Dedicated to…..

My Family & Friends

KEYWORDS:

Low molecular weight gelator, Supramolecular Gels, Polymorphism, Multi-stimuli response, Gel phase crystallization, Concomitant crystallization, Gel screening, Conformational flexibility, In-situ gelation.

Declaration of Academic Integrity

I do hereby declare that this thesis titled "***Understanding Supramolecular Self-assembly of Bis-Urea Functional Low Molecular Weight Gel for Pharmaceutical Crystallization***" represents my ideas in my own words and where other's ideas or words have been included, I have adequately cited and referenced the original sources. I also declare that I have adhered to all principles of academic honesty, integrity and have not misrepresented or fabricated or falsified any idea/data/fact/source in my submission. I understand that any violation of the above will be caused for disciplinary action as per the rules and regulations of the Institute.

Due acknowledgement to all the related data used from different sources in order to support my research findings have been made wherever necessary. All funding agencies have been duly acknowledged for providing research grants to carry out my research work smoothly.

Date: 21-03-2025

Place: Tezpur University

Himanshu Sharma

(Himanshu Sharma)

Department of Chemical Sciences

Tezpur University

तेजपुर विश्वविद्यालय / TEZPUR UNIVERSITY
(संसद के अधिनियम द्वारा स्थापित केंद्रीय विश्वविद्यालय)
(A Central University established by an Act of Parliament)

Dr. Bipul Chandra Sarma
Assistant Professor
Department of Chemical Sciences

Mobile: 91-8399936229
Phone: 03712 275066
Email: bcsarma@tezu.ernet.in

CERTIFICATE FROM SUPERVISOR

This is to certify that the thesis entitled "***Understanding Supramolecular Self-assembly of Bis-Urea Functional Low Molecular Weight Gel for Pharmaceutical Crystallization***" submitted to the School of Sciences, Tezpur University in partial fulfillment for the award of the degree of Doctor of Philosophy in Chemical Sciences is a record of research work carried out by Mr. Himanshu Sharma under my supervision and guidance at Department of Chemical Sciences, Tezpur University, Assam. He has successfully completed the work.

He has fulfilled all the requirements for submitting the thesis for award of the Degree of Doctor of Philosophy in Science. All help received by him from various sources have been duly acknowledged. No part of this thesis has been submitted elsewhere for award of any other degree.

Date: 21/03/2025
Place: Tezpur.

(Dr. Bipul Chandra Sarma)
Supervisor
School of Sciences
Department of Chemical Sciences
Tezpur University
Assam, India – 784028

ACKNOWLEDGEMENT

This journey is all about "One foot inside the boundary of the known, the other is just outside" in the world of gels. This work would not have been possible without the unwavering support and encouragement of numerous individuals. It is with great honour that I convey my profound gratitude to all those who have generously offered their assistance and steadfastly supported me throughout my doctoral journey.

I express my deepest appreciation to my esteemed supervisor, Dr. Bipul Chandra Sarma, for his invaluable suggestions, constant encouragement, and enlightening mentorship throughout my research tenure.

I remain obliged to my doctoral committee members, Prof. Ashim Jyoti Thakur and Dr. Sanjeev Pran Mahanta Sir for their valuable input and suggestions during the development of this thesis. Their thoughtful critiques and expert advice have significantly enriched the quality of my work. I take this opportunity to offer thanks to the Head, Department of Chemical Sciences, Tezpur University for providing access to the departmental facilities for my research work.

My sincere appreciation to all the technical and non-teaching staff of the department of Chemical Sciences, Food Engineering & Technology, and Sophisticated Analytical Instrumentation Centre (SAIC) Tezpur University for their constant help throughout this journey.

I extend my warm thanks to the past and present members of "Pharmaceutical Crystallization and Porous materials Laboratory" Dr. Rajiv Khatioda, Dr. Basanta Saikia, Dr. Pranita Bora, Dr. Nazima Sultana, Dr. Kasturi Sarmah, Dr. Manali Dutta, Dr. Rupa Pegu, Dr. Debabrat Pathak, Dr. Tamrat Yimenu Zeleke, Dr. Kankana Baruah, Bikash, Archita, Priya, Akankshya, Swashat and Neeha for their love and encouragement making this journey fun and beautiful.

I express my heartiest love to my friends Gunajit, Hirak Baishya, Nilutpal, Hirok Sarmah, Sanjay , Tapas, Partha (Barman), Sourav, Barasha, Nilutpol, JP, Priyanka, Suravi, Nitumani, Debanga Da, Nayab, Rituparna, Partha (Churi), Suranjana, Rashmi, Asfi, Sudhamoyee Ba, Sahtaz, Raju, Bikash, Raktim Da, Anup Da, Prantika Ba, Shubham, Veda, Niharika, Harsh Bhaiya for their good wishes, support, and motivation in all aspects of my research journey. I would like to express my gratitude towards Dr. Dhruba Jyoti Talukdar for the unconditional help in providing materials characterization.

I gratefully acknowledge DST-SERB (CRG/2019/004946), Tezpur University for the Research and Innovation Grant (RIG-2020) alongside the Institutional fellowship for providing financial assistance during my Ph.D tenure. I would also like to acknowledge INUP i2i program at CeNSE, IISc Bangalore and centre for Nanotechnology, IIT Guwahati for funding and training.

I am grateful to Prof. Parthasarathi Dastidar and his research group at IACS Kolkata for initial training in gel preparation methods and its characterization techniques. I would also like to acknowledge Dr. Akshai kumar A.S. and Prof. Pankaj Tiwari, IITG for mentorship and research collaboration.

Furthermore, I would like to extend my sincere appreciation and loving gratitude to my family for their tireless efforts in accompanying me on this arduous journey. Their constant prayers and moral support have served as a pillar of support for my professional aspirations. Their belief in my abilities has consistently given me the strength to overcome obstacles encountered during this pursuit.

Thank you everyone.

March, 2025

Tezpur University *Himanshu Sharma*

ABBREVIATIONS AND SYMBOLS

%	percentage
ºC	degree centigrade
µm	micrometer
Å	angstrom
π	pi
µL	microlitre
a.u.	arbitrary unit
cm	Centimeter
1D	One dimensional
3D	Three dimensional
ADA	Aminoacetaldehyde diethyl acetal
API	Active Pharmaceutical Ingredients
ASP	Aspirin
BFDH	Bravais-Fridel, Donnay–Harker
CAF	Crystallization of caffeine
CAQ	Cholesteryl anthraquinone-2-carboxylate
CBZ	Carbamazepine
CS	Clear solution
DBS	Di-benzylidene sorbitol
DEE	Diethyl ether
DMA	Dimethyl acetamide
DMF	Dimethylformamide
DMSO	Dimethylsulfoxide
DSC	Differential scanning colorimetry
ETZ	Ethenzamide
FESEM	Field Emission Scanning Electron Microscopy
FG	Flexible Group

FLU	Flufenamic acid
FBRM	Focused Beam Reflectance Measurement
FT-IR	Fourier Transform Infrared spectroscopy
g	gram
G	Gel
GBH	Glycerol tertiary Butanol and Water
Gr	Grinding
HSP	Hansen solubility parameters
HC	Heat Cool
HRMS	High resolution mass spectrometry
H	hour
HCl	Hydrochloric acid
HSA	Hydroxy Stearic Acid
HH	Hyperhelical
HRXRD	High resolution X-ray diffractometer
IND	Indomethacin
IPE	Di-isopropyl ether
I	Insolubility
IG	Instant gelation
ISB	1,3-bis(2-isocyanato-2-propy) benzene
K	Kelvin
Kcal/mol	Kilocalorie per mole
kV	Kilovolt
LC	Liquid crystalline
LVR	Linear Viscoelastic Regions
LMWG	low molecular weight gel
MFA	Mefenamic acid
MHz	Megahertz
mA	milliampere

mg	milligram
mm	millimetre
mmol	Milli mole
mW	milliwatt
MGC	Minimum Gelation Concentration
nm	nanometre
NCA	Niclosamide
NIF	Nifedipine
NMR	Nuclear Magnetic Resonance
ORTEP	Oak Ridge Thermal-Ellipsoid Plot
PVM	particle vision measurement
Pa	Pascal
pH	potential of hydrogen
PXRD	Powder X-Ray Diffraction
Rad	Radian
Rad/s	Radian per second
SEM	Scanning Electron Microscopy
SAM	Self Assembeld Monolayer
Sh	Shaking
SCXRD	Single Crystal X-Ray Diffraction
SG	Slow gelation
SANS	Small Angle Neutron Scattering
SAXS	Small Angle X-Ray Scattering
S	Solubility
So	Sonication
THF	Tetrahydrofuran
TGA	Thermogravimetric analysis
TLC	Thin Layer Chromatography
TBA	Tolbutamide

TFA	Tolfenamic acid
UV-Vis	Ultraviolet-visible
w/v	Weight / Volume
WAXS	Wide angle X-Ray Scateering

LIST OF FIGURES

Chapter 2

Chapter 3

Chapter 4

Chapter 5

LIST OF SCHEMES

LIST OF TABLES