

Declaration by the Candidate

The candidate certifies that the thesis entitled "Development of multiresponsive carbon dots based aerogel and its application in food analysis" submitted to the School of Engineering, Tezpur University in partial fulfilment for the award of the degree of Doctor of Philosophy in the Department of Food Engineering and Technology is a record of research work carried out by me under the supervision of Prof. Poonam Mishra.

All assistance received from various sources have been duly acknowledged.

No part of this thesis has been submitted elsewhere for the award of any other degree.

Date: 14|11|2025

Place: Texpuor University, Assam

Subhamoy Dhua (Subhamoy Dhua)

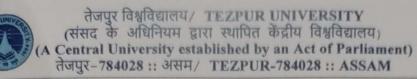
Registration No. TZ203825 of 2021

Roll No. FEP20110

Department of Food Engineering and Technology

School of Engineering, Tezpur University

Napaam 784028, India



(सर्बोत्तम विश्वविद्यालय के लिए कुलाध्यक्ष पुरस्कार, 2016 औरमारत के 100श्रेष्ठ उच्च शिक्षण संस्थानों में पंचम स्थान प्राप्त निश्वविद्यालय) (Awardee of Visitor's Best University Award, 2016 and 5th among India's Top 100 Universities, MHRD NIRF Ranking, 2016)

Dr. Poonam Mishra Professor & Head

Department of Food Engineering and Technology

Email: poonam@tezu.ernet.in Phone: 03712-267007

Certificate of the Supervisor

This is to certify that the thesis entitled "Development of multiresponsive carbon dots based acrogel and its application in food analysis" submitted to the School of Engineering, Tezpur University in partial fulfillment for the award of the degree of Doctor of Philosophy in Food Engineering and Technology is a record of research work carried out by Mr. Subhamoy Dhua under my supervision and guidance.

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.

Signature by Supervisor

Poonam Mishra

Acknowledgements

As I reach the end of my Ph.D. journey, I am overwhelmed with gratitude for all the support, guidance, and encouragement that I have received along the way. This thesis is the result of years of hard work and dedication, as well as the combined efforts of numerous remarkable individuals.

At the outset, I extend my heartfelt gratitude to my supervisor, Prof. Poonam Mishra, Department of Food Engineering and Technology, Tezpur University, Tezpur, Assam, for her invaluable guidance, unwavering support, and expert insights throughout the completion of this thesis. Her encouragement and thoughtful recommendations have been instrumental in shaping this work from its inception to completion, and I deeply appreciate her efforts in organizing my research systematically and within the prescribed timeframe. I am deeply grateful for her love, care, and valuable recommendations throughout the entire process, from beginning to end.

I sincerely thank Hon. Vice-Chancellor Prof. Shambhu Nath Singh, Tezpur University, Tezpur, Assam, for providing me with the opportunity to undertake this research. My gratitude also extends to Prof. S. C. Deka, Dean of the School of Engineering, and Dr. Biren Das, Controller of Examinations, and Prof. Poonam Mishra, HoD, Department of Food Engineering and Technology, Tezpur University, for their continuous support during my thesis work.

I am profoundly grateful to my Doctoral Committee members, Dr. Nishant R. Swami Hulle, and Dr. Amit Baran Das (On Lien) from the Department of Food Engineering and Technology, Tezpur University, for their valuable suggestions and encouragement at various stages of my research.

Heartfelt thanks are due to the members of the Departmental Research Committee for extending all sorts of help and guidance throughout my research work. I acknowledge the faculty members of the Department of Food Engineering and Technology, including Prof. Charu Lata Mahanta (Retd.), Prof. Sankar Chandra Deka, Prof. Manuj Kr. Hazarika, Prof. B. Srivastava, Prof. L. S. Badwaik, Prof. Nandan Sit, Dr. Nishant R. Swami Hulle, Dr. Amit Baran

Das (On Lien), Dr. Tabli Ghosh, Dr. Soumya Ranjan Purohit, and Dr. Nickhil C., for their invaluable input and encouragement throughout my Ph.D. journey. My sincere appreciation extends to the External Committee members, Prof. Bhabesh Deka, Department of Electronics and Communication Engineering and Dr. Rupak Mukhopadhyay, Department of Molecular Biology and Biotechnology, Tezpur University, Assam for their guidance and assistance throughout my research.

I am grateful to the technical staff, Dr. Dipankar Kalita, Dr. Arup Jyoti Das, Mr. Labadeep Kalita, and Mr. Bhaskar J. Kalita, as well as the non-technical staff, Mr. Krishna Borah and Mr. Anjan Keot, for their assistance during my research and departmental work.

I am indebted to the All India Council for Technical Education (AICTE) for providing me with financial assistance through the AICTE Doctoral Fellowship (ADF), and I appreciate the staff of the AICTE cell at Tezpur University, particularly Mr. Debojit Sharma, for diligently handling scholarship-related tasks and addressing any grievances.

I would like to express my sincere gratitude to my batchmates Dr. Arun Kumar Choudhary, Ms. Mwchangti Debbarma, Ms. Pinky Deka, and Ms. Zola Baruah and my dear friends Arunava, Ananya, Sunil, and Asif for their continuous support and encouragement during my research journey.

I extend my heartfelt appreciation to my lab mates (Ms. Manisha Medhi, Dr. Arun Kumar Gupta, Dr. Monica Yumnam, Ms. Parismita Koch, Ms. Krishna Gopalakrishnan, Mrs. A. Sangeeta, Mrs. Mridusmita Burman, Mr. Keshram Dulait, and Ms. Suchitra Keisham), and my juniors for their continuous support throughout my research and the juniors, for their unwavering support throughout my research.

I am deeply grateful to Mr. Tapas Saha, Dr. Kunal Pradhan, Mr. Tamal, Ms. Priyanjana, and Dr. Debanjan Saha, Research Scholars in the Department of Computer Science and Engineering and Molecular Biology and Biotechnology, Tezpur University, for their invaluable support, and I wish to acknowledge the support of all those, whether directly or indirectly involved, whose names may not have been explicitly mentioned.

Lastly, my deepest gratitude goes to my beloved parents, Mr. Madhusudan Dhua and Mrs. Shikha Dhua, my elder brother, Mr. Tanmoy Dhua and my wife Mrs. Puja Rakshit and

my entire family for their unconditional love, encouragement, care, and unwavering support throughout my Ph.D. journey. Their patience, understanding, and motivation have been invaluable in completing this research.

Above all, I am profoundly grateful to the Almighty for the blessings that have guided me throughout my Ph.D. journey.

Subhamoy Dhua

Subhamoy Thua

List of Tables

Table 2.1 Polysaccharide based aerogel, their characteristics, and application21-22
Table 2.2 Corn starch based aerogel, their characteristics, and application23-24
Table 2.3 Protein based aerogel, their characteristics, and application27
Table 2.4 Mucilage based aerogel, their characteristics, and application30
Table 2.5 Hybrid/Composite food aerogel and their potential application32-33
Table 2.6 Functional compound impregnated aerogel
Table 2.7 Carbon dots (CDs) based aerogel, their characteristics and application57-59
Table 3.1 Thermal properties of aerogel
Table 4.1 Thermal properties of freeze and microwave dried aerogel
Table 4.2 Physico-functional and mechanical properties of standardized SCCO ₂ , freeze and microwave dried aerogel
Table 5.1 G/B values of crushed aerogel immersed in water and supernatant water153
Table 6.1 The linearity and the uncertainty of measurement of the calibration curve173
Table 6.2 Accuracy, precision, and % recovery of CDFA
Table 6.3 Comparison of formalin determination in fish fillets through CDFA and Acetylacetone method
Table 6.4 Comparison of formalin determination in raw fish through CDFA and Acetylacetone method

List of Figures

Fig. 2.1 Necessary steps involved in the development of aerogel
Fig. 2.2 Types of Aerogel
Fig. 2.3: Loading of functional compound at different stages of aerogel preparation36
Fig. 2.4: Loading of carbon dots (CDs) at different stages of aerogel preparation46
Fig. 3.1 Development of corn starch based aerogel90
Fig. 3.2 Solvent exchange of hydrogel made of different starch concentrations95
Fig. 3.3 Shrinkage during each stage of solvent transfer (a) and total shrinkage after solvent transfer and drying (b)96
Fig. 3.4 Density and porosity of the developed aerogel96
Fig. 3.5 SEM images of control (a, & b), 5 % glycerol added (c & d) and 10 % glycerol added (e, & f) aerogel
Fig. 3.6 Recompressibility of control (0) and 5, 7.5, and 10 % glycerol added aerogel99
Fig. 3.7 Recompressibility of control (0) and 5, 7.5, and 10 $\%$ glycerol added aerogel99
Fig. 3.8 X-ray diffraction pattern and crystallinity index (CI) of control (0) and 5, 7.5, and 10 % glycerol added aerogel
Fig. 3. 9 Hygroscopicity of control (0) and 5, 7.5, and 10 % glycerol added aerogel102
Fig. 3.10 Water absorption capacity at 30 min (WAC30 min) and 24 h (WAC24 h) of aerogel
Fig. 3.11 Reusability of control (0) and 5, 7.5, and 10 % glycerol added aerogel104
Fig. 3.12 Principal component analysis (PCA) plot of characteristics of aerogel105
Fig. 4.1 Developed aerogel through microwave drying at different power level114
Fig. 4.2 Total shrinkage of the freeze and microwave dried aerogel
Fig. 4.3 Density and porosity of freeze dried (a) and microwave dried (b) aerogel119
Fig. 4.4 SEM images of control (a, b, & c), 5 % glycerol added (d, e, & f) and 10 % glycerol added (g, h, & i) freeze dried aerogel
Fig. 4.5 SEM images of control (a, & b), 5 % glycerol added (c & d) and 10 % glycerol added (e, & f) microwave dried aerogel
Fig. 4.6 FESEM images of 5 % glycerol added (a, & b) and control (c & d) microwave dried aerogel
Fig. 4.7 Compressive strength (a and b) of control (0) and glycerol added (5, 7.5, & 10 %) freeze dried (FD) aerogel.

Fig. 4.8 Compressive strength of control (0) and glycerol added (5, 7.5, & 10 %) microwave dried (MD) aerogel
Fig. 4.9 Recompressibility of control (0) and glycerol added (5, 7.5, & 10 %) freeze dried (FD) aerogel
Fig. 4.10 Recompressibility of control (0) and glycerol added (5, 7.5, & 10) microwave dried (MD) aerogel
Fig. 4.11 X-ray diffraction pattern and crystallinity index (CI) of control (0) and glycerol added (5, 7.5, & 10) freeze dried (FD) aerogel
Fig. 4.12 X-ray diffraction pattern and crystallinity index (CI) of control (0) and glycerol added (5, 7.5, & 10) microwave dried (MD) aerogel
Fig. 4.13 Hygroscopic behaviour of freeze dried (a) and microwave dried aerogel (b)129
Fig. 4.14 Water absorption capacity at 30 min (WAC30 min) and 24 h (WAC24 h) of freeze dried (a) and microwave dried aerogel (b)
Fig. 4.15 Reusability of control (0) and 5, 7.5, & 10 % glycerol added freeze dried (FD) aerogel
Fig. 4.16 Reusability of control (0) and 5, 7.5, & 10 % glycerol added microwave dried (MD) aerogel
Fig. 4.17 Principal component analysis (PCA) bi-plot of the characteristics of freeze dried (FD) and microwave dried (MD) aerogel
Fig. 5.1 Development process of carbon dots (CDs) based aerogel142
Fig. 5.2: Loading of different concentration of carbon dots (CDs) in aerogel142
Fig. 5.3: Fluorescence spectra of CDs developed with ammonia (WA) (a) and without ammonia (WoA) (b)
Fig. 5.4: Fluorescence behaviour (a) and FTIR spectra (b) of the developed CDs148
Fig. 5.5: UV and fluorescence spectra of CDs solution (a) and fluorescence spectra at different concentration of CDs (b)
Fig. 5.6: C-NMR spectra of CDs solution (a) and CDs precursor solution (b)
Fig. 5.7: H-NMR spectra of CDs solution (a) and CDs precursor solution (b)
Fig. 5.8: TEM images, particle size distribution (a) and SAED pattern (b) of CDs151
Fig. 5.9: Appearance and fluorescence nature of the developed aerogel
Fig. 5.10: Loading value and G/B (green/blue) value (a) and fluorescence spectra (b) of the developed CDs based aerogel
Fig. 5.11: Cross-sectional images of transversally and vertically cut aerogel (a) and crushed carbon dots based aerogel (CDA) in water

Fig. 5.12: FTIR spectra of CS (corn starch), CA (control aerogel without CDs), CDA (CDs based aerogel), and CDS (carbon dots solution)
Fig. 5.13: X-Ray diffraction pattern of CA (control aerogel without CDs) and CDA (CDs based aerogel)
Fig. 5.14: DSC (differential scanning calorimetry) thermogram of CDs based aerogel155
Fig. 5.15: Green/blue (G/B) value v/s pH relationship of CDs based aerogel156
Fig. 6.1 Development process of carbon dots based functional aerogel (CDFA)165
Fig. 6.2 Common Carp (<i>Cyprinus carpio</i>) fish
Fig. 6.3 Fluorescence spectra (a) and fluorescence intensity (b) of CDs in the presence of Tollens reagent (TR)
Fig. 6.4 Plot between intensity difference (B–B0) and concentration of FA (a) and images of CDs based aerogel (CDA) (left) and CDs based functional aerogel (CDFA) without (middle) and with FA (right).
Fig. 6.5 Calibration curve of CDFA response to different formalin concentration173

Abbreviations	Full Form
AA	Ascorbic Acid
AChE	Acetylcholinesterase Enzyme
AFM	Atomic Force Microscopy
AgNCs	Silver Nanoclusters
AgNPs	Silver Nanoparticles
ATCh	Acetylthiocholine
ATP	Adenosine Triphosphate
В	Blue Value
BD	Bulk Density
BET	Brunauer-Emmett-Teller
BPEI	Branched Polyethylenimine
BSM	Balangu seed mucilage
CDA	Carbon Dots based Aerogel
CDFA	Carbon Dots based Functional Aerogel
CDs	Carbon Dots
CI	Crystallinity Index
CMs	Carbon Microspheres
CNC	Cellulose Nano Crystals
CNF	Cellulose Nano Fibre
CQD	Carbon Quantum Dot
CV	Cyclic Voltammetry
D	Density
DDL	Diacetyldihydrolutidine
DE	Degree of Esterification
DMF	Dimethylformamide
DPPH	2,2-Diphenyl-1-Picrylhydrazyl
DSC	Differential Scanning Calorimetry
EDC	N-(3-Dimethylaminopropyl)-N'-Ethylcarbodiimide Hydrochloride
EDEA	2,2-(Ethylenedioxy)-bis-(Ethylamine)
EFSA	European Food Safety Authority
FA	Formalin

FD Freeze Dried

FSSAI Food Safety and Standard Authority of India

FT-IR Fourier Transform Infrared Spectroscopy

G Green Value

GA Graphene Aerogel

GAE Gallic Acid Equivalent
GCE Glassy Carbon Electrode

GC-MS Gas chromatography Mass Spectrometry

GO Graphene Oxide

GRAS Generally Recognized as Safe

HPLC High Performance Liquid Chromatography

HRTEM High-Resolution Transmission Electron Microscopy

IARC International Agency for Research in Cancer

IBR Intensity of Backscattered Radiation

IL Ionic Liquid

IUPAC International Union of Pure and Applied Chemistry

LED Light Emitting Diode
LOD Limit of Detection

LOQ Limit of Quantification

MD Microwave Dried

MPL Maximum Permissible Limit

MTMS Methyltrimethoxysilane

MW Molecular Weight

NFC Nanofibrillated Cellulose
NHS N-Hydroxysuccinimide

NMMO N-Methylmorpholine-N-Oxide

NMR Nuclear Magnetic Resonance

OP Organophosphate Pesticides

PCA Principal Component Analysis

PDI Polydispersity Index

PET Photoinduced Electron Transfer

PL Photo Luminescent

PMAA Polymethacrylic Acid

PP Polypropylene

PVA Poly Vinyl Alcohol

QDs Quantum Dots
QY Quantum Yield

RLS Resonance light scattering
RSD Relative Standard Deviation
R² Coefficient of Determination

SAED Single Area Diffraction

SC Supercritical

SCD Supercritical CO₂ Dried SCE Supercritical Extraction

SEM scanning electron microscopy

SP Soy Protein

SSA Specific Surface Area

TBA Tert-Butanol

TEM Transmission Electron Microscopy

TEOS Tetraethyl Orthosilicate
TMAO Trimethylamine Oxide

TR Tollens Reagent

TTA Thenoyltrifluoroacetone

UV Ultraviolet

VOCVolatile Organic CompoundWACWater Absorption CapacityWHOWorld Health Organization

WLE White Light Emitting

XRD X-ray diffraction

YMBE Yerba-Mate Based Extract

ε Porosity