Declaration

I hereby declare that the work presented in this thesis titled *Flexible Organic Transistors with Natural Materials for Eco-friendly Electronics* submitted to the Indian Institute of Technology Jodhpur in partial fulfilment of the requirements for the award of the degree of Doctor of Philosophy, is a bonafide record of the research work carried out under the supervision of Professor Shree Prakash Tiwari. The contents of this thesis in full or in parts, have not been submitted to, and will not be submitted by me to, any other Institute or University in India or abroad for the award of any degree or diploma.



Gargi Konwar
P19EE204

Certificate

This is to certify that the thesis titled *Flexible Organic Transistors with Natural Materials for Eco-friendly Electronics*, submitted by *Gargi Konwar (P19EE204)* to the Indian Institute of Technology Jodhpur for the award of the degree of *Doctor of Philosophy*, is a bonafide record of the research work done by him under my supervision. To the best of my knowledge, the contents of this report, in full or in parts, have not been submitted to any other Institute or University for the award of any degree or diploma.



Prof. Shree Prakash Tiwari
Ph.D. Thesis Supervisor

Abstract

Continuous development of technologies over time has significantly shortened the lifespan of devices, resulting in a substantial increase in electronic waste (E-waste) causing negative impact on environment and ecology. Flexible electronics has emerged as promising technology towards offering solution to this issue by capability of incorporation of nature originated materials in device fabrication through instilling eco-friendliness and potential biodegradability. Organic field-effect transistors (OFET) have been widely investigated as crucial device component for flexible electronics due to its applicability in circuit, sensing, and memory applications. This thesis is an effort to contribute towards flexible biodegradable electronics. Specific emphasis was given on the demonstration of eco-friendly OFETs for circuit and sensing applications with multi-functionality and eventual decomposability, which are essential requirements for smart and sustainable electronics.

To start with, various solution processed biocompatible or natural polymers including polyvinylpyrrolidone, silk fibroin, gelatin, chitosan, egg albumen, and cellulose derivative were explored as promising gate dielectrics in OFETs, either as single layer or in bilayer combination with other polymers or a thin high-*k* hafnium dioxide (HfO_2). All these devices were fabricated on indium tin oxide (ITO) coated glass or polyethylene terephthalate (PET) substrate, where ITO acting as bottom electrode. Widely explored pristine TIPS-Pentacene or TIPS-Pentacene: Polystyrene (PS) blend was utilized to form active layer with gold (Au) top electrodes working as source/drain, to demonstrate p-channel transistor characteristics for -5 V operation. Low voltage operation (-3 V) in flexible OFETs was demonstrated using a pristine layer of solution processed gelatin in combination with TIPS-Pentacene: PS blend. These devices exhibited excellent electrical characteristics with extracted field-effect mobility (μ) value approaching to as high as 3.0 $cm^2\ V^{-1}\ s^{-1}$ with near-zero threshold voltage and low subthreshold swing (*SS*), along with remarkable operational stability as confirmed by various stability tests including bias-stress, repeatability, cyclic, bending and long-term ambient stability. A very high environmental stability over 24 weeks was observed with almost unchanged electrical characteristics. Circuit applicability of these devices was successfully demonstrated through resistive load inverters.

Multifunctionality in OFETs with gelatin/HfO_2 hybrid bilayer gate dielectric and TIPS-Pentacene as semiconductor was demonstrated with multi-parameter sensing capabilities for visible/UV light and humidity, which also led to real time breath rate monitoring, a simple tool for wellness monitoring. Maximum field-effect mobility (μ_{max}) of 2 $cm^2\ V^{-1}\ s^{-1}$ was exhibited with low *SS* value of ~200 mV/dec., and high current on-off (I_{on}/I_{off}) ratio ~10^4 for -5 V operation with excellent electrical, operational, and bending stability. For the enhancement of performance and stability in devices with nature originated dielectrics, a composite of gelatin and chitosan was investigated with chitosan acting as a tuning agent to modulate the gate dielectric properties through controlled crosslinking facilitated by the electrostatic interaction and strong hydrogen bonding between the functional groups of polymers. Devices fabricated with a gelatin: chitosan ratio of 10:5 exhibited better electrical stability upon bending for 100 repeated bending cycles compared to other ratios. Circuit applicability, high bias-stress stability, and shelf life for 24 months indicated overall high performance using the composite gate dielectrics.

Finally, devices were fabricated on paper substrate thorough process optimization for substrate planarization with polyvinyl alcohol (PVA), silver (Ag) as gate electrode, to move towards eco-sustainable electronics. Cyanoethyl cellulose (CEC), a synthesized version of cellulose was comprehensively investigated as a potential gate dielectric in OFET devices for eco-friendliness. Transistors with CEC dielectric and paper substrate were demonstrated in an effort to produce biodegradable systems for eco-sustainable electronics. These devices exhibited high performance with excellent saturation in the output characteristics for -5 V operation along with remarkable electrical and operational stability. High bendability of devices was confirmed upon application of tensile stress through bending along the channel for the bending radii of 12, 7, and

5-mm. Moreover, high cyclic stability was achieved in devices even after keeping the device in high humid environment with relative humidity reaching to ~100 %. High decomposability in water rich soil in ~20 days was observed due to the microorganisms present in soil environment, confirming excellent biodegradability which is highly essential for eco-sustainable electronics. Due to high performance and biodegradability, these paper-based devices can offer huge potential towards the future flexible and green electronics, and can help in minimizing the impact of E-waste on environment and ecology.

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Acknowledgements

I wish to convey my sincere appreciation to all those individuals who have directly or indirectly contributed throughout this journey. Their presence and support have proven to be of immense significance throughout this endeavour.

First and foremost, I would like to express my sincere appreciation to my supervisor, *Prof. Shree Prakash Tiwari*, for his continuous motivation and encouragement, invaluable guidance, and expertise. His consistent support and valuable feedback have significantly influenced my research and development as a scholar. I greatly appreciate the significant time he has dedicated to encouraging me from starting to end of this Ph.D. journey. I express my gratitude to him for being my PhD supervisor and giving me the opportunity to work in FLAME Research Group.

I am also thankful to student research committee (SRC) members, *Prof. Mahesh Kumar, Dr. Satyajit Sahu*, and *Dr. Arpit Khandelwal*, for their constructive criticism and guidance, which have helped me to refine my work and broaden my perspective.

I express my sincere gratitude to my seniors, *Dr. Vivek Raghuwanshi, and Dr. Ajay Kumar Mahato* and my co-authors, *Pulkit Saxena* and *Dr. Sachin Rahi,* for their valuable suggestions and timely guidance during my research.

I would like to acknowledge Visvesvaraya Young Faculty Research Fellowship (YFRF) Scheme, (Ministry of Electronics & IT), Science and Engineering Research Board, Government of India for their financial support in this work, and Indian Nano electronics Users' Programme (INUP-i2i) at Department of Physics, IIT Kharagpur for experimental facilities. In addition, I would like to acknowledge CSIR Foreign Travel Grant (March 2023), and SERB International Travel Support Scheme (July 2024) for the financial support to attend international conferences.

I would like to express my appreciation to my lab mates and juniors *Shalu, Anurag, Anil, Somnath, Nishi, Saqib, Monika, Ritesh, and Saurav*, for their support, and for maintaining a healthy and enthusiastic research environment in the laboratory.

Most importantly, I want to thank my parents *Mr. Durgeswar Konwar* and *Mrs. Ranju Konwar*, and my lovely sisters *Kanika*, *Barlina*, and *Darshna Drisha*, for always believing in me and motivating me to become a better version of myself. Their belief in me, especially during challenging times, has been my source of strength. Their constant push always help me to do beyond my limitations. Lastly, I will always be grateful to the almighty for giving me the strength to complete my research journey on a good note.

Gargi Konwar
PhD Student

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List of Symbols

Symbol	*Description*
ΔV_{TH}	Change in Threshold Voltage
ΔI_{DS}	Change in Drain Current
μ	Field Effect Mobility
μ_{avg}	Average Mobility
μ_{max}	Maximum Mobility
μ_{sat}	Mobility in Saturation regime
A	Effective area
C_i	Capacitance Density
D_{it}	Interface Trap Density
E	Electric Field
E_V	Valence Band
I_{DS}	Drain current
$I_{DS, Lin}$	Drain current in Linear regime
$I_{DS, Light}$	Drain current under Light Irradiation
$I_{DS, Dark}$	Drain current in Dark Condition
$I_{DS, Sat}$	Drain current in Saturation regime
I_{off}	Minimum Current in Off-state
I_{on}	Maximum Current in On-state
k	Dielectric Constant
k_p	Process transconductance
$k_{p, max}$	Maximum process transconductance
P_i	Intensity of Incident Light
L	Length of the Channel
R	Photoresponsivity
R_{bend} or r_{bend}	Bending Radius
R_L	Load Resistance
P	Current modulation
q	Electronic charge
T	Absolute Temperature
t	Transfer Integral
V_{DD}	Drain to Drain Voltage
V_{DS}	Drain to Source Voltage
V_{GS}	Gate to Source Voltage
V_{In}	Input Voltage
V_{out}	Output Voltage
V_{TH}	Threshold Voltage
V_{TO}	Initial Threshold Voltage
W	Width of Transistor Channel
β	Stretched exponential factor
λ	Wavelength of Light Source Used
τ	Relaxation Time

List of Abbreviations

Abbreviation	*Full form*
AFM	Atomic Force Microscopy
Ag	Silver
Al_2O_3	Aluminum oxide
ALD	Atomic Layer Deposition
Au	Gold
BDS	Broadband Dielectric Spectrometer
BGBC	Bottom Gate Bottom Contact
BGTC	Bottom Gate Top Contact
BTBT	2,7-Bis(thien-2-yl)benzo[1,2-b:4,5-b']dithiophene)
CEC	Cyanoethyl Cellulose
CS	Chitosan
CuPc	Copper Phthalocyanine
C8-BTBT	2,7-Dioctyl[1]benzothieno[3,2-b][1]benzothiophene
diF-TES-ADT	2,8-Difluoro-5,11-bis(triethylsilylethynyl)anthradithiophene
DI	De-ionized
DMF	Dimethylformamide
DNTT	Dinaphtho[2,3-*b*:2',3'-*f*]thieno[3,2-*b*]thiophene
DPPTTT	Poly[2,5-(2-octyl-dodecyl)-3,6-diketopyrrolopyrrole-alt-5,5-(2,5-di(thien-2-yl) thieno[3,2-b] thiophene)]
DSC	Differential Scanning Calorimetry
EDL	Electrical double layer
EGOFET	Electrolyte-gated organic field-effect transistor
FET	Field-Effect Transistor
FTIR	Fourier-transform infrared spectroscopy
FWHM	Full Width At Half Maximum
F8T2	Poly(9,9-dioctylfluorene-alt-bithiophene)
HfO_2	Hafnium Dioxide
HOMO	Highest Occupied Molecular Orbital
IPA	Isopropyl Alcohol
ITO	Indium Tin Oxide
KITE	Keithley Interactive Test Environment
LUMO	Lowest Unoccupied Molecular Orbital
MEH-PPV	Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene]
MIM	Metal Insulator Metal
MISM	Metal Insulator Semiconductor Metal
MMT	Million Metric Tons
MOSFET	Metal-oxide-semiconductor field-effect transistor
OFET	Organic Field-Effect Transistor
OLED	Organic Light Emitting Diode
OPV	Organic Photo Voltaic
OSC	Organic Semiconductor
OTFT	Organic Thin Film Transistor
P3HT	Poly(3-hexylthiophene-2,5-diyl)
PANI	Polyaniline
PBS	Polybutylene succinate
PBTTT-14	Poly(2,5-bis(3-alkylthiophen-2-yl)thieno[3,2-b]thiophene)
PCL	Polycaprolactone

PDPP-TTT	Poly[2,5-(2-octyldodecyl)-3,6-diketopyrrolopyrrole-alt-5,5-(2,5-di(thien-2-yl)thieno [3,2-b]thiophene)]
PDMS	Polydimethylsiloxane
PEDOT: PSS	Poly(3,4-ethylenedioxythiophene) with poly(styrenesulfonate)
PEN	Polyethylene napthalate
PES	Polyestersulfone
PET	Polyethylene terephthalate
PGA	Polyglycolic acid
PLA	Polylactic acid
PLGA	Polylactic-co-glycolic acid
PMMA	Poly(methylmethacrylate)
Ppy	Polypyrrole
PS	Polystyrene
PSPD	Position-sensitive photodiode
PTAA	Poly(triaryl amine)
PTFE	Polytetrafluoroethylene
PVA	Polyvinyl alcohol
C-PVA	Cross-linked poly(vinyl alcohol)
PVP	Poly(4-vinylphenol)
PVPy	Polyvinylpyrrolidone
RFID	Radio Frequency Identification
RMS	Root mean square
rpm	Revolutions per minute
R2R	Roll-to-roll
SAM	Self-Assembled Monolayer
SCS	Semiconductor Characterization System
SERB	Science and Engineering Research Board
SF	Silk fibroin
Si_3N_4	Silicon nitride
SMU	Source Measuring Unit
SPM	Scanning probe microscopy
SS	Sub-threshold Swing
TES-ADT	5,11-Bis(triethylsilylethynyl)anthradithiophene
TDMAH	Tetrakis(dimethylamino)hafnium
TFT	Thin Film Transistors
TGA	Thermogravimetric analysis
TGBC	Top Gate Bottom Contact
TGTC	Top Gate Top Contact
TIPS	6,13-Bis(triisopropylsilylethynyl) pentacene
UV	Ultraviolet
VTC	Voltage-transfer characteristic
XRD	X-ray Diffraction
YFRF	Young Faculty Research Fellowship