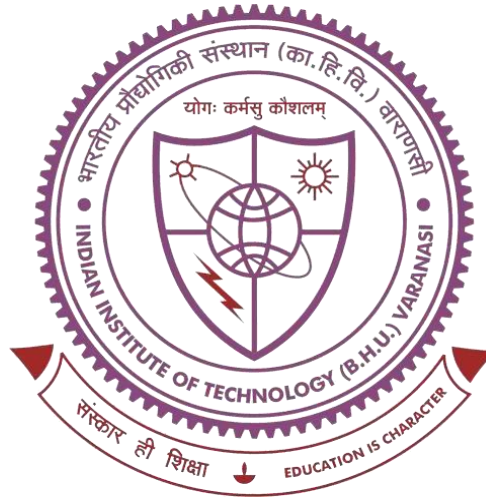


Studies on functionalization, characterization and application of cellulose and its composites



**Thesis submitted in partial fulfilment
For the Award of Degree**

Doctor of Philosophy
in
Biochemical Engineering

by

Rahul Ranjan

**School of Biochemical Engineering
Indian Institute of Technology (BHU) Varanasi
Uttar Pradesh- 221005**

Roll No: 19011001

2024

Dedicated
to
My Beloved Parents

**SCHOOL OF BIOCHEMICAL ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY
(BANARAS HINDU UNIVERSITY)
VARANASI - 221005**



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Dr. Prodyut Dhar
(Supervisor)

**School of Biochemical Engineering
Indian Institute of Technology
(Banaras Hindu University) Varanasi
Dr. Prodyut Dhar, Ph.D.**

**Assistant Professor
School of Biochemical Engineering
Indian Institute of Technology (BHU)
Varanasi-221005, U.P., INDIA**

DECLARATION BY THE CANDIDATE

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(Mr. Rahul Ranjan)

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Dr. Prodyut Dhar, Ph.D.
Assistant Professor
Biochemical Engineering
Institute of Technology (BHU)
Varanasi-221005, U.P., INDIA
School of Biochemical Engineering
Indian Institute of Technology
(Banaras Hindu University) Varanasi


Prof. Vikash Kumar Dubey
(Coordinator)
School of Biochemical Engineering
Indian Institute of Technology
(Banaras Hindu University) Varanasi

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LIST OF ABBREVIATIONS

LCA	Life cycle assessment
NaOH	Sodium hydroxide
MFC	Microfibrillated cellulose
NC	Nanocellulose
TAPPI	Technical Association of the Pulp and Paper Industry
MCC	Microcrystalline cellulose
NFCs	Nanofibrillated cellulose
CNC	Cellulose nanocrystals
BC	Bacterial cellulose
CNFs	Cellulose nanofibers
CMFs	Cellulose microfibrils
KOH	Potassium hydroxide
MoS ₂	Molybdenum disulphide
2D	Two-dimensional
1T	1-Trigonal
2H	2-Hexagonal
3R	3-Rhombohedral
NaClO ₂	Sodium chlorite
H ₂ O ₂	Hydrogen peroxide
NaOCl	Sodium hypochlorite
Na ₂ SO ₃	Sodium sulphite
<i>E. coli</i>	<i>Escherichia coli</i>
<i>S. aureus</i>	<i>Staphylococcus aureus</i>
H ₂ SO ₄	Sulphuric acid
H ₃ PO ₄	Phosphoric acid
H ₃ PO ₃	Phosphorous acid
TEMPO	2,2,6,6-tetramethyl-1-piperidinyloxy
HCl	Hydrochloric acid
HPH	High pressure homogenizer
DS	Degree of substitution
DMF	Dimethylformamide

PA	Phosphoric acid
DAHP	di-ammonium hydrogen phosphate
ADP	Ammonium di-hydrogen phosphate
Des	Deep eutectic solvents
ILs	Ionic liquids
DOX	Doxorubicin
CNF-P	Phosphorylated cellulose nanofiber
UPF	UV protection factor
GSM	Grams per square meter
FESEM	Field emission scanning electron microscopy
Q_e	Highest adsorption capacity
C_e	MB/MG concentration
C_0	Initial MB/MG concentration
V	Volume of the MB/MG solution
W	Gram of adsorbent
SB	Sugarcane bagasse
RH	Rice husk
MB	Methylene blue
MG	Malachite green
MOF	Metal-organic frame work
CO	Carbon monoxide
NO _x	Nitrogen oxides
ROS	Reactive oxygen species
CO ₂	Carbon dioxide
DRH	Delignified rice husk
DSDSB	Delignified sugarcane bagasse
DDDSB-MoS ₂	Delignified sugarcane bagasse-molybdenum di-sulphide
DRH-MoS ₂	Delignified rice husk-molybdenum di-sulphide
FTIR	Fourier transform infrared spectroscopy
XPS	X-ray photoelectron spectroscopy
EDX	Energy-dispersive X-ray spectroscopy
XRD	X-ray diffraction
SEM	Scanning electron microscope

Mo	Molybdenum
S	Sulphur
O	Oxygen
C	Carbon
PFO	Pseudo first order
PSO	Pseudo second order
CB	Conduction band
VB	Valence band
UV-DRS	UV-Vis diffuse reflectance spectroscopy
<i>C.I</i>	Crystallinity index
PC	Phosphorylated cellulose
VC	Viscose cloths
NMR	Nuclear magnetic resonance spectroscopy
AMT	Ammonium molybdate trihydrate
PC@5.5 %	PC gel with solid content of 5.5 %
PC@4.9 %	PC gel with solid content of 4.9 %
PC@4.3 %	PC gel with solid content of 4.3 %
<i>K</i>	Consistency index
<i>N</i>	Power law index
LVR	Linear viscoelastic region
<i>G'</i>	Storage modulus
<i>G''</i>	Loss modulus
FWHM	Full width at half maximum
<i>K_i</i>	Diffusion rate constant
<i>C_i</i>	Boundary layer thickness
PCMo@0.1%	Phosphorylated cellulose printed structures with 0.1g of AMT
PCMo@0.25%,	Phosphorylated cellulose printed structures with 0.25g of AMT
PCMo@0.5%,	Phosphorylated cellulose printed structures with 0.5g of AMT
PCMo@0.75%,	Phosphorylated cellulose printed structures with 0.75g of AMT
PCMo@1%,	Phosphorylated cellulose printed structures with 1g of AMT
<i>T_{onset}</i>	Onset thermal degradation temperature
<i>T_{max}</i>	50 % weight loss
ΔH	Enthalpy

ΔS	Entropy
ΔG	Gibb's free energy
DRS	Delignified rice straw
RSF-45	Rice straw film produced from gel prepared after 45 min of curing
RSF-60	Rice straw film produced from gel prepared after 60 min of curing
RSF-75	Rice straw film produced from gel prepared after 75 min of curing
RSF-90	Rice straw film produced from gel prepared after 90 min of curing
RS	Rice straw
TGA	Thermogravimetric analysis
APTES	(3-Aminopropyl) triethoxy silane
RSA-45	Rice straw aerogel produced from gel prepared after 45 min of curing
RSA-60	Rice straw aerogel produced from gel prepared after 60 min of curing
RSA-75	Rice straw aerogel produced from gel prepared after 75 min of curing
RSA-90	Rice straw aerogel produced from gel prepared after 90 min of curing
PALCAM	Polymyxin Acriflavin Lithium-chloride Ceftazidine Esculin Mannitol
PDA	Potato Dextrose agar
MRS	De Man, Rogosa and sharpe
XLD	Xylose Lysine Deoxycholate
CFU/g	Colony forming unit per gram
PET	Polyethylene terephthalate
TPA	Terephthalic acid
PU foam	Polyurethane foam
VRBA	Violet red bile agar
SCBF-30	Sugarcane bagasse film produced from gel prepared after 30 min of curing
SCBF-45	Sugarcane bagasse film produced from gel prepared after 45 min of curing

SCBF-60	Sugarcane bagasse film produced from gel prepared after 60 min of curing
SCBF-75	Sugarcane bagasse film produced from gel prepared after 75 min of curing
SCBF-90	Sugarcane bagasse film produced from gel prepared after 90 min of curing
PSCB-30	Phosphorylated sugarcane bagasse aerogel produced from gel prepared after 30 min of curing
PSCB-45	Phosphorylated sugarcane bagasse aerogel produced from gel prepared after 45 min of curing
PSCB-60	Phosphorylated sugarcane bagasse aerogel produced from gel prepared after 60 min of curing
PSCB-75	Phosphorylated sugarcane bagasse aerogel produced from gel prepared after 75 min of curing
DPPH	2,2-Diphenyl-1-picrylhydrazyl
PSCB-G	Phosphorylated sugarcane bagasse foam modified with Gallic acid
CC	Charge content
FD	Fossil depletion
POF	Photochemical ozone formation
TA	Terrestrial acidification

PREFACE

Given the global challenges presented by petroleum-based polymers, as well as the need to combat climate change and dwindling fossil fuel resources, there has been a noticeable push on sustainable technologies and research. These challenges have highlighted a crucial effort to shift away from the conventional dependence on polymers derived from petroleum to sustainable and affordable alternatives that are both eco-friendly and renewable. Development of renewable polymers with scalable processes for commercial utilization is another focus point towards developments in sustainable polymers. In line with the research progress in renewable polymers, there exists sustainable development goals (SDGs) such as clean water and sanitation (SDG6), industry, innovation and infrastructure (SDG 9), life below water (SDG 14), life on land (SDG 15), climate action (SDG 13), responsible consumption and production (SDG 12), sustainable cities and communities (SDG 11), and affordable and green energy (SDG 8) by United Nations. There has been a strong push to find alternative methods for producing polymers to address environmental concerns. As a result, there has been an intensive attempt to investigate biopolymers as potential solutions. In this field, agricultural biomass and waste offer exciting opportunities to create valuable products while reducing environmental harm. Cellulose is an extremely promising option for biopolymer production due to its remarkable properties such as mechanical strength, crystallinity, hydrophilicity, biodegradable, sustainable, environmentally friendly, non-toxic, affordable, and widely accessible. This thesis focuses on utilising different agro-wastes to extract cellulose, their *in-situ* modification, and phosphorylation *via* green routes for development of value-added products. The present thesis consists of 8 chapters in which chapter 4 to chapter 7 discuss the methodologies performed. The cellulose extraction and phosphorylation have been performed using an industrially viable, scalable, and green approach. The phosphorylated cellulose processed with the sustainable routes were 3D-printing, moulding, solution casting, freeze drying, and spinning to fabricate complex structure, cups and plates, films, aerogels, and threads. This thesis explores various sustainable solutions derived from abundantly available agricultural biomass to value-added products, specifically emphasizing sugarcane bagasse (SB), rice husk (RH), viscose

cloth, and rice straw for addressing urgent environmental challenges and provide practical advancements in different sectors. In chapter 4, the process of delignification of SB and RH is revealed, resulting in the extraction of cellulose that can be used as an effective adsorbent for methylene blue (MB) and malachite green (MG) dyes, respectively. In addition, using molybdenum salt and thiourea treatments, the resulting cellulose is converted into highly effective adsorbents and photocatalysts for removing dyes. These adsorbents also demonstrate antibacterial properties against *Escherichia coli* and *Staphylococcus aureus* for scalable and continuous wastewater dye and bacterial decontamination with potential practical applicability. The chapter introduces a unique strategy for the up-conversion of agricultural biomass into value-added bio-adsorbents, which can effectively and economically address the remediation of dyes with simultaneous microbial decontamination from polluted wastewater streams. Chapter 5 demonstrates an important development in sustainable material innovation using viscose cloth for gel production through phosphorylation. The phosphorylated gels exhibit improved properties, including higher water absorption, swelling capacity, and rheological attributes. Additionally, they show significant enhancements in their ability to remediate MB. The addition of MoS₂ salts to PC also showed excellent shear-thinning properties. It facilitated the *in-situ* growth of MoS₂ to introduce photocatalytic and antibacterial properties, enhancing the rate of MB degradation and pathogens removal from wastewater. This chapter provides a green, facile and energy-efficient strategy for fabricating PCs with easy processability through additive manufacturing techniques for producing value-added products, opening up new avenues for high-performance applications including continuous photodegradation and antibacterial properties. Chapter 6 of the thesis explores the process of transforming rice straw waste into useful materials like food packaging films, aerogels, and thermal boxes. The films and aerogel boxes demonstrate remarkable mechanical strength, thermal stability, and flame-retardant properties using phosphorylation, solution casting and freeze-drying techniques. Furthermore, the development of biodegradable cups and aerogel boxes showcases the versatility and environmental friendliness of materials made from rice straw. The rice straws have been processed to form films (heat sealing), cups (anti-fizzing and solvothermal stability) and aerogels properties (thermal insulation, low thermal conductivity) and can act as potential replacement of non-biodegradable and

highly flammable polyethylene and polyurethane, and polystyrene. In Chapter 7, we explore the potential of sugarcane bagasse and demonstrate the production of phosphorylated films, aerogels, and fruit foams that offer a wide range of functionalities. These materials possess transparency, mechanical strength, flame retardancy, thermal stability, and biodegradability, making them highly suitable for a wide range of packaging applications. The foams prepared with sugarcane bagasse biomass can become a suitable mechanically stable alternative for damage protection in perishable fruit products, as a potential replacement for non-biodegradable polystyrene foams used currently. It is worth mentioning that the use of these materials for packaging paneer and apples highlights their ability to prolong shelf life and provide environmentally friendly alternatives to traditional packaging materials. Together, this thesis highlights the importance of utilising agricultural biomass and waste to create sustainable materials that can be used in a wide range of applications. This not only helps address pressing environmental issues and UN SDGs, but also promotes innovation and progress in different industries. This thesis aims to make a valuable contribution towards a scalable, facile, low-cost, utilization of abundantly available waste biomass for making high value products with lower carbon footprints which can be utilized in the direction of the global efforts in achieving a sustainable and resilient future.

