





TEZPUR UNIVERSITY
(A Central University Established by an Act of Parliament)
Napaam, Tezpur-784028, Sonitpur, Assam, India

DECLARATION

I do hereby declare that the thesis entitled “**Exploration of Polymeric Hydrogels: Potential Applications as Adhesives, Photocatalysts, and Electroactive Materials**”, is the result of investigations carried out by me in the Department of Chemical Sciences, Tezpur University, India. I also declare that I have adhered to all principles of academic honesty and integrity and have not misrepresented or falsified any idea/data/source in my submission. I have also acknowledged all sources, wherever the work described is based on the findings of other investigators. Neither this work as a whole nor any part of it has been submitted to any other University or Institute for academic credit.

Date: 20-05-2025

Place: Tezpur University, Tezpur

(Asfi Ahmed)

Department of Chemical Sciences
Tezpur university



TEZPUR UNIVERSITY

(A Central University Established by an Act of Parliament)

Napaam, Tezpur-784028, Sonitpur, Assam, India

Dr. Swapan Kumar Dolui
Professor
Department of Chemical Sciences

Tell (O): +91 (3712) 275052
Fax (O): +91 (3712) 267006
Email: dolui@tezu.ernet.in

CERTIFICATE FROM THE SUPERVISOR

This is to certify that the thesis entitled “**Exploration of Polymeric Hydrogels: Potential Applications as Adhesives, Photocatalysts, and Electroactive Materials**” submitted to Tezpur University, in the Department of Chemical Sciences, under the School of Sciences, in partial fulfillment for the award of the degree of Doctor of Philosophy in Science is a record of research work carried out by **Ms. Asfi Ahmed** under my supervision and guidance.

She has fulfilled all the requirements for submitting the thesis for the award of the degree of Doctor of Philosophy in Science. All help and assistance she received from various sources have been duly acknowledged. No part of this thesis has been reproduced elsewhere for any award or other degree.

Date: 20-05-2025

Place: Tezpur University, Tezpur

(Prof. Swapan Kumar Dolui)

Professor

Department of Chemical Sciences

School of Sciences

Tezpur University

Assam, India-784028



TEZPUR UNIVERSITY

(A Central University Established by an Act of Parliament)

Napaam, Tezpur-784028, Sonitpur, Assam, India

Dr. Utpal Bora
Professor
Department of Chemical Sciences

Tell (O): +91 (3712) 275052
Fax (O): +91 (3712) 267006
Email: ubora@tezu.ernet.in

CERTIFICATE FROM THE CO-SUPERVISOR

This is to certify that the thesis entitled “**Exploration of Polymeric Hydrogels: Potential Applications as Adhesives, Photocatalysts, and Electroactive Materials**” submitted to Tezpur University, in the Department of Chemical Sciences, under the School of Sciences, in partial fulfillment for the award of the degree of Doctor of Philosophy in Science is a record of research work carried out by **Ms. Asfi Ahmed** under my supervision and guidance.

She has fulfilled all the requirements for submitting the thesis for the award of the degree of Doctor of Philosophy in Science. All help and assistance she received from various sources have been duly acknowledged. No part of this thesis has been reproduced elsewhere for any award or other degree.

Date: 29-04-2025

Place: Tezpur

(Prof. Utpal Bora)

Professor

Department of Chemical Sciences

School of Sciences

Tezpur University

Assam, India-784028



TEZPUR UNIVERSITY
(A Central University Established by an Act of Parliament)
Napaam, Tezpur-784028, Sonitpur, Assam, India

CERTIFICATE FROM THE EXTERNAL EXAMINER AND ODEC

This is to certify that the thesis entitled “**Exploration of Polymeric Hydrogels: Potential Applications as Adhesives, Photocatalysts, and Electroactive Materials**” submitted to Tezpur University, in the Department of Chemical Sciences, under the School of Sciences, in partial fulfillment for the award of the degree of Doctor of Philosophy in Science has been examined by us on and found to be satisfactory.

The committee recommends awarding Ms. Asfi Ahmed the degree of Doctor of Philosophy.

Supervisor

Co-Supervisor

External Examiner

Date:

Date:

Date:

PREFACE

The escalating demand for polymeric materials to fulfill human needs has led to the synthesis of polymeric hydrogels. Their unique ability to absorb and retain water or other biological fluids up to a thousand times their weight and their tunable functionality make them highly valuable for a wide range of applications including both biomedical and industrial fields. In addition, many hydrogels are biocompatible, biodegradable, and soft in nature, thus possessing similarities to natural tissue. Furthermore, they are responsive to different environmental stimuli. Due to these remarkable properties, hydrogels are well suited for numerous medical practices and other applications such as tissue adhesion, wound healing, controlled release of drugs, actuators, etc. Moreover, their tremendous water-absorbing property and the presence of different functional groups allow them to absorb and chemically bind organic pollutants and hence are applicable in water remediation processes.

The present thesis focuses on developing and evaluating different hydrogel materials and highlights their potential applications as adhesives, photocatalysts, and electric field-responsive materials. The contents of this thesis are compiled into six chapters which include a general introduction of hydrogel discussing their potential applications in the said fields, followed by comprehensive experimental studies of hydrogel in those areas. Finally, the major findings of this thesis work and its future scope have been discussed in the last chapter.

This study is expected to contribute valuable insights to the rapidly evolving field of hydrogels and open new avenues for further research in this field.

Place: Tezpur University, Napaam, Tezpur

Asfi Ahmed

Date:

ACKNOWLEDGEMENT

It is an immense pleasure to convey my sincere gratitude to every individual I encountered during my PhD tenure. It is indeed a long journey, and along with dedicated work, it requires patience and resilience. However, this work would not have been possible to undertake and accomplish without the encouragement and support of numerous people.

At the very beginning, I acknowledge the blessings of the Almighty in the successful completion of this research work. His divine guidance has illuminated my path and shielded me from unforeseen difficulties.

I would like to express my heartfelt gratitude to my supervisor, Prof. Swapan K. Dolui for his insightful guidance and unwavering support throughout this challenging PhD journey. His profound knowledge, patience, and belief in my abilities have been invaluable in shaping the direction of my research. I shall be forever indebted to him for all his lessons which not only enhanced my research work but also provided me with valuable life lessons that will continue to guide me in my future endeavors.

I extend my sincere gratitude to my co-supervisor, Prof. Utpal Bora for his constant support and valuable suggestions during my PhD journey. His mentorship and guidance were crucial in overcoming challenges in submitting my thesis and ensuring the successful completion of my PhD.

I am immensely grateful to my Doctoral Committee member, Prof. Ashim Jyoti Thakur, for his advice and suggestions. I am also thankful to Prof. Panchanan Puzari, Head of the Department, and Prof. Ruli Bora, former Head of the Department, for giving me the opportunity to conduct my research work and for granting me access to the necessary technical facilities in this Department.

My sincere gratitude goes to the Vice-Chancellor, Tezpur University for the positive campus environment and excellent infrastructure for the academic community.

I would like to acknowledge all the faculty members of this Department who have inspired me to improve my academic growth throughout this journey. I am also grateful to the Technical Staff of the Department of Chemical Sciences, SAIC, Tezpur University,

SAIF, CSIR-NEIST, Jorhat, and CRF, IIT Ropar for their assistance and cooperation in providing analytical support for my research.

I would like to express my appreciation to the office staff of the Department of Chemical Sciences, Tezpur University, for their assistance in various administrative matters throughout my PhD journey. I am especially thankful to the Non-Technical Staff for their timely assistance.

I would also like to acknowledge Prof. Manabendra Mandal and Dr. Muzamil Ahmad Rather from the Department of Molecular Biology and Biotechnology, Tezpur University, Prof. Bodhisatwa Das and Anwesha Mukherjee from the Department of Biomedical Engineering, Indian Institute of Technology, Ropar, for their assistance in the biocompatibility experiments.

I am deeply grateful to Tezpur University for the financial support provided through the Research Innovation Grant and the Institutional Fellowship.

I would like to express my profound appreciation to all my past and present lab members Dr. Kiranjyoti Mohan, Dr. Jayashree Nath, Dr. Junali Handique, Dr. Anindita Bora, Dr. Simanta Doley, Dr. Priyankamoni Saikia, Dr. Shahnaz Ahmed, Dr. Suman Lahkar, and Dr. Kankana Barua for their love and support throughout the journey. I also acknowledge Arzu Almin, Dibyashree Dolakasharia, Manash Jyoti Kalita, Prastuti Saikia, and Jayanta Bordoloi for their help during my PhD. I am also grateful to the M.Sc. project student in our laboratory Priyanka Ray who worked with me on her M.Sc. project.

My sincere gratitude goes to Deepsikha Chaliha, Dr. Dipika Konwar, Ankita Rakshit, Dr. Sudhamoyee Katakya, Debajani Duarah, Bhaktadeep Chaliha, Nayab Hussain, and Dr. Raktim Abha Saikia for their unwavering support and warm wishes during the entire journey.

I am grateful to Mrs. Sutapa Dolui and Dr. Anindita Dewan for their motivation and care throughout the journey.

I am truly thankful to the warden, caretakers, and all the members of Pobitora Women's Hostel for making my stay on campus feel like home.

Finally, my loving gratitude goes to my parents, Mr. Abdul Motin and Mrs. Morzina Begum, and my sister Mrs. Arfi Begum for their countless sacrifices, moral support, and unconditional love. Their unwavering care and prayers have always been the foundation of my strength. Lastly, I am deeply grateful to my two uncles for their never-ending help, support, and care, and thankful to other family members and well-wishers for their love and encouragement.

Date:

Place:

Asfi Ahmed

ABBREVIATIONS AND SYMBOLS

AAc	Acrylic acid
AAm	Acrylamide
AMPS	2-Acrylamido-2-methylpropane sulphonic acid
APS	Ammonium persulfate
a.u.	Arbitrary unit
q_e	Adsorption capacity
BioMEMS	Biomedical microelectrochemical systems
BQ	Benzoquinone
CHN	Carbon, Hydrogen, Nitrogen
cm	Centimeter
CO ₂	Carbon dioxide
CV	Crystal violet
3D	Three-dimensional
DA	Dopamine
DC	Dopamine chrome
DQ	Dopamine quinone
DDS	Drug delivery system
DOPA	3,4-dihydroxyphenylalanine
DMEM	Dulbecco's Modified Eagles Medium
DMSO	Dimethyl sulfoxide
DSC	Differential Scanning Calorimetry
EAH	Electroactive hydrogel
EDTA	Ethylenediamine tetraacetic acid
EDX	Energy-dispersive X-ray spectroscopy
e^-	Electron
Em	Emission
Eq.	Equation
Etc.	et cetera
eV	Electron volt
Ex	Excitation
FBS	Fetal bovine serum

FTIR	Fourier Transform Infrared Spectroscopy
g-C ₃ N ₄	Graphitic Carbon Nitride
g	Gram
HCl	Hydrochloric acid
h	Hour(s)
h ⁺	Hole
H ₂ O	Water
IPA	Isopropanol
KBr	Potassium bromide
kg	Kilogram
KPa	Kilo Pascal
kV	Kilo Volt
DL	Leucodopamine
LED	Light emitting diode
MB	Methylene blue
MBA	<i>N,N'</i> -Methylenebisacrylamide
min	Minute
mA	Milliampere
mg	Milli gram(s)
MJm ⁻³	Megajoule per cubic meter
mL	Millilitre
MO	Methyl orange
MPa	Mega Pascal
MTT	(3-[4,5-dimethylthiazol-2-yl]-2,5 diphenyl tetrazolium bromide)
N ₂	Nitrogen
NaCl	Sodium Chloride
NaOH	Sodium hydroxide
nm	Nanometer
OH ⁻	Hydroxyl radical
O ₂	Oxygen
[•] O ₂ ⁻	Superoxide
OD	Optical density
PAAc	Poly(acrylic acid)

PBS	Phosphate buffered saline
PANi	Polyaniline
PDA	Polydopamine
pH	Potential of hydrogen
K ₂ S ₂ O ₈	Potassium persulphate
PL	Photoluminescence
PTFE	Polytetrafluoroethylene
RhB	Rhodamine
rpm	Revolutions per minute
s	Second
SEM	Scanning electron microscopy
·SO ₄	Sulphate
TEM	Transmission electron microscopy
TGA	Thermogravimetric analysis
UTM	Universal Testing Machine
UV	Ultraviolet
UV-Vis	Ultraviolet-Visible
Vis	Visible
V	Volt
VB	Vitamin B ₁₂
wt%	Weight percentage
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction
θ	Theta
%	Percentage
°C	Degree Centigrade
°	Degree
α	Alpha

LIST OF FIGURES

Chapter 1

Figure 1.1	Different generations of hydrogels over the years	1.3
Figure 1.2	Classification of hydrogels based on different aspects	1.4
Figure 1.3	Different types of stimuli-responsive hydrogel	1.5
Figure 1.4	Graphical representation of swelling behavior of pH-responsive hydrogel at different pH conditions	1.7
Figure 1.5	Graphical representation of bending behavior of an electroactive hydrogel	1.8
Figure 1.6	Pictorial representation showing different properties of hydrogel	1.10
Figure 1.7	Schematic diagram of the mechanism involved in hydrogel adhesion	1.13
Figure 1.8	Different bio-applications of adhesive hydrogel	1.14
Figure 1.9	Schematic diagram showing removal of pollutants in water	1.19

Chapter 2

Figure 2.1	Molecular structure of dopamine	2.2
Figure 2.2	FTIR spectra of (a) monomers, (b) hydrogel; (c) (i) dopamine grafted gelatin-co-poly(acrylic acid) and (ii) gelatin-co-poly(acrylic acid)	2.10
Figure 2.3	XRD spectra of gelatin and hydrogel	2.11
Figure 2.4	TGA spectra of hydrogel	2.11
Figure 2.5	SEM morphology of air dried (a) surface of dried hydrogel, (b) cross-sectional area of swelled hydrogel, (c,d) optical microscopic image of the hydrogel showing porous structure	2.12
Figure 2.6	(a)XPS survey spectra of the hydrogel, (b) c1s, (c) o1s, (d) n1s spectra	2.13
Figure 2.7	(a) Stress-strain curve and (b) young's modulus of the hydrogel at a dopamine concentration of 3wt%, 2wt%, and 1wt%; (c) digital photograph of the hydrogel showing unstretched and stretched hydrogel	2.14
Figure 2.8	Digital photograph of the hydrogel showing (a) adhesion to	2.15

different materials under both dry (upper) and submerged conditions (lower), (b) hydrogel holding weight of 550 g when adhered to two glass surfaces joints (c) hydrogel lifting a weight of 400g when attached to human skin (d) adhesive strength of the hydrogel on different materials under both dry and wet conditions (e) repeatable peel-off test on human skin

Figure 2.9	(a) Digital photograph of the hydrogel showing self-healing behaviour, (b) stress-strain curve of the self-healed hydrogel, (c) schematic representation of the adhesive mechanism of the hydrogel to tissue surface	2.17
Figure 2.10	Swelling behaviour of the hydrogel by varying the composition of dopamine	2.18
Figure 2.11	Haemolysis percentage study of the hydrogel	2.19

Chapter 3

Figure 3.1	Schematic diagram showing crosslinking of polymers to form hydrogel	3.8
Figure 3.2	FTIR Spectra of (a) hydrogel at different DA content and (b) monomers	3.9
Figure 3.3	(a), (b) SEM images of the hydrogel; (c) elemental mapping of carbon, oxygen, and nitrogen of the hydrogel	3.10
Figure 3.4	(a) TGA spectra at different DA content and (b) XRD spectra of hydrogel	3.11
Figure 3.5	Swelling behavior of the hydrogel with (a) varying crosslinker content and (b) at different pH	3.12
Figure 3.6	Tensile strength of the hydrogel at different content of (a) DA, (d) MBA; (b) Young's modulus and (c) toughness of hydrogel with varying content of DA; (e) Young's modulus and (f) toughness of hydrogel with varying content of MBA	3.13
Figure 3.7	Digital image showing (a) a piece of hydrogel lifting a weight of 2.4 kg, (b) showing transparency and flexibility of hydrogel (c) repeatable stretching of the hydrogel	3.14
Figure 3.8	(a) Tensile strength of the hydrogel before and after self-healing; (b) Digital and microscopic image of the hydrogel during self-	3.15

	healing; (c) Schematic diagram of the hydrogel showing self-healing mechanism	
Figure 3.9	(a) Graphical image showing lap-shear adhesion test; (b) optical images showing adhesion of the hydrogel to different substrates; (c) Digital images showing adhesion to human skin; (d) Schematic diagram showing possible interactions of hydrogel with biological tissue surface; (e) adhesive strength of the hydrogel under both dry and wet conditions; (f) Repeatable adhesion of the hydrogel up to 4 cycles	3.17
Figure 3.10	Hemocompatibility of the hydrogel	3.19
Figure 3.11	FTIR spectra of the undegraded and degraded hydrogel	3.20
Figure 3.12	Figure 3.12 (a) cell viability assay of the hydrogel extract at different dilutions, (b) Representative live/dead staining images of L929 fibroblast cells after incubation with the hydrogel extract for 3 days	3.21
Figure 3.13	FTIR spectra of the Vitamin B12 loaded and unloaded hydrogel	3.22
Figure 3.14	Cumulative drug release percentage of the hydrogel at pH of (a) 5.8 and (b) 7.4	3.23

Chapter 4

Figure 4.1	FTIR spectroscopy of (a) bulk g-C ₃ N ₄ , (b) AAm, AMPS monomers and CN5 hydrogel, (c) CN0 hydrogel and CN5 hydrogel	4.10
Figure 4.2	Powder X-ray diffraction pattern of (a) bulk g-C ₃ N ₄ , (b) CN hydrogel and blank hydrogel	4.11
Figure 4.3	(a) TEM image of bulk g-C ₃ N ₄ powder, SEM images of (b) bulk g-C ₃ N ₄ powder, (c) CN0 hydrogel, (d) cross-section CN5 hydrogel	4.12
Figure 4.4	(a, c) UV-Visible absorbance spectra and (b, d) Band gap energy of the synthesized bulk g-C ₃ N ₄ , CN0, and CN5 hydrogel	4.13
Figure 4.5	PL spectra of (a) bulk g-C ₃ N ₄ , (b) CN5 and CN0 hydrogel	4.14
Figure 4.6	TGA spectra of CN5 hydrogel	4.15
Figure 4.7	Swelling behaviour of the hydrogel at g-C ₃ N ₄ concentration of	4.16

	1wt%, 3wt% and 5wt% at pH values of 1, 7 and 10	
Figure 4.8	(a) Compressive strength of the CN hydrogel by varying the composition of g-C ₃ N ₄ , and (b) Repetitive compressive strength of the hydrogel by performing various cycle, and (c) Digital photograph of the hydrogel when compressed with a sharp object	4.17
Figure 4.9	UV-Visible absorbance spectra of the dye treated (a) with hydrogel under light irradiation, (b) with hydrogel under dark condition, (c) with hydrogel, g-C ₃ N ₄ powder for a definite time, (d) Relative removal efficiencies of the dye by the hydrogel as a function of irradiation time under different light condition	4.19
Figure 4.10	Graphical representation of the mechanism of dye removal by the hydrogel	4.20
Figure 4.11	Relative removal efficiencies of the dye by the hydrogel as a function of irradiation time under varying content of g-C ₃ N ₄	4.20
Figure 4.12	Dye removal efficiency of the hydrogel at pH 1, pH 7, and pH 10	4.21
Figure 4.13	Fit plot of (a) pseudo-first order, (b) pseudo-second-order kinetics model for dye degradation	4.22
Figure 4.14	Dye removal efficiency of the hydrogel under the influence of different scavengers	4.23
Figure 4.15	UV-Vis absorbance spectra showing removal of (a) MB dye, (b) RhB dye, (c) MO dye, (d) Removal efficiency of hydrogel towards MB and CV dyes, (e) UV-Vis absorbance spectra showing removal of mix dye of CV/MO under light irradiation at different time intervals, and (f) Digital image showing selective degradation performance by hydrogel from a CV/MO mix dye solution	4.24
Figure 4.16	Repeatable removal efficiency of the hydrogel up to 5 cycles (b) FTIR spectra of the hydrogel before and after recycling and (b) SEM morphology of the hydrogel after recycling	4.26
Figure 4.17	Effect on adsorption amount and removal efficiency of CV dye by the CN3 hydrogel with different adsorbent dosage	4.27
Figure 4.18	Effect on adsorption amount and removal efficiency of CV dye by the CN3 hydrogel at different contact time	4.27

Figure 4.19	Effect on adsorption amount and removal efficiency of CV dye by the CN3 hydrogel with different initial concentration of (a) CV, (b) MB, and (c) RhB dyes	4.29
Figure 4.20	Linear fit plot of (a) Langmuir adsorption isotherm, (b) Freundlich adsorption isotherm model for adsorption of CV dye by the CN hydrogel	4.30
Figure 4.21	Fit plot for (a) Pseudo-first order and (b) Pseudo-second order kinetics for adsorption of CV dye by CN hydrogel with different adsorbent dosage	4.31

Chapter 5

Figure 5.1	Schematic diagram showing the electrodes dipped in an electrolyte solution connected to a DC power source with a strip of hydrogel placed in between the electrodes	5.7
Figure 5.2	Schematic diagram showing hydrogel formation	5.8
Figure 5.3	FTIR spectra of (a) precursors and (b) SAA-gCN-2 hydrogel	5.9
Figure 5.4	SEM image of (a) SAA-gCN-0 and (b) SAA-gCN-2; EDX of (c) SAA-gCN-0 and (d) SAA-gCN-2	5.10
Figure 5.5	TGA spectra of the hydrogels with and without g-C ₃ N ₄	5.11
Figure 5.6	Swelling behavior of hydrogels with time as a function of (a) different ionic strength of NaCl, (b) PANI content, and (c) g-C ₃ N ₄ content	5.13
Figure 5.7	Tensile strength of hydrogels (a) without g-C ₃ N ₄ (SAA-gCN-0) and with g-C ₃ N ₄ (SAA-gCN-1.2) hydrogels, (b) with varied content of g-C ₃ N ₄	5.14
Figure 5.8	(a) Young's modulus and (b) toughness of the hydrogel at varied content of g-C ₃ N ₄	5.15
Figure 5.9	Cyclic compressive strength of SAA-gCN-1.2 hydrogel	5.16
Figure 5.10	(a) Maximum bending angle as a function of ionic strength, (b) rate of bending actuation at varied ionic strength	5.17
Figure 5.11	Rate of bending actuation at varied thickness	5.18
Figure 5.12	Repeatable bending actuation of the hydrogel on cyclically applying electric field	5.18

LIST OF TABLES

Chapter 1

Table 1.1	Table showing various interactions and reactions of dopamine	1.16
-----------	--	------

Chapter 2

Table 2.1	Detailed composition of the hydrogel with varied compositions of DA	2.4
Table 2.2	Chemical compositions of C, N, and O	2.13
Table 2.3	Comparison of the adhesive strength of the synthesized hydrogel-based adhesive under wet condition with previously synthesized hydrogel reported in literature.	2.16

Chapter 3

Table 3.1	Detailed composition of the hydrogels with varying DA concentration	3.3
-----------	---	-----

Chapter 4

Table 4.1	Detailed composition of the CN hydrogels	4.5
Table 4.2	Elemental composition of CN0 and CN5 hydrogel obtained from CHN analysis.	4.13
Table 4.3	Compressive stress along with strain and Young's modulus are shown with varying compositions of g-C ₃ N ₄	4.17
Table 4.4	Isotherm parameters for adsorption of CV dye by CN hydrogel	4.30
Table 4.5	Kinetic parameters for adsorption of CV dyes by CN hydrogel with different adsorbent dosage	4.31

Chapter 5

Table 5.1	Table 5.1. Detailed compositions of the precursors used in the formation of hydrogels	5.4
-----------	---	-----

LIST OF SCHEMES

Chapter 2

Scheme 2.1	Plausible mechanism involved in the formation of hydrogel	2.8
-------------------	---	-----

Chapter 3

Scheme 3.1	Plausible mechanism of hydrogel formation	3.8
-------------------	---	-----

Chapter 4

Scheme 4.1	Plausible mechanism of hydrogel formation	4.9
-------------------	---	-----

Chapter 5

Scheme 5.1	Plausible mechanism involved in the formation of hydrogel	5.7
-------------------	---	-----