may come from equilibrium conditions or previous simulations will provide a more informed guess. For boundaries at the contacts, Dirichlet or Neumann boundary conditions are applied (prescribed potential or prescribed current density). At the interfaces of materials, the potentials and current densities are continuous. The numerical iterative methods such as the Newton-Raphson Method and Gummel Iteration solve the coupled systems of nonlinear algebraic equations from discretization. The Newton-Raphson Method linearizes the nonlinear equations about the guess and solves the linear system iteratively. The Poisson's equation for potential and the continuity equations for carriers are solved alternatively. The total device current is computed from the numerically integrated current densities across the cross sectional area of the device. The total device current is derived at a corresponding potential. The I-V curve is computed by varying actual potential application and repeating the solution. They will also analyze and predict the performance of new architectures, save significant human resources, time, and money on experiment costs.

$$\frac{\partial \epsilon_0 \epsilon}{\partial x} \frac{\partial \Psi}{\partial x} = -q (p - n + N_D - N_A + \frac{\rho_{def}}{q}) \quad (1)$$

$$\frac{\partial J_n}{\partial x} - Un + G = \frac{\partial n}{\partial t} \quad (2)$$

$$\frac{\partial J_p}{\partial x} - Up + G = \frac{\partial p}{\partial t} \quad (3)$$

$$J_n = - \frac{\mu_n n}{q} \frac{\partial E_{Fn}}{\partial x} \quad (4)$$

$$J_p = + \frac{\mu_p p}{q} \frac{\partial E_{Fp}}{\partial x} \quad (5)$$

In the above-mentioned equations, the charge is denoted by q . The fundamental charge of an electron is approximately 1.602×10^{-19} C. It is a constant used in all the equations involving charge carriers (electrons and holes). Ψ denotes the electrostatic potential which

represents the electric potential energy per unit charge at a point in the semiconductor. It is a solution to the Poisson equation and influences the movement of charge carriers. The symbols p , n , p_t , and n_t stand for free holes and electrons concentrations (number of electrons or holes per unit volume), trapped holes and trapped electrons which represent holes and electrons captured by traps, respectively. N_D (donors that have released an electron and become positively charged) and N_A (acceptors that have captured an electron and become negatively charged) denote ionized donor-like and ionized acceptor-like doping concentrations, respectively. Here, ϵ signifies absolute permittivity, while ϵ_0 denotes permittivity in free space which represents the ability of the material to permit electric field lines. It influences the electric potential distribution within the material. J_n and J_p signify electron and hole current densities which are derived from the drift and diffusion components of carrier transport, respective while U_n denotes electron recombination rate, U_p is the rate of hole recombination (The rate at which electrons and holes recombine and annihilate each other), and G is the generation rate (The rate at which electron-hole pairs are generated, typically by absorption of photons in the context of solar cells). In contrast to what has been described so far, E_{Fn} and E_{Fp} stand for the electron and hole quasi-Fermi levels, whereas μ_n and μ_p indicate the electron and hole mobility, respectively. In SCAPS-1D software, understanding the concepts of assumed Fermi energy levels (E_{Fn} , E_{Fp}) and the conductivity of electrons and holes (n , p) is crucial for analyzing the behavior of semiconductor devices. In thermodynamic equilibrium, the Fermi level is the energy level at which the probability of finding an electron is 50 %. It represents the chemical potential for electrons in the system. In non-equilibrium conditions (e.g., when a device is illuminated or under bias), separate quasi-Fermi levels are defined for electrons (E_{Fn}) and holes (E_{Fp}). When there are non-equilibrium conditions, the energy level E_{Fn} corresponds to the distribution of electrons in the conduction band, and the corresponding energy level E_{Fp} corresponds to the distribution of holes in the valence band. These quasi-Fermi levels are crucial for determining the carrier concentrations (n and p) and the current flow in the device. In equilibrium, E_{Fn} and E_{Fp} coincide with the overall Fermi level (E_F), as there is no net generation or recombination. Under illumination or bias, E_{Fn} and E_{Fp} split, reflecting the different energy distributions of electrons and holes. The splitting is a measure of the deviation from equilibrium and is related to the applied voltage and light-generated carriers. The conductivity attributed to holes is increased by higher hole concentration (p) and mobility (μ_p), whereas the conductivity attributed to electrons is increased by higher electron concentration (n) and mobility (μ_n).

Al [Cathode]
ZnONPS [ETL]
P3HT : IC₆₀BA [Active Layer]
PEDOT : PSS [HTL]
ITO [Anode]
Glass Substrate

Figure 1. Simulated bulk heterojunction polymer solar cell structure.

Figure 1 depicts the configuration of the simulated bulk heterojunction structure, which is Glass substrate/ITO/PEDOT: PSS/P3HT:IC₆₀BA/ZnONPS/Al. Table 1 shows the material parameters used in the simulation, including electron affinity (χ), bandgap energy (E_g), and relative permittivity (ϵ_r). The mobilities for holes and electrons are μ_p and μ_n , respectively. N_D and N_A represent donor and acceptor density, respectively. N_v and N_c represent the effective

density of states in the valence and conduction bands, respectively, whereas N_t represents the defect density. The anode and cathode work functions are set at 4.8 and 4.2 eV, respectively. The thermal velocity is set to 10^7 cm/s, and the capture cross section for holes and electrons is set to 1×10^{-25} cm² across all layers. As already mentioned, the main output parameters of solar cell are V_{oc} , J_{sc} , FF and PCE. The required material parameters for simulation and absorption spectra are gathered from the literature [26 - 28, 29 - 37]. Table 2 shows the comparative evaluation of experimental and simulated values for calibrating SCAPS 1-D

Table1. Simulation parameters of bulk heterojunction polymer solar cell.

Parameter	HTL	Active layer	ETL
Thickness [nm]	27	250	46
E_g (eV)	1.6	1.68	3.2
χ (eV)	3.4	3.880	4.6
ϵ_r	3.0	3.3	9.0
μ_n (cm ² /vs)	10^1	10^7	10^7
μ_p (cm ² /vs)	400	10^7	10^7
N_A (cm ⁻³)	10^{15}	0	0
N_D (cm ⁻³)	0	3.2×10^{18}	1.1×10^{18}
N_C (cm ⁻³)	2.2×10^{15}	1.6×10^{20}	2.2×10^{19}
N_V (cm ⁻³)	1.8×10^{18}	10^{19}	1.8×10^{19}
N_t (cm ⁻³)	10^{15}	10^{16}	10^{15}

Table 2. Evaluation of simulated parameters with experimental results.

Parameter	Experimental	Simulated
V_{oc} (V)	0.84	0.85
J_{sc} (mA/cm ²)	8.36	8.21
FF (%)	59	57
PCE (%)	4.1	3.97

3. RESULTS AND DISCUSSION

3.1. Effect of active layer thickness

The active layer thickness is varied between 300 nm to 700 nm. As thickness increases, V_{oc} , J_{sc} and PCE increase, but FF decreases. The corresponding graph is given in Figure 2. V_{oc} increases due to the production of more charge carriers. Variation in J_{sc} accounts for more light absorption and FF declines due to increase in series resistance and recombination. Variation in PCE accounts for the combined effect of all other output parameters

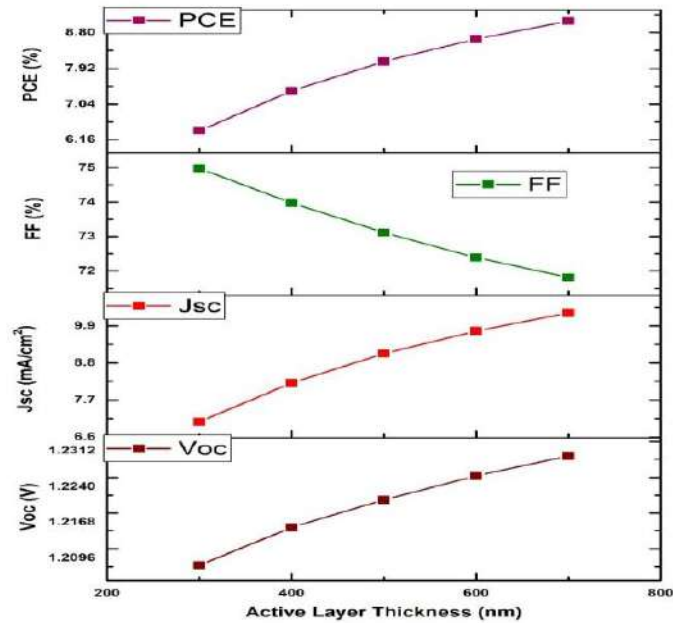


Figure 2. Influence of active layer thickness on cell performance.

3.2. Effect of shallow defect density of active layer

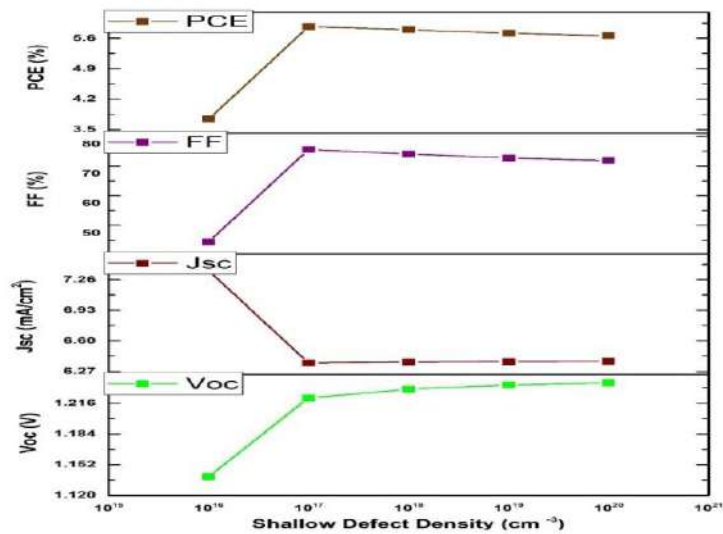


Figure 3. Influence of shallow defect density on cell performance.

The shallow defect density of the active layer is increased from 10^{16} cm^{-3} to 10^{20} cm^{-3} . Voc is found to increase with shallow defect density, although FF and PCE vary similarly. Jsc is found to decrease with increasing shallow defect density. Optical techniques based on sub-bandgap absorption or emission can also provide insights into the defect density in the active layer. . It was discovered that optimal performance occurs at 10^{17} cm^{-3} , with an efficiency of

5.87 %. These variances are depicted in Figure 3. Shallow defect density can act as trapping sites, leading to non-radiative recombination which impacts the Voc. The Jsc is lowered by high defect density due to increased recombination and trapping of charge carriers. FF is decreased due to increased recombination and localized resistive losses, at high defect density. Reductions in Voc, Jsc, and FF results in the variation of PCE. Improved material purity, optimized fabrication processes, and careful control of the polymer morphology helps in reducing shallow defect density for better performance

3.3. Effect of temperature

The temperature is changed from 300 K to 360 K. The optimal performance is achieved at 300 K. The variations of output parameters are depicted in Figure 4.

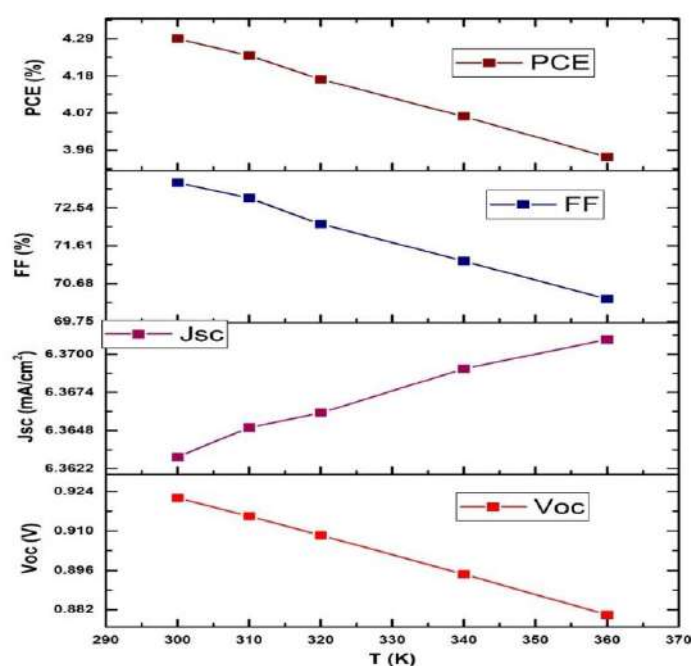


Figure 4. Influence of temperature on cell performance.

At higher temperatures, the materials carrier concentrations, mobility of charge carriers, resistance, and bandgap changes. As temperature rises, the intrinsic carrier concentration in the semiconductor increases, leading to a reduction in the separation of the quasi-Fermi levels. This results in a decrease in Voc. Additionally, increased temperature can enhance the recombination rates of charge carriers, further lowering Voc. Jsc increases with increasing temperature due to enhancement in the mobility of charge carriers and slightly increase in absorption of photons. Elevated temperatures increase the series resistance and decrease the shunt resistance in the solar cell leading to the reduction in FF. The diode ideality factor also increases with temperature, negatively impacting the FF. Since PCE is a product of Voc, Jsc, and FF, the overall efficiency tends to decrease with rising temperature. Maintaining the efficiency of polymer solar cells requires proper thermal management, which includes choosing materials with improved thermal stability to lessen the negative impacts of temperature rise, using heat sinks, and optimising cell

design. In polymers, thermally generated carriers increase with temperature, enhancing the overall carrier concentration.

3.4. Effect of electron affinity of active layer

The electron affinity of the active layer ranges from 3.5 eV to 4 eV. Simulation revealed an optimal value of 5.75 % efficiency for 3.7 eV electron affinity. This is due to the enhancement in exciton dissociation, charge transport and reduction in recombination losses. Figure 5 shows a graphical illustration. If the electron affinity of the acceptor is high, it ensures a larger energy difference, potentially increasing V_{oc} . However, if the electron affinity is too high, it may also increase recombination rates, which could negate the benefits. A higher electron affinity can facilitate better electron transport to the electrode, reducing recombination within the active layer. This enhances the overall charge collection efficiency, contributing to higher J_{sc} and FF. Conversely, if the electron affinity is not optimized, it can lead to poor charge transport and higher recombination losses. Proper electron affinity ensures optimal energy level alignment, facilitating efficient charge transfer. Optimized electron affinity enhances charge transport and reduces recombination, improving J_{sc} and FF. Well-matched electron affinity reduces recombination losses, enhancing overall device efficiency.

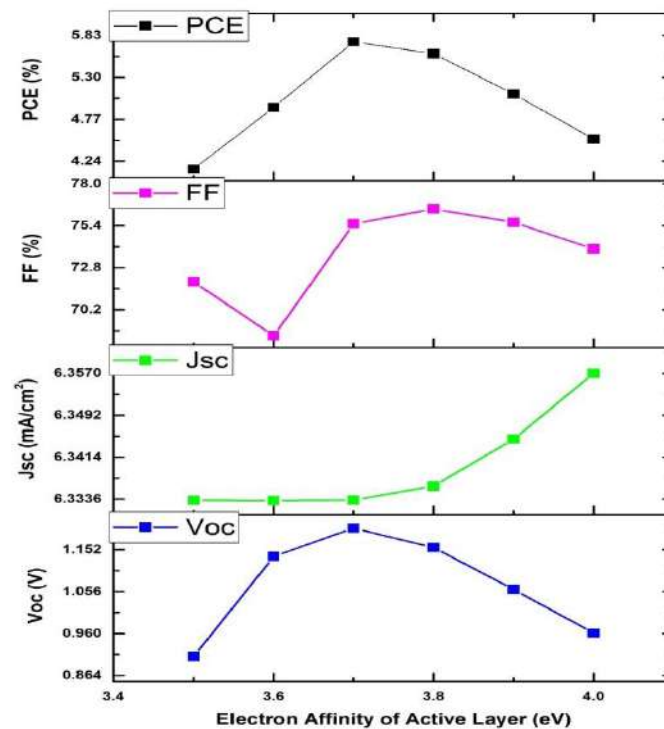


Figure 5. Influence of electron affinity of active layer on cell performance.

3.5. Effect of carrier concentration of active layer

The carrier concentration of the active layer is increased from 10^{17} cm^{-3} to 10^{21} cm^{-3} . In this study, we discovered that V_{oc} reaches its maximum value of 1.2013 V, whereas PCE reaches its maximum value of 5.76 % at 10^{18} cm^{-3} . J_{sc} achieves its maximum value of 6.333860 mA/cm^2 ,

whereas FF achieves its greatest value of 76.08 % at 10^{21} cm^{-3} . Figure 6 shows the corresponding graph. Increased doping concentration can lead to better alignment of energy levels, reducing recombination losses and potentially increasing Voc. Proper doping can enhance the electrical conductivity and mobility of charge carriers, improving their transport to the electrodes and increasing Jsc

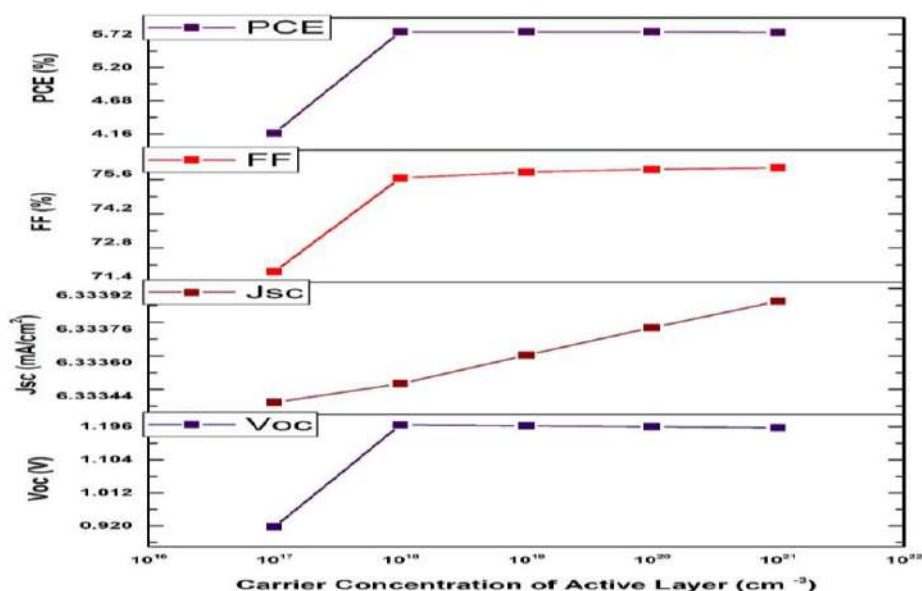


Figure 6. Effect of carrier concentration of active layer on cell performance.

Appropriate doping levels can decrease the series resistance (by increasing conductivity) and minimize shunt pathways (by reducing defects), thereby increasing the FF. Optimal doping improves Voc, Jsc, and FF, leading to higher PCE. To maximize the performance of polymer solar cells, it's crucial to optimize the doping concentration. This involves finding a balance where the doping is sufficient to improve electrical properties and charge transport but not so high as to introduce significant recombination losses or defects.

3.6. Influence of thickness of hole transport layer

The thickness of hole transport layer [HTL] is changed from 25 nm to 45 nm. The optimized values are obtained for a thickness of 30 nm. The corresponding diagram is given in Figure 7. An optimal HTL thickness ensures efficient hole transport and extraction, maintaining a high built-in potential and resulting in a higher Voc. An appropriately thick HTL allows for efficient hole transport to the anode without significant recombination losses, leading to a higher Jsc. A very thick HTL increases series resistance, reducing the FF. An optimal HTL thickness improves Voc, Jsc, and FF, leading to a higher PCE.

3.7. Effect of band gap of HTL

The band gap of HTL varies from 1.4 eV to 1.8 eV. It was discovered that Voc, FF, and PCE rise with the band gap and achieve optimal performance at 1.8 eV. Figure 8 depicts the variations in the output parameters. Voc is a direct consequence of bandgap, with a higher

bandgap resulting in higher Voc and a lower rate of radiative recombination. Jsc drops with increasing bandgap due to reduced production of electrons, as few photons have enough energy. An HTL with an appropriate band gap ensures proper energy level alignment with the active layer, minimizing energy losses during hole extraction resulting in a higher Voc. A mismatched band gap can create energy barriers, hindering hole extraction and leading to increased recombination, which reduces Jsc. A properly aligned band gap minimizes recombination and series resistance, leading to a higher FF. An optimal HTL band gap ensures efficient charge extraction and minimal recombination, maximizing Voc, Jsc, and FF, and thus PCE. To maximize the performance of polymer solar cells, the HTL band gap should be optimized to align well with the energy levels of the active layer.

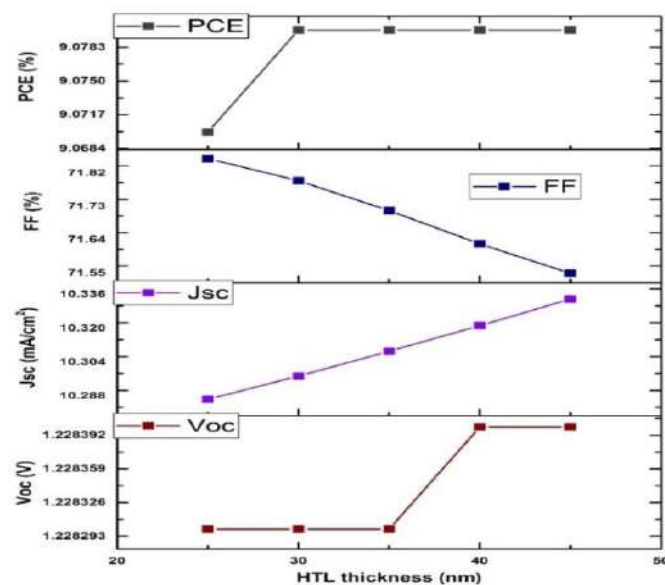


Figure 7. Influence of thickness of HTL.

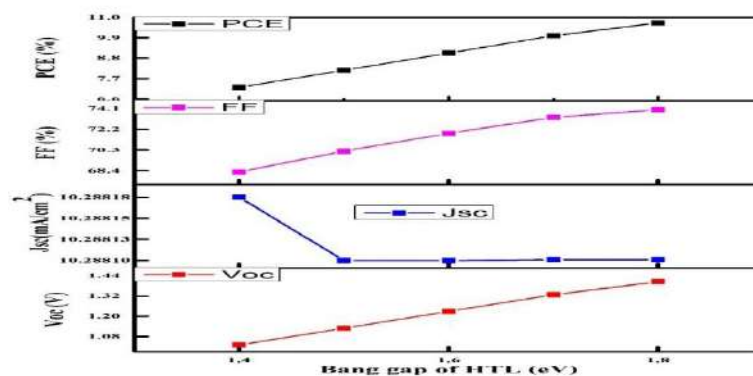


Figure 8. Influence of Band Gap of HTL on cell performance.

3.8. Influence of electron affinity of HTL

The electron affinity of HTL ranges from 3.2 eV to 3.6 eV. The optimal performance is achieved at 3.4 eV. Figure 9 shows the respective changes in the output parameters. An HTL with suitable electron affinity will align its energy levels with the active layer, preventing electron leakage and recombination at the HTL/anode interface. If the electron affinity is mismatched, it can cause inefficient charge separation and increased recombination losses, reducing J_{sc} . Proper electron affinity reduces recombination losses and ensures efficient charge transport, leading to a higher FF at 3.4 eV. A high electron affinity can increase recombination rates and resistive losses, reducing the FF. An optimal electron affinity ensures efficient charge separation and transport, maximizing V_{oc} and FF, leading to a higher PCE at 3.4 eV. Mismatched electron affinity can result in suboptimal V_{oc} , J_{sc} , and FF, thereby reducing the overall PCE. To maximize the performance of polymer solar cells, the electron affinity of the HTL should be optimized to ensure proper energy level alignment with the active layer and the anode

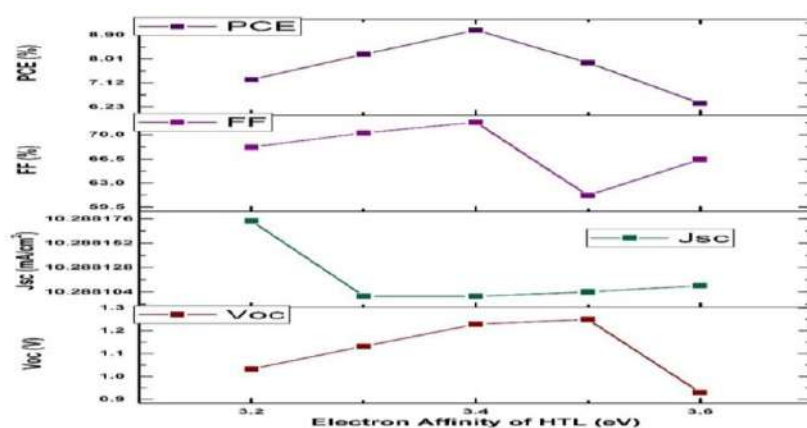


Figure 9. Effect of electron affinity of HTL on cell performance.

3.9. Effect of effective density of states [CB] of HTL

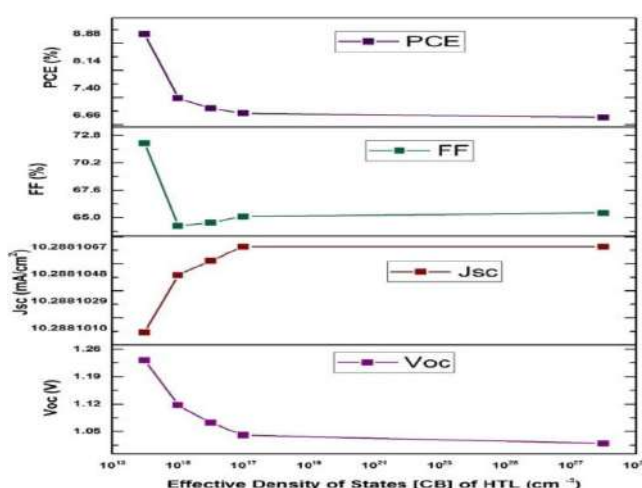


Figure 10. Influence of density of states [CB] of HTL.

The effective density of states of conduction band of HTL is changed from 10^{14} cm^{-3} to 10^{18} cm^{-3} . In this case, Voc and PCE decrease as the density of states increases. Jsc is found to increase while FF initially decreases, then increases. The optimized performance is obtained at 10^{14} cm^{-3} . The corresponding graph is given in Figure 10.

3.10. Influence of effective density of states [VB] of HTL

The effective density of states of valence band of HTL is changed from 10^{16} cm^{-3} to 10^{20} cm^{-3} . Here, the Voc, FF and PCE decrease as the effective density of states in the valence band of HTL increases. Jsc is found to increase with the effective density of states in the valence band of HTL. The optimized results are obtained at 10^{16} cm^{-3} . The results are shown in Figure 11.

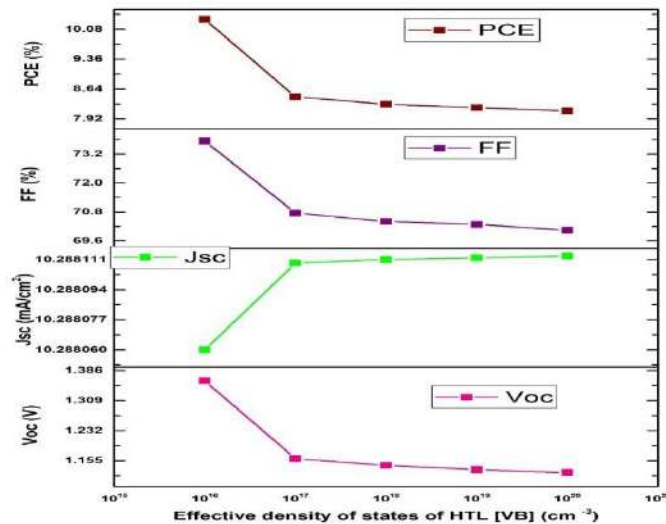


Figure 11. Influence of density of states [VB] of HTL.

3.11. Effect of shallow acceptor density of HTL

There is an increase in the shallow acceptor density of states of HTL from 10^{14} cm^{-3} to 10^{18} cm^{-3} . In this case, it is discovered that Voc increases as HTL acceptor density increases, whereas FF and PCE drop as HTL shallow acceptor density increases. An acceptor density of 10^{14} cm^{-3} yields the best results. The graphs are displayed in Figure 12. The Voc is enhanced by improving the built-in potential and reducing recombination losses. The Jsc is increased with efficient hole transport and minimal recombination. The FF is reduced due to increase in series resistance and recombination. The PCE is maximized when Voc, Jsc, and FF are optimized through balanced HTL doping density. To maximize the performance of polymer solar cells, the doping density in the HTL should be optimized. This involves finding a balance where the HTL is doped enough to improve hole mobility and transport while minimizing recombination and resistive losses. Achieving this balance ensures that the solar cell operates efficiently with high Voc, Jsc, FF, and PCE.

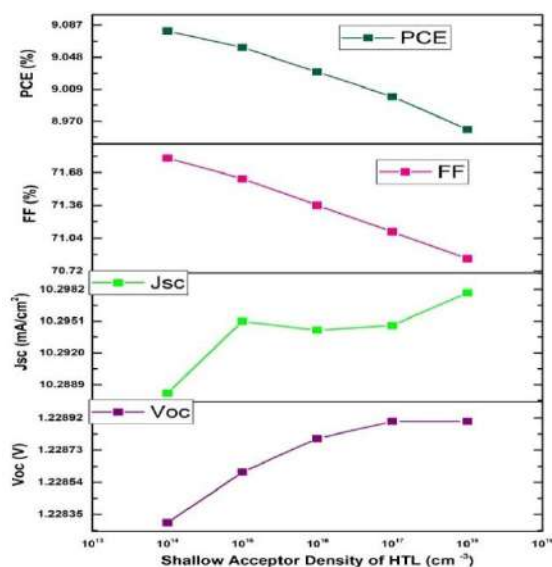


Figure 12. Effect of shallow acceptor density of HTL.

3.12. Influence of defect density of HTL

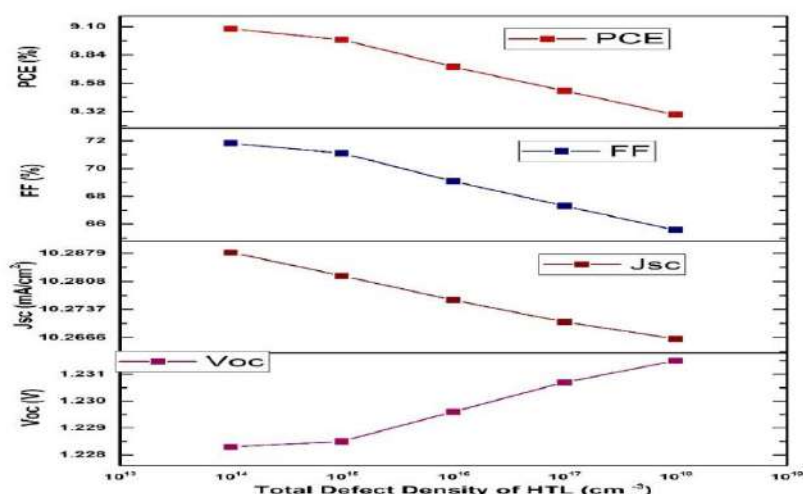


Figure 13. Effect of total defect density of HTL on cell performance.

The total defect density of HTL is changed from 10^{14} cm^{-3} to 10^{18} cm^{-3} . Here, the Voc is found to increase, while the other output parameters are found to decrease as the total defect density of HTL increases. The optimized performance is obtained at a defect density of 10^{14} cm^{-3} . The corresponding graph is given in Figure 13. In polymer solar cells, defects in the HTL can act as recombination centers where charge carriers (electrons and holes) can recombine. When the defect density increases, it can lead to more effective trapping of electrons that have diffused into the HTL. This trapping can reduce the rate of recombination of photogenerated charge carriers at the HTL interface, leading to a lower overall recombination rate in the cell. Reduced recombination of charge carriers helps maintain a higher separation of charges, thereby

increasing the quasi-Fermi level splitting and resulting in a higher Voc. A high defect density can trap charge carriers, reducing their mobility and increasing recombination rates, which lowers the Jsc. High defect density increases the series resistance due to impeded charge transport and introduces more shunt pathways through increased recombination, both of which reduce the FF. Lower defect density ensures efficient charge transport and collection, minimal recombination losses, and optimal series and shunt resistances, thereby maximizing Voc, Isc, and FF, and consequently the PCE. Higher defect density has the opposite effect, reducing all these parameters and thereby lowering the PCE. Careful material selection, processing conditions, and interface engineering are adopted to reduce the defect density

4. CONCLUSIONS

Numerical investigations were conducted on a fullerene BHJ PSC employing the ITO/PEDOT:PSS/P3HT:IC₆₀BA/ZnONPs/Al structure. These studies were validated by comparing the simulated results with experimental data from the literature. Remarkably, the actual device performance closely mirrored the simulated outcomes. Our study examines the impact of parameters such as hole transport layer characteristics, absorber layer thickness, doping concentration, defect density, and temperature on cell performance, specifically focusing on configurations devoid of an electron transport layer (ZnO NPs). Through optimization, we enhanced parameters to augment the power conversion efficiency (PCE). Notably, the optimization process included fine-tuning transport layer properties. The resultant cell performance with optimized numerical parameters yielded: Voc = 1.4158 V, Jsc = 10.294741 mA/cm², FF = 74.30 %, and PCE = 10.83 %. The results are encouraging and may open up future research in this field.

Acknowledgements. The authors would like to acknowledge Pr.M. Burgelman, University of Gent, Belgium for providing the SCAPS 1-D software for the simulation study.

CRedit authorship contribution statement. All authors contributed equally to this work.

Declaration of competing interest. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

REFERENCES

1. Mohd Yusoff A. R., Silva W. J., Schneider F. K. - Understanding the pH-dependent behaviour of graphene oxide aqueous solutions on organic photovoltaic performance, *Solar Energy Materials and Solar Cells* **194** (2019) 62-66.
2. Cui C., Wong W. Y., Li Y. - Improvement of open-circuit voltage and photovoltaic properties of 2d-conjugated polymers by alkylthio substitution, *Energy & Environmental Science* **7** (7) (2014) 2276-2284.
3. Singh T. J., Singh S., Islam S. M., Mahala P., Singh K. J. - Flexible organic solar cells with graphene/pedot: Pss schottky junction on pet substrates, *Optik* **181** (2019) 984-992.
4. Urbina A., Abad J., Romero A. J. F., Lacasa J. S., Colchero J., González-Martínez J. F., Rubio-Zuazo J., Castro G. R., Gutfreund P. - Neutron reflectometry and hard x-ray photoelectron spectroscopy study of the vertical segregation of pcbm in organic solar cells, *Solar Energy Materials and Solar Cells* **191** (2019) 62-70.

5. Riede M., Spoltore D., Leo K. - Organic solar cells-the path to commercial success, *Advanced Energy Materials* **11** (1) (2021) 2002653.
6. Deibel C., Dyakonov V. - Polymer–fullerene bulk heterojunction solar cells, *Reports on Progress in Physics* **73** (9) (2010) 096401.
7. Xu M., Wei C., Zhang Y., Li H., Ma J., Lin J., Wang S., Xue W., Wei Q., Xie L., *et al.* - Ultrafast photoexcitation dynamics behavior of hydrogen-bonded polyfluoreneol, *Chinese Chemical Letters* **35** (1) (2024) 108279.
8. Ghoreishi F., Ahmadi V., Samadpour M. - *Journal of Power Sources* **271** (2014) 195.
9. Ghadiri E., Taghavinia N., Zakeeruddin S. M., Gratzel M., Moser J. E. - Enhanced electron collection efficiency in dye-sensitized solar cells based on nanostructured TiO₂ hollow fibers, *Nano letters* **10** (5) (2010) 1632-1638.
10. He Y., Zhao G., Peng B., Li Y. - High-yield synthesis and electrochemical and photovoltaic properties of indene-c70 bisadduct, *Advanced Functional Materials* **20** (19) (2010) 3383-3389.
11. Bendenia C., Merad-Dib H., Bendenia S., Hadri B. - Numerical modelisation of zno interfacial layer on p3ht: Pcbm based organic photovoltaic bulk heterojunction devices, *Optik* **174** (2018) 167-172.
12. Hazra A., Mal I., Samajdar D., Das T. - Analytical modelling of organic solar cells with scattering interface, *Optik* **168** (2018) 747-753.
13. Sun Y., Cui C., Wang H., Li, Y. - Efficiency enhancement of polymer solar cells based on poly (3-hexylthiophene)/indene-c70 bisadduct via methylthiopheneadditive, *Advanced Energy Materials* **1** (6) (2011) 1058-1061.
14. He Y., Chen H. Y., Hou J., Li Y. - Indene- c60 bisadduct: a new acceptor for high-performance polymer solar cells. *Journal of the American Chemical Society* **132** (4) (2010) 1377-1382.
15. Zhao G., He Y., Li Y. - 6.5 % efficiency of polymer solar cells based on poly (3-hexylthiophene) and indene-c60 bisadduct by device optimization, *Advanced Materials* **22** (39) (2010) 4355-4358
16. Tan Z., Qian D., Zhang W., Li L., Ding Y., Xu Q., Wang F., Li Y. - Efficient and stable polymer solar cells with solution-processed molybdenum Oxideinterfacial layer, *Journal of Materials Chemistry A* **1** (3) (2013) 657-664.
17. G rtner S., Christmann M., Sankaran S., R hm H., Prinz E. M., Penth F., P utz A., T ureli A. E., Penth B., Baumst mmeler B., *et al.* - Eco-friendly Fabricationof 4 % efficient organic solar cells from surfactant-free p3ht: Icba Nanoparticledispersions, *Advanced materials* **26** (38) (2014) 6653-6657.
18. Oliveira E. F., Silva L. C., Lavarda F.C. - Modifying electronic properties of Icbathrough chemical substitutions for solar cell applications, *Structural Chemistry* **28** (2017) 1133-1140.
19. Halls J., Walsh C., Greenham N. C., Marseglia E., Friend R. H., Moratti S., Holmes A. - Efficient photodiodes from interpenetrating polymer networks, *Nature* **376** (6540) (1995) 98-500.
20. Yu G. J., Gao J. C., Hummelen F. Wudi, and A. J. Heeger - *Science* **270** (1995) 1789.

21. Peet J., Kim J. Y., Coates N. E., Ma W. L., Moses D., Heeger A. J., Bazan G. C. - Efficiency enhancement in low-bandgap polymer solar cells by Processing with alkane dithiols, *Nature materials* **6** (7) (2007) 497-500.
22. Lee J. K., Ma W. L., Brabec C. J., Yuen J., Moon J. S., Kim J. Y., Lee K., Bazan G. C., Heeger A. J. - Processing additives for improved efficiency from bulk heterojunction solar cells, *Journal of the American Chemical Society* **130** (11) (2008) 3619-3623.
23. Loew N., Komatsu S., Akita H., Funayama K., Yuge T., Fujiwara T., Ihara M. - TiO₂ as electron-extraction-layer in reverse type p3ht/icba organic solar cells, *ECS Transactions* **58** (45) (2014) 77.
24. Matavulj P., Islam M., Zivanovi C. S. - Proceedings of the international Conference on numerical simulation of optoelectronic devices, (2017) 133-134.
25. Gopalan S. A., Gopalan A. I., Vinu A., Lee K. P., Kang S. W. - A new optical lectrical integrated buffer layer design based on gold nanoparticles tethered thiol containing sulfonated polyaniline towards enhancement of solar cell performance, *Solar Energy Materials and Solar Cells* **174** (2018) 112-123.
26. Burgelman M., Nollet P., Degraeve S. - Modelling polycrystalline Semiconductorsolar cells, *Thin solid films* **361** (2000) 527-532.
27. Xu B., Sai-Anand G., Gopalan A. I., Qiao Q., Kang S. W. - Improving hotovoltaic properties of p3ht: Ic60ba through the incorporation of small molecules, *Polymers* **0** (2) (2018) 121.
28. Nithya K., Sudheer K. - Numerical modelling of non-fullerene organic solar Cell with high dielectric constant itic-oe acceptor, *Journal of Physics Communications* **4** (2) (2020) 025012.
29. Heo S. W., Lee E. J., Seong K. W., Moon D. K. - Enhanced stability in polymer solar cells by controlling the electrode work function via modification of indium tin oxide, *Solar energy materials and solar cells* **115** (2013) 123-128.
30. Loser S., Bruns C. J., Miyauchi H., Ortiz R. P., Facchetti A., Stupp S. I., Marks T. J. - A naphthodithiophene-diketopyrrolopyrrole donor molecule for efficient solution- processed solar cells. *Journal of the American Chemical Society* **133** (21) (2011) 8142-8145.
31. Li G., Yao Y., Yang H., Shrotriya V., Yang G., Yang Y. - "Solvent annealing" effect in polymer solar cells based on poly (3-hexylthiophene) and methanofullerenes, *Advanced Functional Materials* **17** (10) (2007) 1636-1644.
32. He Z., Liu F., Wang C., Chen J., He L., Nordlund D., Wu H., Russell T. P., Cao Y. - Simultaneous spin-coating and solvent annealing: Manipulating the active layer morphology to a power conversion efficiency of 9.6 % in polymer solar cells, *Materials Horizons* **2** (6) (2015) 592-597.
33. Zhu X., Lu K., Xia B., Fang J., Zhao Y., Zhao T., Wei Z., Jiang L. - Improving the performances of random copolymer based organic solar cells by adjusting the film features of active layers using mixed solvents, *Polymers* **8** (1) (2015) 4.
34. Lu L., Chen W., Xu T., Yu L. - High-performance ternary blend polymer solar cells involving both energy transfer and hole relay processes, *Nature communications* **6** (1) (2015) 7327.

35. Stylianakis M. M., Konios D., Petridis C., Kakavelakis G., Stratakis E., Kymakis E. - Ternary solution-processed organic solar cells incorporating 2D materials, *2D Materials* **4** (4) (2017) 042005.
36. Ezealigo B. N., Nwanya A. C., Simo A., Osuji R. U., Bucher R., Maaza M., Ezema F. I. - Optical and electrochemical capacitive properties of copper (i) iodide thin film deposited by silar method, *Arabian Journal of Chemistry* **12** (8) (2019) 5380-5391.
37. Liu J., Zhang Y., Liu C., Peng M., Yu A., Kou J., Liu W., Zhai J., Liu J. - Piezo-phototronic effect enhanced uv photodetector based on cui/zno double-Shell grown on flexible copper microwire, *Nanoscale research letters* **11** (2016) 1-7.

A Study on the influence of different HTL and ETL layers on the performance of a P3HTIC₆₀BA bulk heterojunction organic solar cell

C D Ramabadran^{1,2*} and K S Sudheer^{1*} 

¹Department of Physics, Opto-electronic Device Simulation Research Lab, Christ College(Autonomous), University of Calicut, Irinjalakuda, Thrissur 680125, Kerala, India

²Department of Physics, Government College, Chittur, Palakkad 678104, Kerala, India

Received: 25 May 2023 / Accepted: 27 May 2024

Abstract: In photovoltaic research, bulk heterojunction organic solar cells have garnered significant interest as light harvesters. This increased attention underscores the importance of advance research in organic solar cell development. The present study considers an organic bulk heterojunction solar cell with P3HT:IC₆₀BA as the active layer. Simulation studies are conducted using SCAPS 1D software. Initially, the software is standardized by comparing the experimental data of the solar cell structure, ITO/PEDOT:PSS/P3HT:IC₆₀BA/ZnONPS/Al with the simulated characteristics of the solar cell. Subsequently simulation research is done to evaluate the impact of different hole transport layers (HTL) and electron transport layers (ETL) on device performance. The HTL materials considered in this study include PEDOT:PSS, CuI, and Cu₂O while the ETL materials include ZnO NPs, SnO₂, TiO₂, IGZO and PCBM. According to the results, the cell structure employing Cu₂O as the HTL and IGZO as the ETL performed better than all other combinations. The cell structure is further optimized to analyze the impact of material parameters on device performance.

Keywords: Bulk heterojunction organic solar cell; Fullerene; HTL; ETL; SCAPS -1D

1. Introduction

Non-conventional energy sources have gained significant importance in today's world due to the depletion of traditional fossil fuels and the sharp rise in energy demand. In such a scenario, photovoltaics play a crucial role. The significance of organic solar cells in this perspective increases when one takes into account the production costs, stability, and complexity of inorganic silicon solar cells. Moreover, roll-to-roll printing made possible by solution processing is a feasible method for large-scale production, making it the most straightforward and economical way to fabricate solar cells. The use of dye-sensitized solar cells (DSSCs), one of the most promising methods for lowering solar cell production costs serves as a solution technique for this [1].

Small molecule organic or polymer solar cells along with Quantum dot-sensitised solar cells (QD-SSCs) have also gained popularity due to their unique properties [2–5].

Among all photovoltaic technologies, bulk heterojunction organic solar cells (BHJOSCs) are highly sought after for their potential to enable low-cost, rapid and lightweight production. Tang claimed to have developed the first effective organic photovoltaic device in 1986 [6]. In this, a BHJ structure combines materials from the donors and acceptors to form a bicontinuous interpenetrating network with large interfacial areas for efficient exciton dissociation. While Tang's friend and colleagues demonstrated the BHJ structure for all polymer blends with polymer donor and polymer acceptor for successful photogeneration [7], Heeger and colleagues first proposed the BHJ structure for polymer fullerene blends [8]. Eventually, it was shown that additives could optimize the acceptor and donor domain sizes [9, 10]. Since then the BHJ structure has become the standard for organic solar cells.

In order to improve the PCE (power conversion efficiency) of BHJ OSCs and solve the issue linked to OSC

C.D. Ramabadran and K.S. Sudheer have contributed equally to the work.

*Corresponding author, E-mail: ramabadrancd@christcollegeijk.edu.in; sudheersebastian@christcollegeijk.edu.in; sudheersebastian@yahoo.com

using poly(3-hexylthiophene): [6,6] Phenyl-C61-butyric acid methyl ester (P3HT: PCBM) mix with a lower open-circuit voltage (Voc) of 0.6 V, Li et al. used an indene-C60 bisadduct (IC₆₀BA) as the electron acceptor in BHJ OSCs [11] to achieve improved results. Meanwhile the P3HT:IC₆₀BA pair-based OSCs demonstrated a significant Voc of 0.84 V and a higher PCE [12–14]. Further research into the IC₆₀BA's capacity to generate high-performance PSCs is currently underway [15–17]. The electron and hole transport layers (ETL and HTL) are critical for improving the properties of organic solar cells by reducing carrier recombination at the surface [18–20]. One of the primary advantages of including an effective HTL and ETL in the OSC design is that it reduces the creation of undesirable space charges. Therefore it becomes evident that long-term sustainability requires finding the proper combination of transport layers for the OPV [Organic Photo voltaic] device's interface stability.

In this study, the SCAPS 1-D software [21] simulates BHJOSC with P3HT:IC₆₀BA (poly(3- hexylthiophene):indene-C60 bis adduct) as active layer. The program is standardized by comparing the simulated and experimental results [22]. Several electron transport layer (ETL) and hole transport layer (HTL) materials are utilized to simulate the structure of the solar cell. The most suitable and efficient BHJOSC design consists of a combination of cuprous oxide (Cu₂O) as the HTL and indium gallium zinc oxide (IGZO) as the ETL. The device is further optimized to analyze the effect of various material parameters on performance.

2. Device simulation methodology

Throughout the study, we used the 1D solar cell capacitance simulator SCAPS 1-D [21] software created by Marc Burgelman at the University of Gent in Belgium. SCAPS one of the best simulation tools has been utilized successfully for solar cell device modelling. Previous simulations have been carried out for fullerene and non-fullerene BHJ, perovskite, and tandem solar cells (Abdelaziz et al., 2019, 2020; Gupta and Dixit, 2018; Bahrami et al., 2019). The application is separated into multiple panels, allowing the user to alter parameters and make judgments on the outcome. For illumination we employed a 1.5AM spectrum.

The software numerically solves the generic semiconductor equations provided below by Burgelman et al [23] to create a working point solution in a steady state. This software program is based on Poisson's and continuity differential equations for holes and electrons [24–27]. The fundamental semiconductor equations like the Poisson equation, continuity equations, drift and diffusion current

equations are used to determine the electrical performance of electronic devices, shortly after applying stimulus to them. This stimulus eventually takes the form of voltages, photons, or thermal energy. This implies that we may use these equations to compute the electrical performance of the devices and obtain I-V relationships for them. Consequentially, this serves as the theoretical foundation for analyzing and interpreting the measured performance of such devices. They can also be used to evaluate and estimate the performance of new device structures and aid in the reduction of expenses regarding experimental labour, which in turn results in significant savings in both effort and expense. They can be utilized to obtain the best device structure parameters and make mathematical analysis easier. These differential equations delineate the material's local microscopic behaviour. The Poisson equation represents the local change in the electric field owing to volumetric charge density at a given point within the material [Eq. 1]. The continuity equations express the conservation of electrons and holes at a position in the material [Eqs. 2 and 3]. In this the electron and hole currents are either driven by an electric field (drift current) or a concentration gradient (diffusion current) [Eqs. 4 and 5].

$$\left(\frac{\partial \epsilon_0 \epsilon}{\partial x} \frac{\partial \Psi}{\partial x} \right) = -q \left(p - n + N_D - N_A + \frac{\rho_{def}}{q} \right) \quad (1)$$

$$\frac{-\partial J_n}{\partial x} - U_n + G = \frac{\partial n}{\partial x} \quad (2)$$

$$\frac{-\partial J_p}{\partial x} - U_p + G = \frac{\partial p}{\partial x} \quad (3)$$

$$J_n = \frac{-(\mu_n n)}{q} \frac{\partial E_{Fn}}{\partial x} \quad (4)$$

$$J_p = \frac{-(\mu_p p)}{q} \frac{\partial E_{Fp}}{\partial x} \quad (5)$$

In the above mentioned equations, the charge is denoted by q , and the electrostatic potential is denoted by Ψ . The symbols p , n , p_t , and n_t represent free holes and electrons, trapped holes, and trapped electrons respectively. N_D and N_A represent ionised donor-like and ionised acceptor-like doping concentrations. ϵ denotes absolute permittivity, while ϵ_0 denotes permittivity in a free space. The terms J_n and J_p refer to electron and hole current densities. U_n denotes the rate of electron recombination, G is the generation rate, and U_p is the rate of hole recombination. In contrast E_{Fn} and E_{Fp} are the electron and hole quasi-Fermi level values whereas n and p represent the electron and hole mobility [27].

The software is calibrated using a solar structure that includes P3HT:IC₆₀BA as the active layer, PEDOT PSS as the HTL, and ZnO NPS as the ETL [22]. Fig. 1 depicts the framework of the BHJOSC for standardizing the SCAPS

1D software. Table 1 compares the experimental and simulation values for calibrating SCAPS 1-D.

Figure 2 illustrates the general structure used for simulations with various ETL and HTL materials. Figure 3 presents the JV curves for both experimental and simulated results. Table 2 displays the physical and technological parameters of the simulated cell layers with χ , E_g , and ϵ_r representing electron affinity, bandgap energy, and relative permittivity. The electron and hole mobilities are

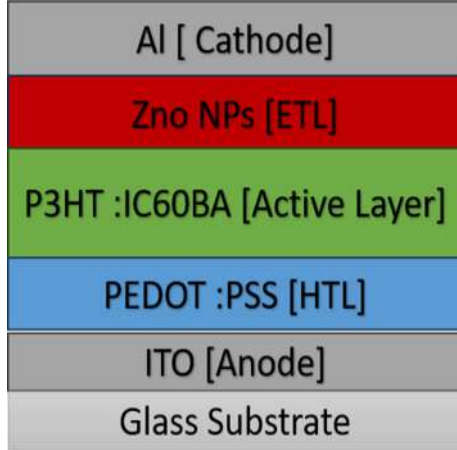


Fig. 1 Device structure of bulk heterojunction organic solar cell

Table 1 Evaluation of simulated parameters with experimental results

Parameters	Experimental	Simulated
Voc (V)	0.84	0.85
J_{sc} (mA/cm ²)	8.36	8.21
FF (%)	59	57
PCE (%)	4.1	3.97

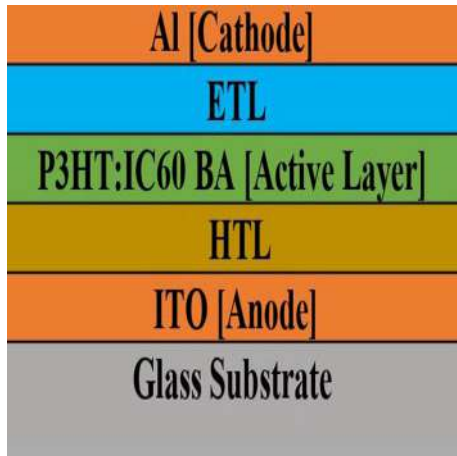


Fig. 2 Generalised structure for simulations with various ETL and HTL

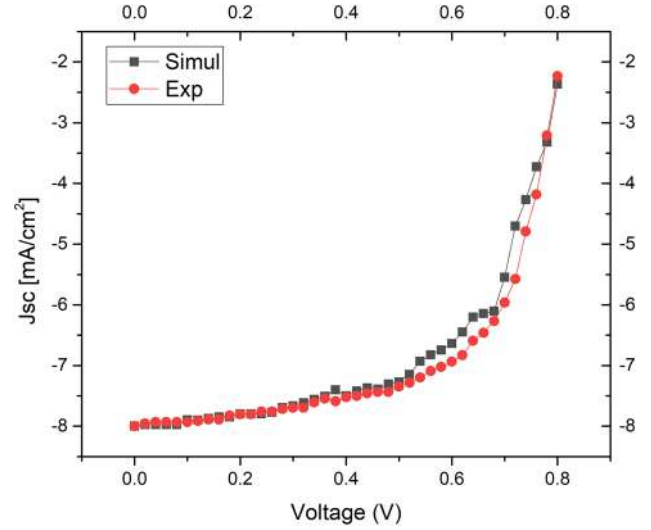


Fig. 3 Comparison of experimental and simulated JV graphs

Table 2 Simulation parameters of bulk heterojunction organic solar cell

Parameters	HTL	Active layer	ETL
Thickness [nm]	27	250	46
E_g (eV)	1.6 [25]	1.68 [31]	3.2 [14]
χ (eV)	3.4 [25]	3.880 [32]	4.6 [29]
ϵ_r	3.0 [25]	3.3 [22]	9.0 [14]
μ_n cm ² /vs	10 ¹ [25]	10 ⁷ [22]	10 ⁷ [29]
μ_p cm ² /vs	400	10 ⁷	10 ⁷ [29]
N_A (cm ⁻³)	10 ¹⁵ [25]	0 [33]	0
N_D (cm ⁻³)	0 [25]	3.2×10^{18} [31]	1.1×10^{18} [14]
N_C (cm ⁻³)	2.2×10^{15} [24]	1.6×10^{20} [32]	2.2×10^{19} [29]
N_V (cm ⁻³)	1.8×10^{18} [24]	10 ¹⁹ [33]	1.8×10^{19} [29]
N_t (cm ⁻³)	10 ¹⁵ [24]	10 ¹⁶ [31]	10 ¹⁵ [30]

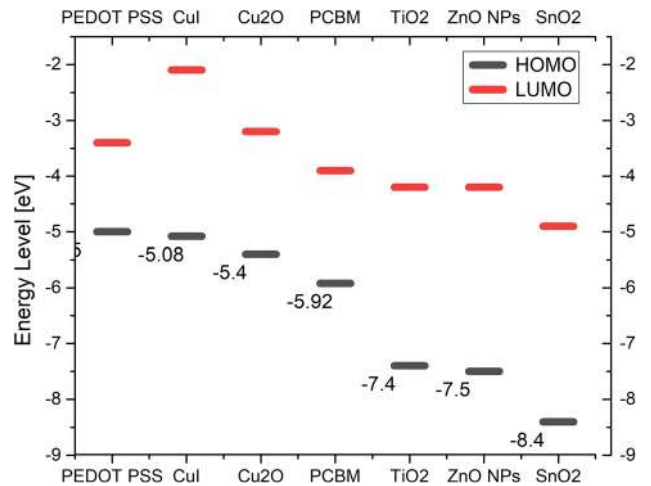


Fig. 4 Energy level diagram of HTL and ETL layers

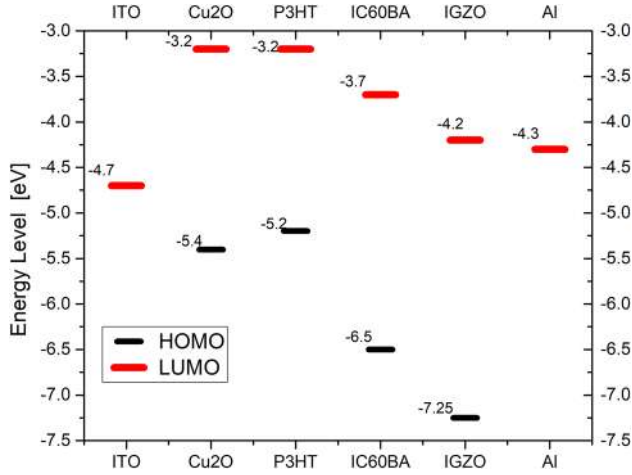


Fig. 5 Energy level diagram of optimised organic solar cell structure

represented as (μ_n) and (μ_p) . N_D and N_A denote donor and acceptor density. N_v and N_c shows the effective density of states in the valence and conduction bands, while N_t represents the defect density [25]. The anode and cathode work functions are set at 4.8 and 4.2 eV respectively. The surface recombination velocity for electrons on the anode is set at 10^7 cm/s, whereas for holes it is fixed at 10^5 cm/s. In the cathode electrons have a surface recombination

velocity of 10^5 and holes have a value of 10^7 centimeters per second. The thermal velocity is set at 10^7 cm/s, and the capture cross-section of both holes and electrons are set at 1×10^{-25} cm^2 across all layers. The primary output parameters of a solar cell are V_{oc} (open circuit voltage), J_{sc} (short circuit current), FF (fill factor), and PCE (power conversion efficiency). The needed material parameters for simulation and absorption spectra are obtained from the literature [14, 19, 20, 22, 28–38].

The solar cell structure is simulated with different HTL and ETL layers, and the most efficient combination is chosen. The thickness of HTL and ETL is maintained at 27 and 46 nm, respectively. The HTL layers considered are Poly (3,4-ethylene dioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS), Copper Iodide (CuI), Cuprous oxide (Cu_2O), and the ETL layers considered are Zinc oxide nano particles (ZnO NPs), Phenyl-C61-butyric acid methyl ester (PCBM), Tin dioxide (SnO_2), Titanium dioxide (TiO_2), and Indium gallium zinc oxide (IGZO). The device parameters and absorption profiles of these layers were taken from the literature by Nithya.K.S and Sudheer.K.S [27], Md. Samiul Islam Sadek et al. [12, 39, 40]. Figure 4 depicts the energy level diagram for the various HTL and ETL layers under consideration.

Table 3 Device parameters of HTL layers

Parameters	PEDOT: PSS	CuI	Cu_2O
Thickness [nm]	27	27	27
Eg (eV)	1.6 [27]	2.98 [27]	2.17 [39]
χ (eV)	3.4 [27]	2.1 [27]	3.2 [39]
ϵ_r	3.0 [27]	6.5 [27]	7.1 [39]
μ_n (cm^2/vs)	1.0	1.69×10^{-4} [27]	200 [39]
μ_p (cm^2/vs)	4×10^2 [27]	1.69×10^{-4} [27]	800 [39]
N_C (cm^{-3})	2.2×10^{15} [27]	10^{22} [27]	2.5×10^{18} [39]
N_V (cm^{-3})	1.8×10^{19} [27]	10^{22} [27]	1.8×10^{19} [39]
N_t (cm^{-3})	10^{15} [27]	10^{15} [27]	10^{14} [3]

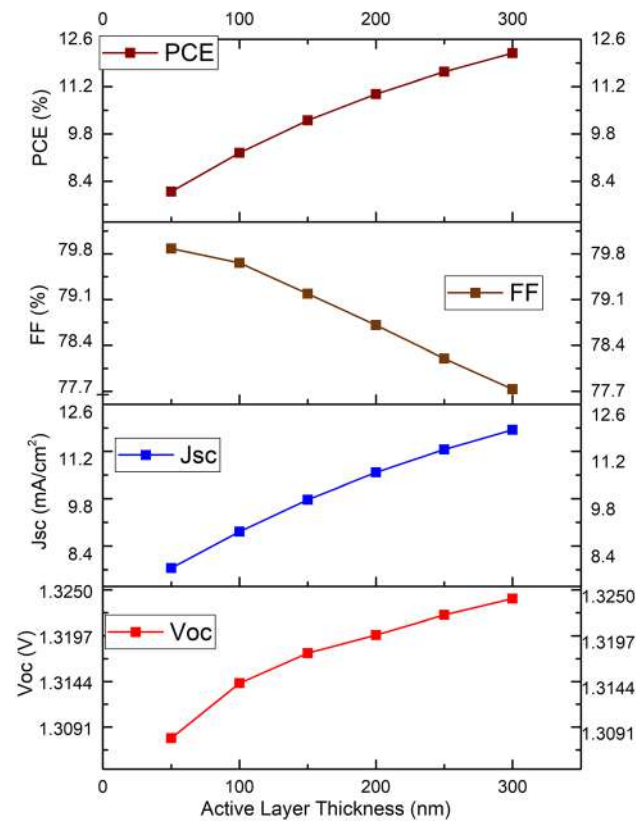
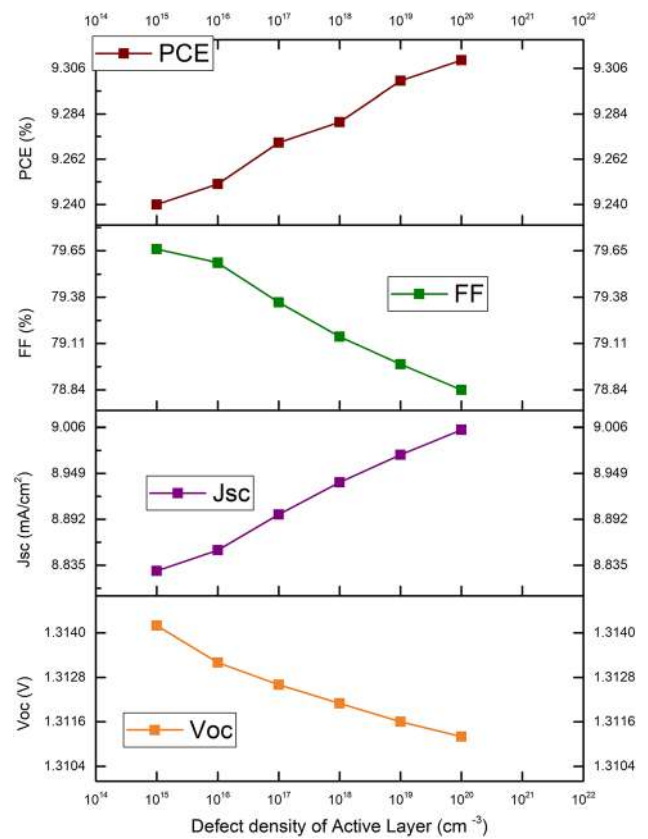
Table 4 Device parameters of ETL layers

Parameters	ZnO NPs	PCBM	SnO_2	TiO_2	IGZO
Thickness [nm]	46	46	46	46	46
Eg (eV)	3.2 [14]	1.9 [41]	3.6 [27]	3.2 [27]	3.05 [27]
χ (eV)	4.6 [29]	3.9 [41]	4.4 [27]	4.1 [27]	4.0 [27]
ϵ_r	9.0 [14]	4.0 [41]	9.0 [27]	9.0 [27]	9.0 [27]
μ_n (cm^2/vs)	2×10^2 [14]	0.5 [27]	240 [27]	2.0 [27]	100 [27]
μ_p (cm^2/vs)	5.0	1.5×10^{-2} [41]	25 [27]	1.0 [27]	2.5 [27]
N_C (cm^{-3})	2.2×10^{19} [29]	1.8×10^{20} [41]	4.36×10^{18} [27]	2.2×10^{18} [27]	10^{19} [27]
N_V (cm^{-3})	1.8×10^{19} [29]	2.2×10^{21} [27]	2.52×10^{19} [27]	10^{19} [27]	10^{19} [27]
N_t (cm^{-3})	10^{15} [30]	10^{14} [41]	10^{15} [27]	10^{15} [39]	10^{14} [27]

Table 5 Device performance of OSC with different combinations of HTL and ETL

HTL	ETL	Voc [Volts]	Jsc [mA/cm ²]	FF %	PCE %
PEDOT:PSS	SnO ₂	0.9596	9.30	59.98	5.35
	TiO ₂	0.8750	9.50	63.48	5.27
	IGZO	0.9646	9.78	60.26	5.68
	ZnO NPs	0.8546	8.20	56.65	3.97
	PCBM	0.9798	12.46	59.58	7.27
CuI	SnO ₂	0.8797	7.63	66.20	4.45
	TiO ₂	0.6467	7.86	59.14	3.00
	IGZO	0.9104	12.79	62.06	7.22
	ZnO NPs	0.7586	6.47	61.72	3.03
	PCBM	0.6468	7.87	59.13	3.01
Cu ₂ O	SnO ₂	1.3190	10.75	78.34	11.11
	TiO ₂	1.3213	10.93	78.47	11.34
	IGZO	1.3221	11.26	78.20	11.64
	ZnONPs	1.1985	12.13	72.75	10.57
	PCBM	1.3261	11	78.41	11.41

Bold values indicate the combinations used for the simulation study and the combination expressed in optimized performance at best [Cu₂O as HTL and IGZO as ETL]


Fig. 6 Influence of active layer thickness on cell performance

Fig. 7 Influence of defect density of active layer on cell performance

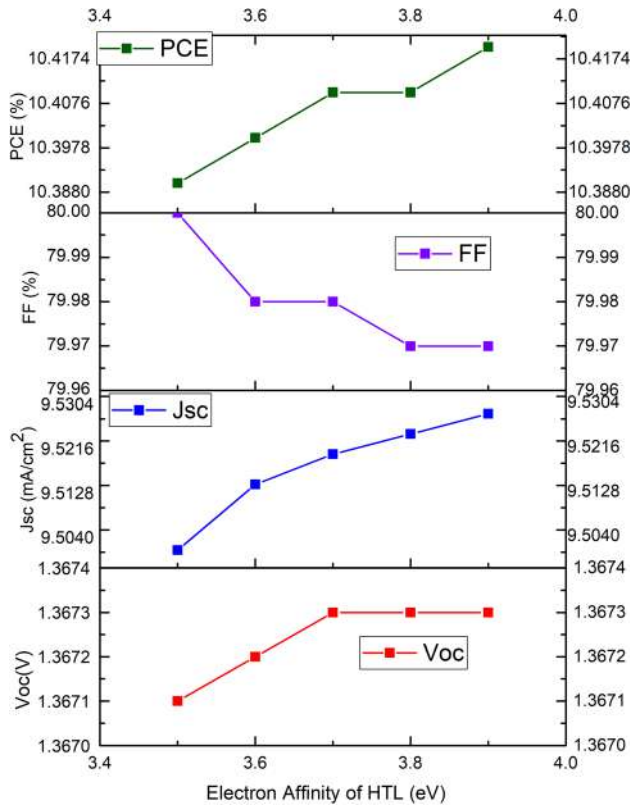


Fig. 8 Influence of electron affinity of HTL on cell performance

3. Results and discussion

The organic solar cell fuelled with P3HT:IC₆₀BA as the active layer is simulated with various HTL and ETL layers. As a result of these simulations, the optimal cell structure was identified and created using Cu₂O as the hole transport layer (HTL) and IGZO as the electron transport layer (ETL). The energy level diagram of the optimized organic solar cell structure is given in Fig. 5. The following sections outline the various strategies employed to simulate the generalized structure to develop an optimal model.

3.1. Simulation studies of P3HT:IC₆₀BA organic solar cell with different HTL and ETL Layers

During the study, different HTL and ETL materials are employed to simulate bulk heterojunction organic solar cell with P3HT:IC₆₀BA as active layer. The functional properties of the organic cell are examined for each combination. Tables 3 and 4 list the simulation parameters for the various HTL and ETL layers.

Table 5 shows the output parameters for the various HTL and ETL layer combinations. We found that the optimized cell structure with Cu₂O as HTL and IGZO as ETL outperformed the other combinations. The structure yielded 1.3221V as Voc, 11.26mA/cm² as Jsc, 78.20% as FF, and

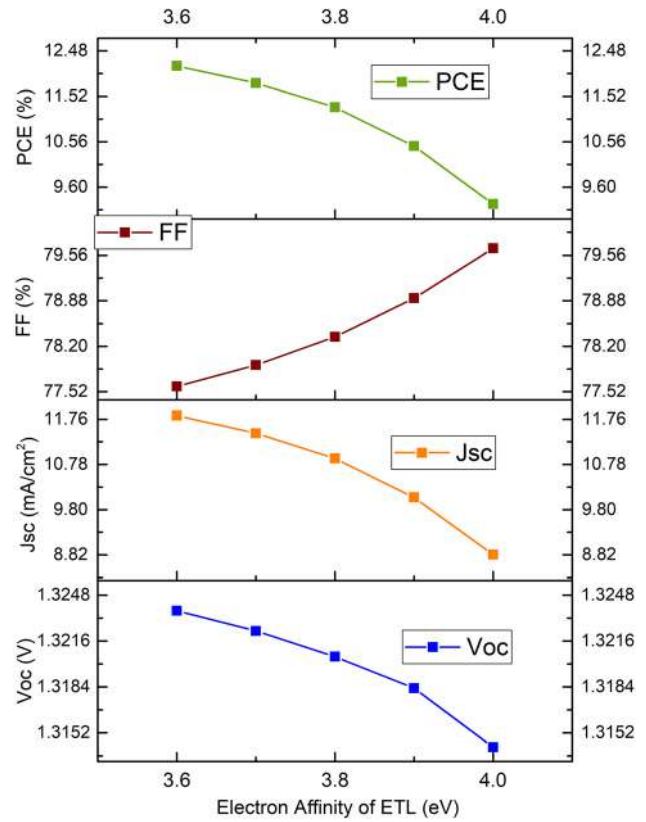


Fig. 9 Influence of electron affinity of ETL on cell performance

11.64% as PCE as outputs. The performance is determined by the band alignment of the materials used in the cell structure as well as other device-specific parameters.

Using Cu₂O as HTL and TiO₂ as ETL results in the highest FF of 78.47%. In the coupling of PEDOT:PSS as the HTL and ZnONPs as the ETL, FF has a minimum value of 56.65%. The power conversion efficiency varies from 3% for CuI as HTL and TiO₂ as ETL to a maximum of 11.64% for Cu₂O as HTL and IGZO as ETL. The optimized organic cell structure using Cu₂O as HTL and IGZO as ETL is further examined after changing various cell-related variables. Simulation studies on the organic solar cell construction employing P3HT:IC₆₀BA as the active layer aid in achieving optimal results under various circumstances.

4. Optimization of the cell structure ITO/Cu₂O/P3HTIC₆₀BA/IGZO/Al

4.1. Influence of active layer thickness on cell performance

As shown in Fig. 6, the active layer thickness is increased from 50 nm to 300 nm, and the effect on cell parameters is

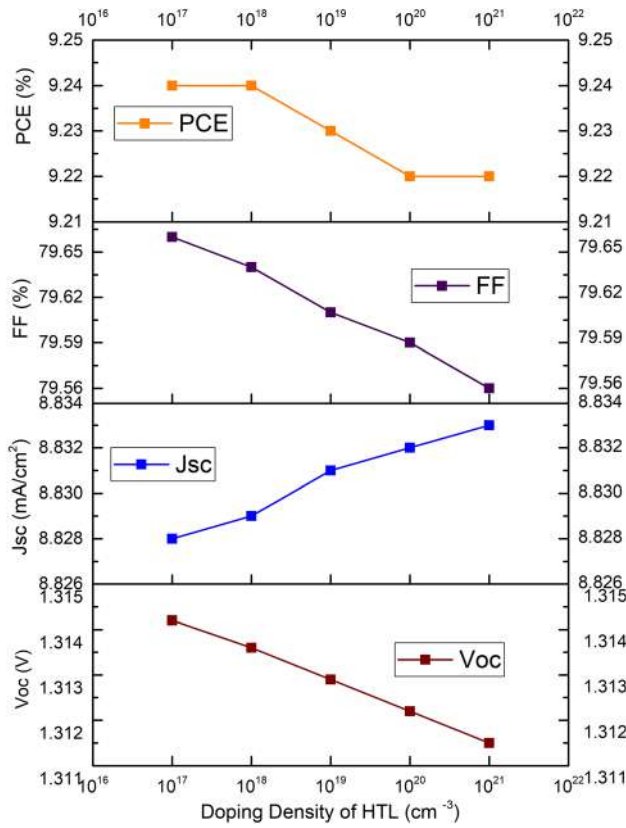


Fig. 10 Influence of doping density of HTL on cell performance

investigated. Jsc reaches a maximum value of 11.84 mA/cm² at a thickness of 300 nm, whereas Voc reaches a maximum value of 1.3240V at the same thickness. The FF reduces with thickness and peaks at 79.88% at 50 nm, while efficiency increases with thickness and peaks at 12.19% at 300 nm. These data show how the thickness of the active layer impacts light absorption and thus solar cell efficiency.

The difference between the HOMO level of the donor and the LUMO level of the acceptor determines Voc values. To promote exciton dissociation at the interface, a thick layer allows for increased electron and hole trapping in the film as well as considerable photon absorption which results in a rise in Voc with thickness [42].

In this simulation, Jsc increases with active layer thickness because nearly all electron-hole pairs generated by photons are effectively collected at the electrode before recombination. This indicates a higher production of charge carriers. The fill factor (FF) diminishes in thicker films due to uneven charge extraction rates for holes and electrons [43]. This demonstrates that efficiency will increase because a thicker active layer allows for the formation of a continuous network throughout, producing the

highest number of dissociated excitons and delaying charge recombination due to Coulombic attraction at the interface.

4.2. Effect of active layer defect density on cell performance

Figure 7 demonstrates how the active layer defect density influences the functionality of organic cells. The graph shows that Voc declines with defect density and peaks at 10¹⁵cm⁻³. Jsc achieves its peak value of 9.003mA/cm² at a defect density of 10²⁰cm⁻³.

While PCE is found to grow with defect density and finds an optimal value of 9.31% efficiency for defect density of 10²⁰cm⁻³, FF is found to decrease with defect density of the active layer and peaks at 10¹⁵cm⁻³ with a value of 79.66%.

The defect concentrations in the absorber layer also result in faster film deterioration, decreased stability, and poor device performance due to increased pinhole generation and recombination [44]. More defects result in shorter charge carrier diffusion lengths and an increase in recombination carriers in the absorber layer, hence defect density has a direct impact on efficiency [45].

4.3. Effects of HTL's electron affinity on cell performance

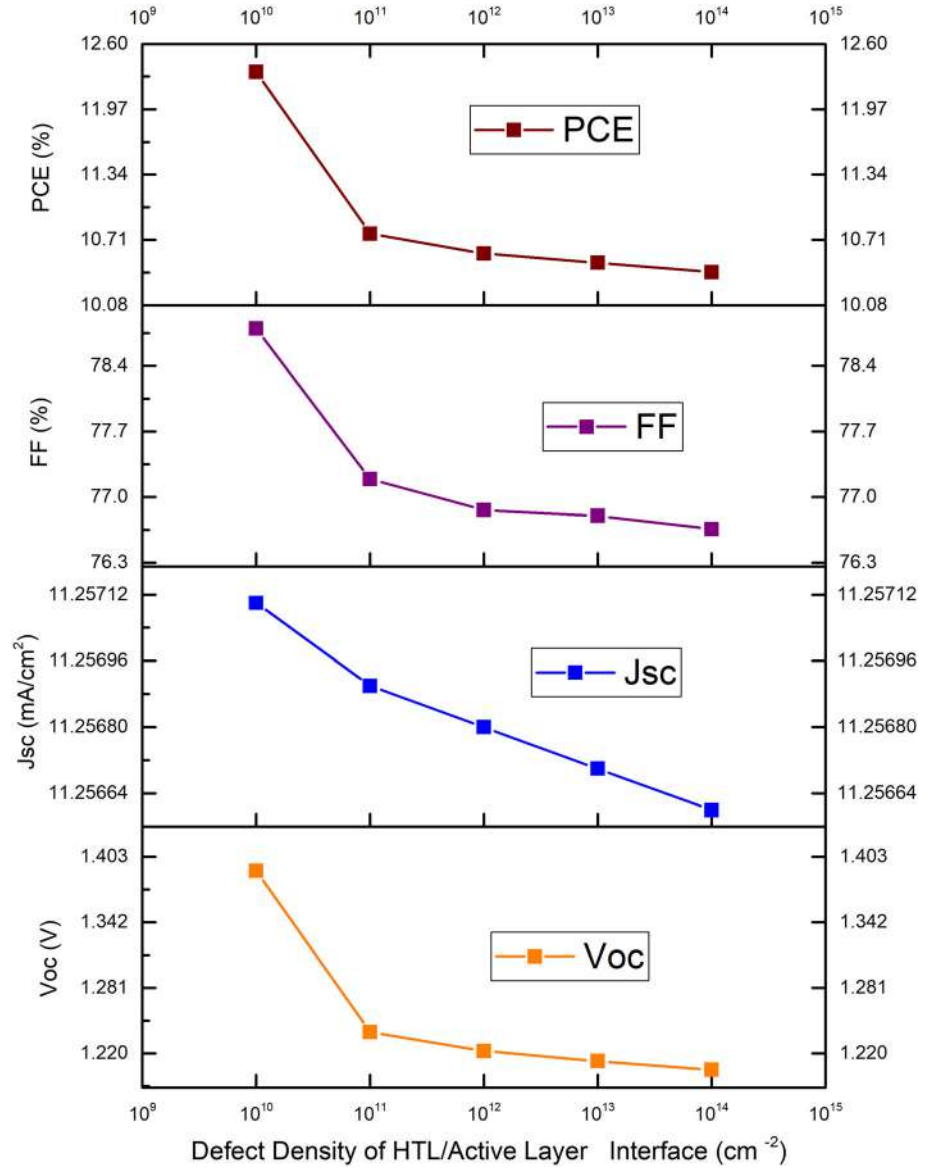
Figure 8 illustrates the influence of the HTL's electron affinity on cell performance. The electron affinity is closely related to the lowest unoccupied molecular orbital (LUMO) of the hole transport layer representing the energy (measured in eV) required to move a free electron from the lowest level of that orbital to the vacuum level.

According to the study, Voc, Jsc, and PCE vary similarly, while FF behaves differently. The optimized values of Voc and Jsc were discovered to be 1.3673V and 9.527 mA/cm² at 3.9eV. At 3.5 eV, the optimal FF value is 80%, while at 3.9 eV, the optimal PCE value is 10.42%.

4.4. Electron affinity of ETL's impact on cell performance

Throughout the research the impacts of changing ETL's electron affinity from 3.6 to 4 eV on cell performance were explored. FF has a maximum value of 79.66% at 4 eV, Voc has an optimal value of 1.3237V at 3.6 eV electron affinity, Jsc has an optimum value of 11.842 mA/cm², and PCE has an optimized value of 12.16% at 3.6 eV. Figure 9 shows the variations.

Fig. 11 Influence of defect density of HTL/Active layer interface on cell performance



4.5. Influence of doping density of HTL on cell performance

The significance of the doping density of HTL on cell performance is stated here. When the doping density of HTL was increased from 10^{17} to 10^{21} cm⁻³, Voc, FF, and PCE fell accordingly. The greatest values of these output parameters found at a doping density of 10^{17} cm⁻³ are 1.3142V, 79.66%, and 9.24% respectively. Jsc increased by 8.833 mA/cm² at the optimal doping density of 10^{21} cm⁻³. Fig. 10 shows the relevant graphs. The electric field intensity at the HTL/absorber interface is altered by the doping density of HTLs, which regulates the performance of the solar cell. The device gets more conductive as the HTL doping density increases, lowering the series resistance [24, 30].

4.6. Effects of HTL/active layer interface defect density on cell performance

The research testifies that changing the defect density of the HTL/Active Layer Interface from 10^{10} to 10^{14} cm⁻² revealed a drop in Voc, Jsc, FF, and PCE. We got values of 1.3899V, 11.2571 mA/cm², 78.7% and 12.33% respectively for output parameters when the defect density becomes 10^{10} cm⁻². Figure 11 shows the comparable graph. The rate of change of fill factor is slower than the rates at which Jsc and Voc are dropping, resulting in a significantly lower PCE at output [27]. At the HTL/Active interface, output parameters steadily decrease as defect density increases.

Fig. 12 Influence of defect density active layer/ETL interface on cell performance

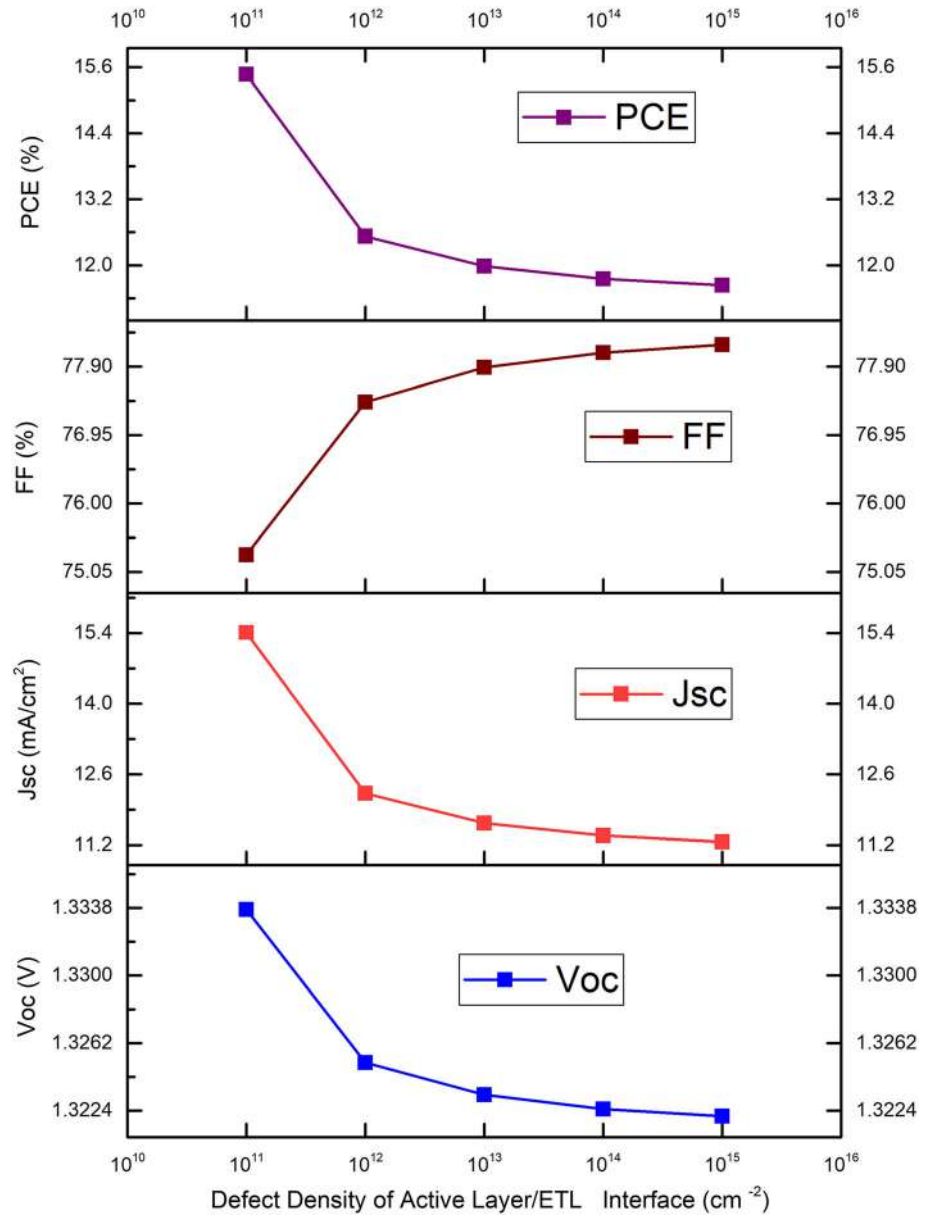


Table 6 Optimized material parameters

Parameters	HTL	Active layer	ETL
Doping density [cm^{-3}]	10^{17}	10^{16}	10^{20}
Thickness [nm]	30	300	46

Table 7 Comparison of organic solar cell with standard and optimized designs

Parameters	Initial	Optimised
Voc [V]	0.84	1.1873
Jsc [mA/cm^2]	8.36	22.094
FF [%]	59	53.74
PCE [%]	3.9	14.10

4.7. Effects of active Layer/ETL interface defect density on cell performance

Changing the defect density of the Active Layer/ETL Interface from 10^{11} to $10^{15} cm^{-2}$ resulted in a drop in Voc, Jsc, and PCE, while FF is increased. At a defect density of

$10^{11} cm^{-2}$, the best values for Voc, Jsc, and PCE are 1.3337V, 15.411mA/cm², and 15.47% respectively. The best FF result was 78.20% at $10^{15} cm^{-2}$. Figure 12 depicts

the equivalent graphs. Increased defect density at interfaces increases the possibility of trap recombination resulting in device deterioration. A lower interface defect density will result in better output performance [27].

Table 6 displays the improved material properties. Table 7 compares the device's performance for the initial and upgraded values. The simulation produces better results than the experimental structure.

5. Conclusions

The work emphasizes the importance of selecting appropriate charge transport layers to design highly efficient bulk heterojunction organic solar cells ensuring favourable interfacial stability and improved device performance. This study examines a bulk heterojunction organic solar cell with P3HT:IC₆₀BA as the active layer complemented by various HTL and ETL layers. The design showcasing cuprous oxide as the HTL (hole transport layer) and Indium gallium zinc oxide as the ETL (electron transport layer) respectively attains the highest power conversion efficiency (PCE). Ensuring compatible charge transport layers facilitates the seamless transition of charge carriers towards their respective electrodes. It's crucial to have a relatively low energy gap for the active layer to ensure smooth and efficient absorption of incident photons. The updated design is further refined to meticulously analyze the impact of various material parameters on the output performance. The optimized parameters for the new structure are $V_{oc} = 1.1873V$, $J_{sc} = 22.09 \text{ mA/cm}^2$, $FF = 53.74\%$, and $PCE = 14.10\%$. The findings hold significant promise for the OPV community, offering valuable insights for designing highly efficient bulk heterojunction organic solar cells in the near future.

Acknowledgements The authors would like to acknowledge Pr.M.Burgelman, University of Gent, Belgium for providing SCAPS 1-D software for the simulation study

References

1. N Z S M G Ghadiri, E Taghavinia and M Moser *Nano letters* **10** 1632 (2010)
2. F Ghoreishi, V Ahmadi and M Samadpour *Journal of Power Sources* **271** 195 (2014)
3. M Riede, D Spoltore and K Leo *Advanced Energy Materials* **11** 2002653 (2021)
4. C Deibel and V Dyakonov *Reports on Progress in Physics* **73** 096401 (2010)
5. A J Heeger *Chemical Society Reviews* **39** 2354 (2010)
6. C W Tang *Applied physics letters* **48** 183 (1986)
7. Halls et al *Nature* **376** 6540 (1995)
8. Yu et al *Science* **270** 1789 (1995)
9. Peet et al *Nature materials* **6** 497 (2007)
10. Lee et al *Journal of the American Chemical Society* **130** 3619 (2008)
11. He et al *Journal of the American Chemical Society* **132** 1377 (2010)
12. Loew et al *ECS Transactions* **58** 77 (2014)
13. S G S Ganesamoorthy and P Ramasamy *Solar Energy Materials and Solar Cells* **161** 102 (2017)
14. Gopalan et al *Solar Energy Materials and Solar Cells* **174** 112 (2018)
15. Huang et al *Nature Photonics* **7** 479 (2013)
16. Venkatesan et al *Nano Energy* **12** 457 (2015)
17. Nemnes et al *Applied Surface Science* **424** 264 (2017)
18. Kim et al *Applied Physics Letters* **91** (2007)
19. Heo et al *Solar energy materials and solar cells* **115** 123 (2013)
20. Irwin et al *Proceedings of the National Academy of Sciences* **105** 2783 (2008)
21. M Burgelman, P Nollet and S Degraeve *Thin solid films* **361** 527 (2000)
22. Xu et al *Polymers* **10** 121 (2018)
23. Burgelman et al University of Ghent: Ghent, Belgium (2016)
24. Abdelaziz et al *Optical Materials* **91** 239 (2019)
25. K Nithya and K Sudheer *Journal of Physics Communications* **4** 025012 (2020)
26. Raoui et al *Solar Energy* **193** 948 (2019)
27. K Nithya and K Sudheer *Optical Materials* **123** 111912 (2022)
28. K A P Omer, Bushra Mohamed and Almantas, in, 37th IEEE photovoltaic specialists conference *IEEE* **2011** 000734 (2011)
29. Li et al *Advanced Functional Materials* **17** 1636 (2007)
30. He et al *Materials Horizons* **2** 592 (2015)
31. Zhu et al *Polymers* **8** 4 (2015)
32. Lu et al *Nature communications* **6** 7327 (2015)
33. Stylianakis et al *2D Materials* **4** 042005 (2017)
34. Ezealigo et al *Arabian Journal of Chemistry* **12** 5380 (2019)
35. Liu et al *Nanoscale research letters* **11** 1 (2016)
36. O Madelung Springer Science & Business Media, (2004)
37. E Jouad et al *Solar Energy Materials and Solar Cells* **141** 429 (2015)
38. M I Hossain, F H Alharbi and N Tabet *Solar Energy* **120** 370 (2015)
39. Sadek et al *Journal of Engineering Studies and Research* **1** 25 (2021)
40. N D Lam *Engineering Research Express* **2** 025033 (2020)
41. Raj et al *Materials Today: Proceedings* **47** 1564 (2021)
42. Abdelaziz et al *Solar Energy* **211** 375 (2020)
43. Lin et al *Solar energy materials and solar cells* **122** 264 (2014)
44. S Rai, B Pandey and D Dwivedi *Optical Materials* **100** 109631 (2020)
45. L Et-taya, T Ouslimane and A Benami *Solar Energy* **201** 827 (2020)

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

RESEARCH ARTICLE | DECEMBER 15 2023

Study of influence of inorganic Cul hole transport layer on bulk heterojunction P3HT:IC60BA polymer solar cells using SCAPS 1-D

C. D. Ramabadran ; K. S. Sudheer



AIP Conf. Proc. 2901, 040001 (2023)

<https://doi.org/10.1063/5.0178644>



CrossMark



APL Energy
First Articles Online!
Read Now

 AIP
Publishing

Study of Influence of Inorganic CuI Hole Transport Layer on Bulk Heterojunction P3HT:IC60BA Polymer Solar Cells Using SCAPS 1-D

C D Ramabadran ^{1, 2 a)}, K S Sudheer ^{1 b)}

¹Opto-electronic device simulation research lab, Department of Physics,
Christ College (Autonomous), University of Calicut, Irinjalakuda, Kerala - 680125, India
²Govt. College, University of Calicut, Chittur, Palakkad, Kerala, India

^{a)} Corresponding Author: ramabadrancd@christcollegeijk.edu.in

^{b)} sudheersebastian@christcollegeijk.edu.in

Abstract: Bulk heterojunction Polymer solar cells [BHJPSCs] are considered as a promising option in the field of organic solar cells due to their features like low cost, light weight, ease in fabrication and roll-to-roll compatibility. In this work we considered P3HT:IC60BA based BHJ PSC and studied the influence made by CuI Hole transport layer on the solar cell parameters. CuI is preferred over PEDOT:PSS hole transport layer which is commonly used due to its features like wider band gap, low cost and high stability. On optimization, the new structure gives Voc of 1.2170 V, Jsc of 6.9243 mA/cm², Fill factor of 73.95 %, Power conversion efficiency of 6.23 %. The results are really encouraging and promising.

INTRODUCTION

In Photovoltaics, the overall performance of Organic solar cells has progressed a lot in current years[1-4]. Bulk Heterojunction Polymer Solar Cells [BHJ PSCs] are one off the alternatives withinside the subject of nonconventional energy sources due to their features like low cost, ease of fabrication, light weight and easily availability [5-7]. Hole Transport Layer [HTL] performs an essential function in the performance of a BHJ PSCs. Usually PEDOT:PSS is considered as HTL due to its features like high conductivity, transparency and high work function [8-9], it's instability and influence of moisture and electron mobility are not consistent [10-13]. Hole transport Layer has an important role in taking holes to the respective electrodes while blocking electrons. CuI as a hole transport layer for BHJ solar cells is shown to have greater performance than the traditional structure with PEDOT:PSS as HTL [14].

The BHJ PSC with P3HT:IC60BA as Active Layer has shown a high Voc of 0.84 V and a higher PCE [15-17]. So far, no computational research were carried out to for the usage of CuI as HTL in BHJ PSC with P3HT:IC60BA as Active Layer and simulation research is a beneficial for developing a high-overall performing BHJ PSC in the future.

In this study, we looked at the SCAPS 1-D software [35] with an illumination of AM 1.5 G spectrum. SCAPS 1-D is used to simulate BHJ PSC with P3HT:IC60 BA blend as active layer, CuI serves as the hole transport layer, and ZnO Nanoparticles as the electron transport layer (ETL). Numerous forms of solar cells, including polymer solar cells, have been modelled using SCAPS. The software is calibrated by experimental paper on BHJ PSC [18] and device modelling is used to investigate how CuI affects the calibrated BHJ PSC performance.

Device Structure and Modelling

SCAPS 1D software is standardized by successfully simulating the polymer solar cell structure [18] given in Fig 1. The simulated output matches the experimental findings for BHJ PSC [18] as given in Table 1. This

demonstrates that the simulation parameters are identical to the device's real-world values, and the software is fully calibrated for simulating BHJ PSC.

TABLE 1. Evaluation of simulated parameters with experimental results

Parameters	Experimental	Simulated
Voc (V)	0.84	0.955
Jsc (mA/cm ²)	8.36	6.280
FF (%)	65.0	69.90
PCE (%)	4.22	4.19

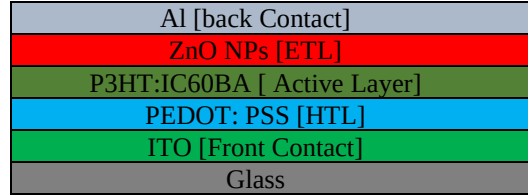
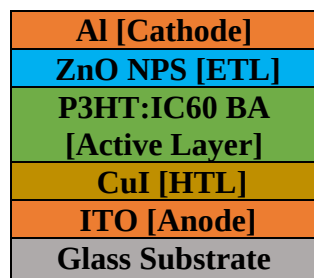


FIGURE 1. P3HT:IC60BA Bulk Heterojunction Polymer Solar Cell Device Structure

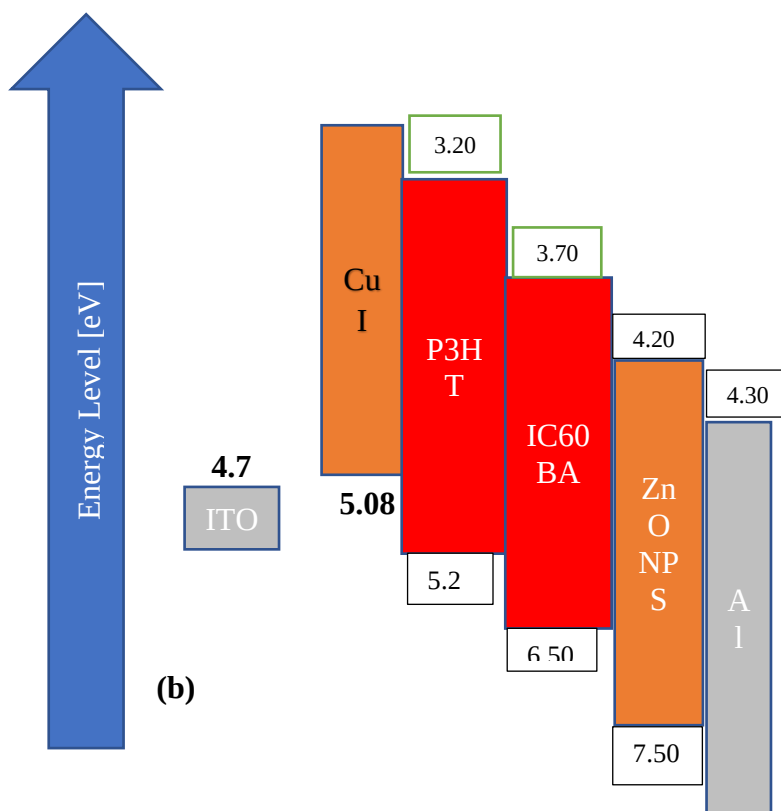
Bulk heterojunction structure [Fig 2a] of polymer solar cell using the Glass/ITO/CuI/P3HT:IC60BA/ZnO NPs/Al is simulated using SCAPS 1-D. Figure 2b depicts the energy band alignment. At the Active/CuI interface, the valence band offset is +0.12 eV, allowing holes to readily travel to the back metal contact, while the conduction band offset at the ZnO NPs/Active interface is +0.5 eV, allowing produced photoelectrons to migrate to the front contact.

A summary of the configuration's simulation parameters is provided in Table 2. The parameters provided: Energy bandgap, relative permittivity, electron affinity, electron and hole mobilities, and defect density are all represented by the symbols E_g , ϵ_r , μ_n , μ_p , and N_t , respectively. The effective densities of the conduction band and valence band are N_C and N_V , while the densities of the acceptor and donor materials are N_A and N_D , respectively [33]. All layers have the same values for the parameters that aren't listed in the table. The typical energy of the neutral gaussian distribution defect is chosen to 0.1 eV. The electron and hole capture cross sections are adjusted to $9 \times 10^{-15} \text{ cm}^2$ and the thermal velocity of both carriers is fixed at 10^7 cm/s . [34]. Two defect interfaces (HTL/Active and ETL/Active) are defined at a concentration of 10^9 cm^{-2} . The work function of the front electrode is set to 4.7 eV, while that of the back electrode is at 4.125 eV. The absorption profile of active layer and HTL were obtained from literature [18]. The simulation does not take into account optical reflectance, and to optimise the numerical parameters used in the study, we used the control variable method.

We determined the solar cell output parameters by employing the parameters listed in Table 2 as open circuit voltage (Voc), short circuit current density (Jsc), fill factor (FF), and power conversion effectiveness (PCE). Figure 3 illustrates the J-V curve. By refining the characteristics of the polymer solar cells, the calibrated structure is used to create BHJ PSC with inorganic CuI as the HTL.



(a)



(b)

FIGURE 2. (a) Simulated cell structure (b) Energy Level band diagram

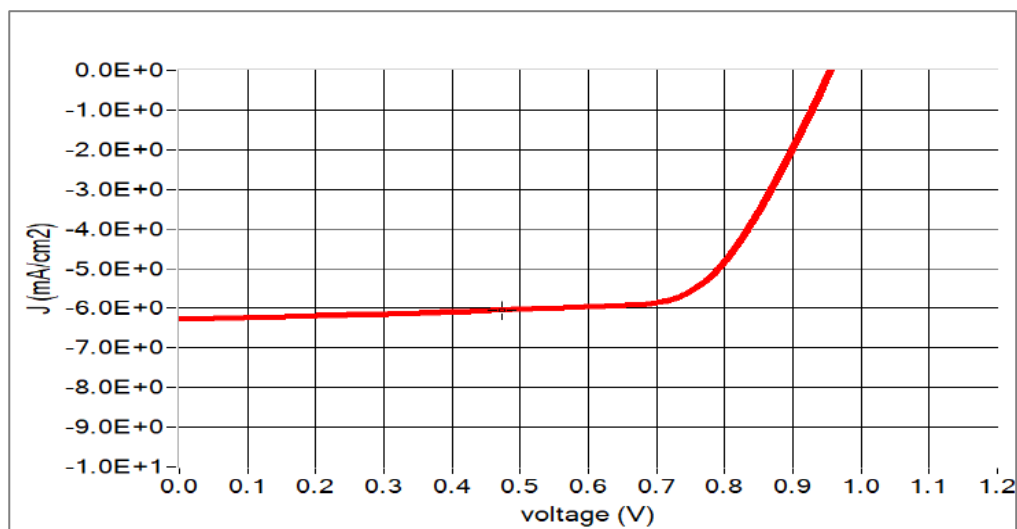


FIGURE 3. Simulated J-V Curve

TABLE 2. Simulation Parameters used

Parameters	HTL	Active layer	ETL
Thickness [nm]	40	250	46
E _g (eV)	2.98 [27]	1.68 [24]	3.2 [21]
χ (eV)	2.1 [28]	3.880 [25]	4.6 [22]
ε _r	6.5 [29]	3.3 [18]	9 [21]
μ _n (cm ² /vs)	1 x 10 ⁷	1 x 10 ⁷ [18]	1 x 10 ⁷ [22]
μ _p (cm ² /vs)	1 x 10 ⁷ [30]	1 x 10 ⁷ [18]	1 x 10 ⁷ [22]
N _A (cm ⁻³)	2x10 ¹⁸ [31]	0 [26]	0 [23]
N _D (cm ⁻³)	0	3.2x 10 ¹⁸ [24]	1.1 x 10 ¹⁸ [21]
N _C (cm ⁻³)	1x10 ²²	1.6x 10 ²⁰ [25]	2.2x10 ¹⁹ [22]
N _V (cm ⁻³)	1x10 ²² [32]	1x10 ¹⁹ [26]	1.8x10 ¹⁹ [22]
N _t (cm ⁻³)	1x10 ¹⁵	1x10 ¹⁶ [24]	1 x 10 ¹⁵ [23]

RESULTS AND DISCUSSION

Active layer thickness's impact

A polymer solar cell's output performance is significantly influenced by the thickness of its active layer. As illustrated in Fig. 4, the output is examined when the active layer thickness is changed from 50 to 400 nm. J_{sc}, PCE and Voc all increase when the active layer is 400 nm thick, reaching maximum values of 8.24487 mA/cm², 4.13 %, and 0.8298 V. Increased thickness may be responsible for this by boosting light absorption and photogenerated carriers. As thickness increases, more photons are absorbed and hence PCE increases. J_{sc} rises because of a higher likelihood of exciton dissociation or a lower rate of recombination of non-geminate charge couples. When the thickness is increased, FF first rises before beginning to fall. The maximum value of 62.29 % occurs at a thickness of 150 nm. This could be due to the cell's series resistance increasing with thickness

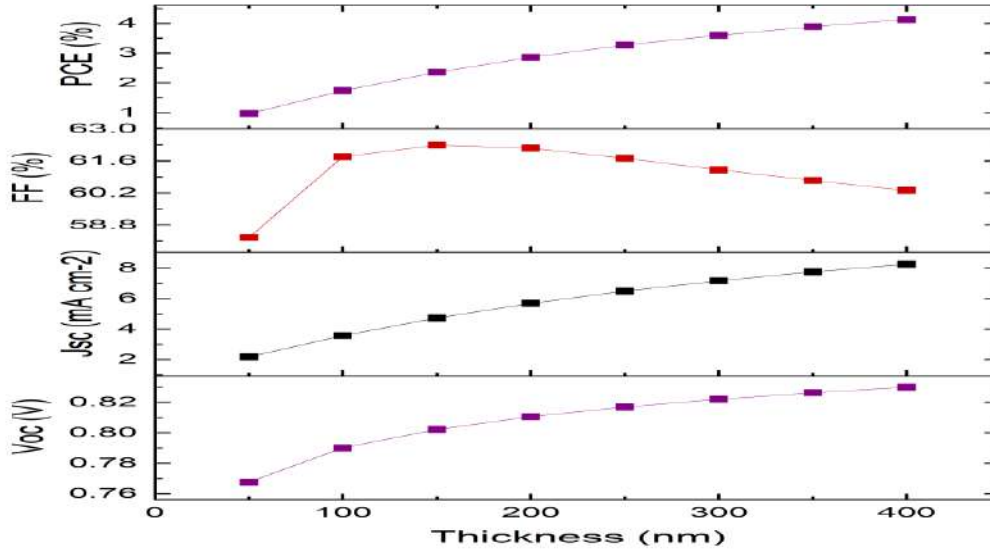


FIGURE 4. Active layer thickness's impact on output parameters

Effect of HTL (CuI) properties

On the thickness of HTL

The output of BHJ PSC is heavily influenced by the hole transporting layer. Better hole transport and charge collection at the electrodes are made possible by a superior HTL selection. On solar cell performance, the effects of HTL layer [CuI] parameters including thickness, doping concentration, electron affinity, and mobility were examined. The HTL layer thickness was increased from 50 to 400 nm, and Fig. 5. illustrates the impact on output parameters. From the graph we find that Voc, Jsc and PCE increases, while for FF it is found to decrease initially with HTL thickness and then increases. The highest possible Voc, Jsc and PCE occurs at a thickness of 250nm

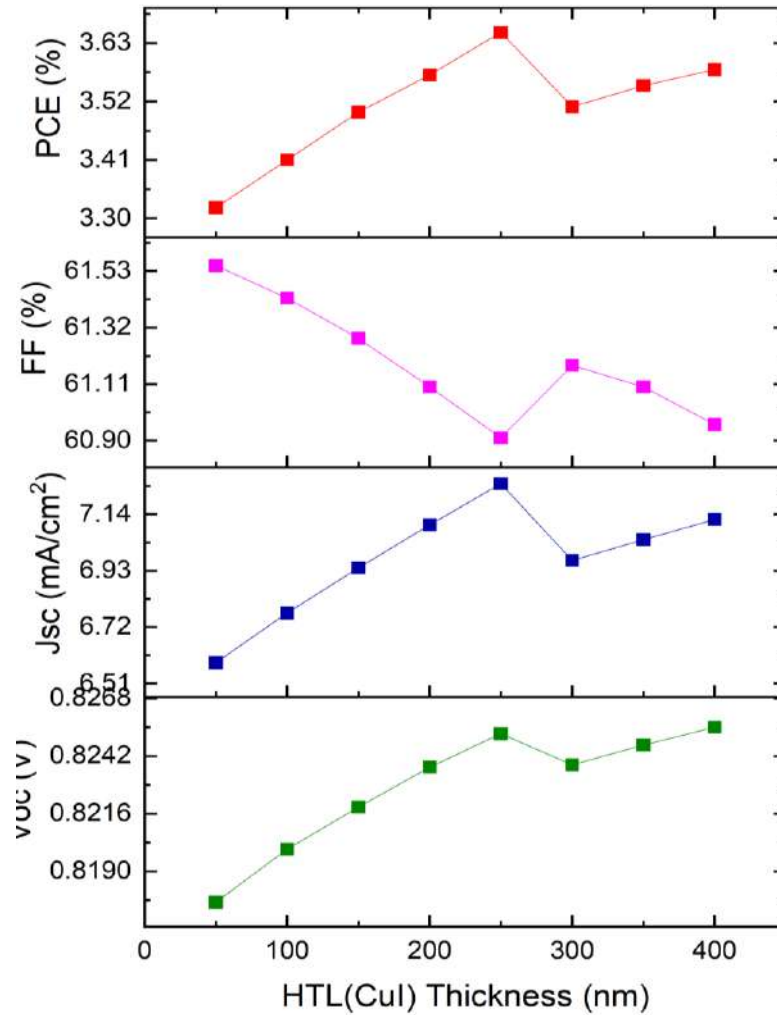


FIGURE 5. Effect of output parameters on HTL [CuI] layer thickness

On doping density of HTL layer

Figure 6 depicts the effect of hole transport layer doping density on output characteristics. As can be seen, raising the HTL doping concentration leads to increase of Voc, FF and PCE while Jsc is found to decrease

initially and then increases. This can be explained by the fact that conductivity has risen due to increase in concentration.

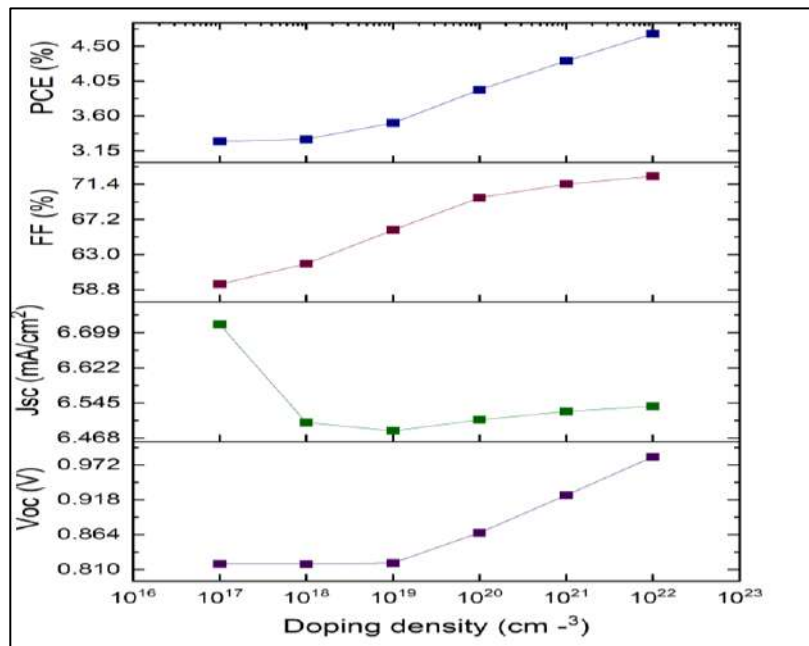


FIGURE 6. Effect of output parameters on HTL [CuI] doping density

On Hole Mobility of HTL

Figure 7 depicts the effect of hole mobility. The HTL layer's hole mobility is raised from 10^{-6} to $1 \text{ cm}^2/\text{Vs}$ and the outcomes are investigated. The output appears to validate the material's increased conductivity due to the mobility effect. The PCE is maximum at $10^{-3} \text{ cm}^2/\text{Vs}$

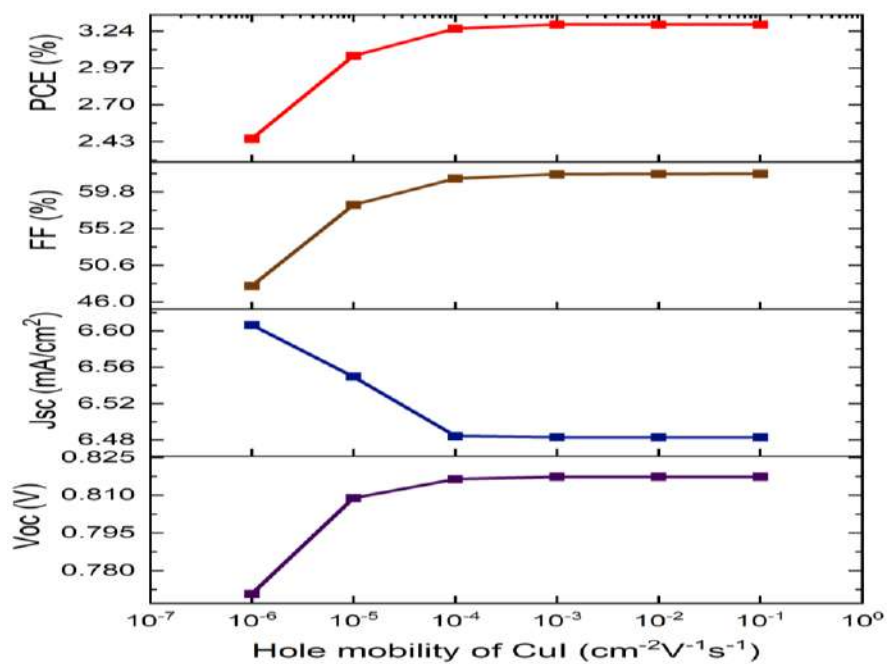


FIGURE 7. Effect of HTL [CuI] Hole Mobility on output parameters

On Electron Affinity of HTL

HTL's electron affinity is changed between 1.8 and 2.6 eV, and the influence on output performance is displayed in Fig.8. At 2.4 eV, PCE of 3.56 % and Voc of 0.9234V are attained for the best value. At 2.8V, an FF of 61.82 % is obtained. The BHJ PSC's active and transport layers benefit from better energy alignment and improved charge transport when the bandgap effect is reduced by the best electron affinity

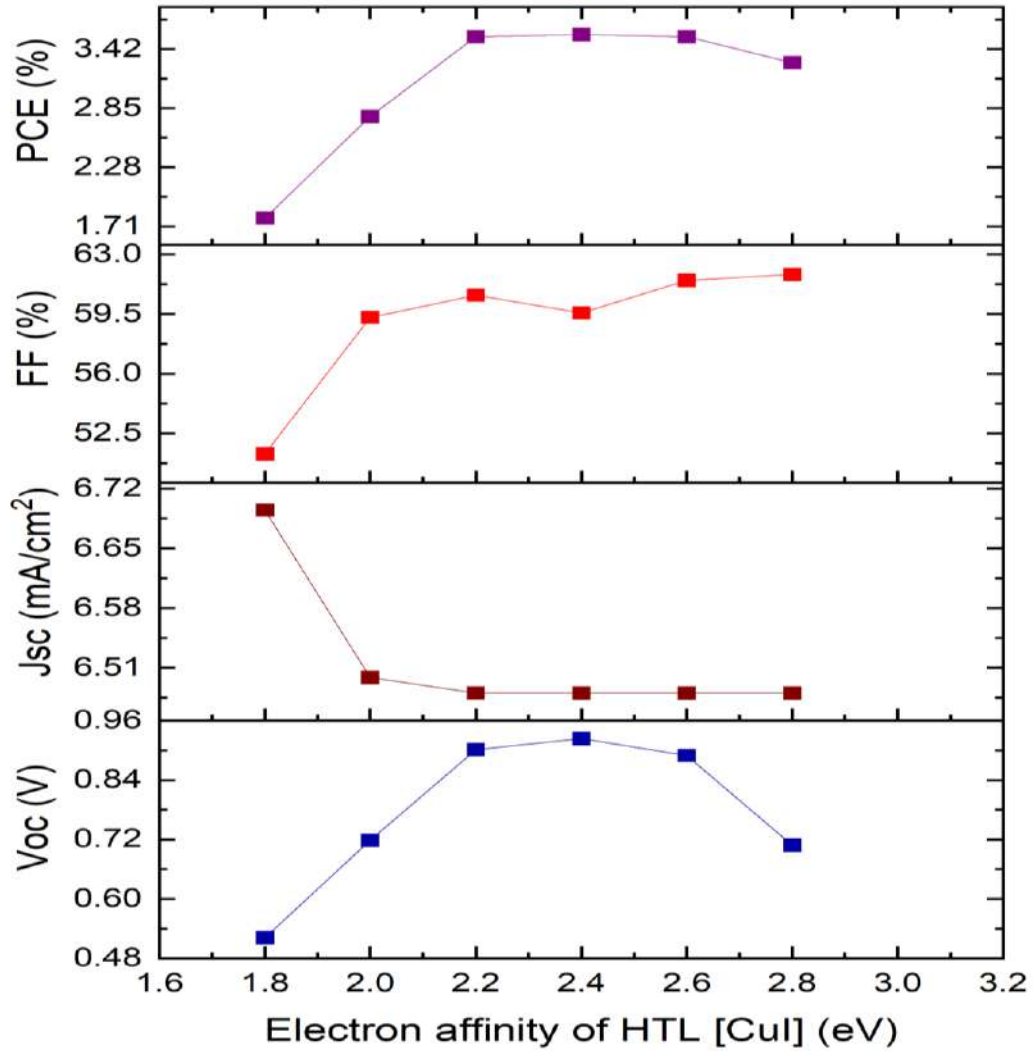


FIGURE 8. Effect of output parameters and HTL [CuI] Electron Affinity

Effect of defect density at interface layers [HTL/Active]

One interface has been considered: HTL/Active. The defect densities of the HTL/Active layer contact are varied from 10^7 cm^{-2} to 10^{12} cm^{-2} in the simulations. Increased defect density results in more traps and lowers cell production. For better output, low interface defect densities are desirable. The effects of changing defect density at the HTL/Active layer contact are depicted in Fig 9. The optimized values are obtained at 10^7 cm^{-2}

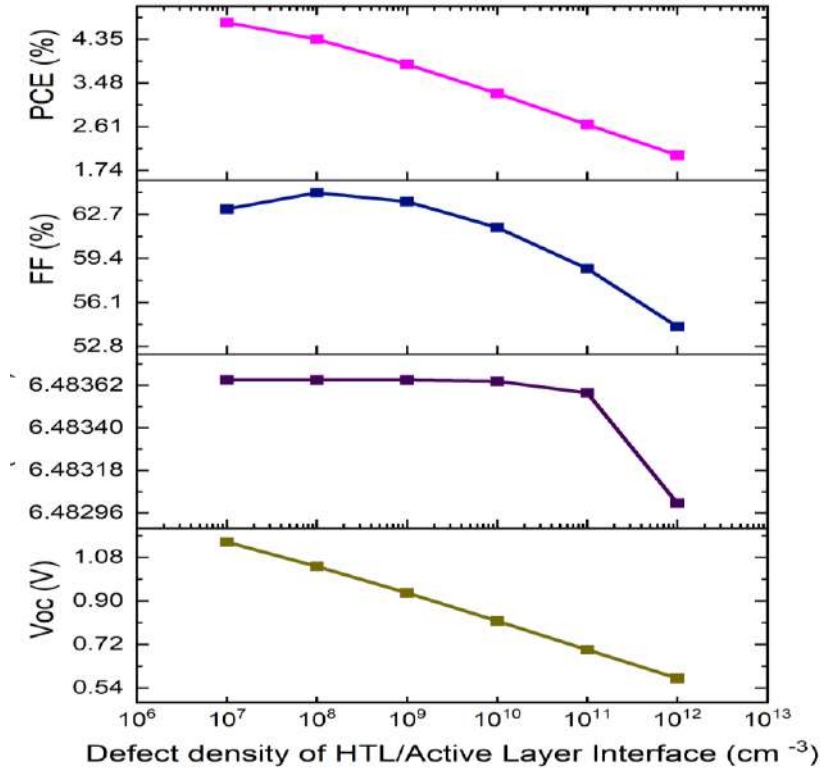


FIGURE 9. Effect of HTL/Active Layer Interface Defect Density on Output Parameters

CONCLUSION

In this work, the impact of key HTL parameters on the functionality of the BHJ PSC was examined. The results demonstrated that the cell performance can be improved by increasing thickness to 400 nm and reducing defect density. The defect density at the HTL/Active contact influences the output parameters. To boost efficiency in the simulation, the transport layer's characteristics are optimised. With the optimised numerical parameters, the cell performance is $V_{oc} = 1.2170$ V, $J_{sc} = 6.9243$ mA/cm², $FF = 73.95\%$, and $PCE = 6.23\%$. The parameters for the optimised device performance are provided in Table 3. The improved outcome encourages the usage of CuI as a substitute HTL on BHJ polymer solar cell. After optimization and simulation, we successfully constructed a BHJ PSC with inorganic CuI HTL with good performance.

TABLE 3. Optimized Device Performance Parameters

Device Parameters	Thickness Nm	χ eV	μ_p cm ² /Vs	Final
Voc [V]	0.8252	0.9234	0.8173	1.2170
Jsc [mA/cm ²]	7.2543	6.4802	6.4829	6.9243
FF [%]	60.91	59.57	62.01	73.95
PCE [%]	3.65	3.56	3.29	6.23

ACKNOWLEDGMENTS

Pr. M. Burgelman, University of Gent, Belgium, generously provided the SCAPS program to the authors.

REFERENCES

1. K. M. Coakley and M. D. McGehee, *Chem. Mater* **16**, 4533–4542 (2004).
2. B. A. Gregg and M. C. Hanna, *J. Appl. Phys.* **93**, 3605–3614 (2003).
3. M. A. Green, K. Emery, Y. Hishikawa, W. Warta and E. D. Dunlop, *Progress Photovolt.: Res. Appl.* **23**, 1–9 (2015).
4. M. C. Scharber and N. S. Sariciftci, *Prog. Polym. Sci.* **38** 1929–1940 (2013).
5. L. Lu, T. Zheng, Q. Wu, A. M. Schneider, D. Zhao and L. Yu, *Chem. Rev.* **115**, 12666–12731 (2015).
6. S. Venkatesan, E. Ngo, D. Khatriwada, C. Zhang and Q. Qiao, *ACS Appl. Mater. Interfaces*, **7**, 16093–16100 (2015).
7. G. Li, R. Zhu and Y. Yang, *Nat. Photonics*, **6**, 153–161 (2012).
8. F. Zhang, M. Johansson, M. R. Andersson, J. C. Hummelen and O. Inganäs, *Adv. Mater.* **14**, 662–665 (2002).
9. G. Li, V. Shrotriya, J. Huang, Y. Yao, T. Moriarty, K. Emery and Y. Yang, *Nature Materials* **4**, 80–84 (2010).
10. K. Wong, H. Yip, Y. Luo, K. Wong, W. Lau, K. Low, H. Chow, Z. Gao, W. Yeung and C. Chang, *Appl. Phys. Lett.* **80**, 2788–2790 (2002).
11. M. Kemerink, S. Timpanaro, M. M. de Kok, E. A. Meulenkaamp and F. J. Touwslager, *J. Phys. Chem. B* **108**, 18820–18825 (2004).
12. H. Yan, P. Lee, N. R. Armstrong, A. Graham, G. A. Evmenenko, P. Dutta and T. J. Marks, *J. Am. Chem. Soc.* **127**, 3172–3183 (2005).
13. Y. H. Kim, S. H. Lee, J. Noh and S. H. Han, *Thin Solid Films* **510**, 305–310 (2006).
14. W. Sun, H. Peng, Y. Li, W. Yan, Z. Liu, Z. Bian and C. Huang, *J. Phys. Chem. C* **118**, 16806–16812 (2014).
15. N. Loew, S. Komatsu, H. Akita, K. Funayama, T. Yuge, T. Fujiwara and M. Ihara, *ECS Trans.* **58**, 77–88 (2014).
16. A. Petrović, J. Gojanović, P. Matavulj, M. Islam, S. Živanović, “Temperature dependence of P3HT:ICBA polymer solar cells”, *Proceedings of the International Conference on Numerical Simulation of Optoelectronic Devices (NUSOD) (Copenhagen, Denmark, 2017)* pp. 133–134.
17. S. A. Gopalan, A. I. Gopalan, A. Vinu, K. P. Lee and S. W. Kang, *Sol. Energy Mater. Sol. Cells* **174**, 112–123 (2018).
18. B. Xu, S. A. Gopalan, A. I. Gopalan, Q. Qiao and S. W. Kang, *Polymers*, **10**, 121 (2018).
19. F. Parenti, F. Tassinari, E. Libertini, M. Lanzi and A. Mucci, *ACS Omega* **2**, 5775–5784 (2017).
20. S. A. Gopalan, A. I. Gopalan, K. P. Lee, S. Venkatesan, B. H. Kang, S. W. Lee, J. S. Lee, Q. Qiao, D. H. Kwon and S. W. Kang, *Sol. Energy Mater. Sol. Cells* **141**, 275–290 (2015).
21. S. A. Gopalan, A. I. Gopalan, A. Vinu, K. P. Lee and S. W. Kang, *Sol. Energy Mater. Sol. Cells*, **174**, 112–123 (2018).
22. G. Li, Y. Yao, H. Yang, V. Shrotriya, G. Yang and Y. Yang, *Adv. Funct. Mater.* **17**, 1636–1644 (2007).
23. Z. He, F. Liu, C. Wang, J. Chen, L. He, D. Nordlund, H. Wu, T. P. Russell and Y. Cao, *Mater. Horiz.* **2**, 592–597 (2015).
24. X. Zhu, K. Lu, B. Xia, J. Fang, Y. Zhao, T. Zhao, Z. Wei and L. Jiang, *Polymers* **8**, 4 (2016).
25. L. Lu, W. Chen, T. Xu and L. Yu, *Nat. Commun.* **6**, 7327 (2015).
26. M. M. Stylianakis, D. Konios, C. Petridis, G. Kakavelakis, E. Stratakis and E. Kymakis, *2D Mater.* **4**, 042005 (2017).
27. B.N. Ezealigo, A.C. Nwanya, A. Simo, R. U. Osuji, R. Bucher, M. Maaza and F. I. Ezema, *Arab. J. Chem.* **12**, 5380–5391 (2019).
28. J. Liu, Y. Zhang, C. Liu, M. Peng, A. Yu, J. Kou, W. Liu, J. Zhai and J. Liu, *Nanoscale Res. Lett.* **11**, 281 (2016).
29. O. Madelung, *III-V compounds, Semiconductors: Data Handbook* (Springer, 2004), pp. 71–172.
30. Z. El Jouad, M. Morsli, G. Louarn, L. Cattin, M. Addou and J.C. Bernède, *Sol. Energy Mater. Sol. Cells*

- 141**, 429–435 (2015).
31. M. I. Hossain, F. H. Alharbi and N. Tabet, [Sol. Energy](#) **120** 370–380 (2015).
 32. B. M. Omer, A. Khogali, A. Pivrikas, “Amps-1d modeling of p3ht/pcbm bulk-heterojunction solar cell”, 37th IEEE Photovoltaic Specialists Conference (2011) pp. 000734–000743
 33. K. S. Nithya and K. S. Sudheer, [J. Phys. Commun.](#) **4**, 025012 (2020)
 34. K. S. Nithya and K. S. Sudheer, [J Optik](#) 217, 164790 (2020)
 35. M. Burgelman, P. Nollet and S. Degrave, [Thin Solid Films](#) 361, 527–532 (2000).
 36. M. Burgelman, K. Decock, A. Niemegeers, J. Verschraegen, S. Degrave, Scaps Manual, 2016 (February).