

**DEVELOPMENT OF SUSTAINABLE
BIOMATERIALS FOR BIOMETHANE
AND CARBON DIOXIDE
ADSORPTION**

KOMALKANT ADLAK



CENTRE FOR RURAL DEVELOPMENT AND TECHNOLOGY

INDIAN INSTITUTE OF TECHNOLOGY DELHI

JULY 2024

© Indian Institute of Technology Delhi (IITD), New Delhi, 2024

DEVELOPMENT OF SUSTAINABLE BIOMATERIALS FOR BIOMETHANE AND CARBON DIOXIDE ADSORPTION

by

KOMALKANT ADLAK

CENTRE FOR RURAL DEVELOPMENT AND TECHNOLOGY

Submitted

**In fulfilment of the requirements of the degree of Doctor of
Philosophy**

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

JULY 2024

*Dedicated to my
grandparents, parents,
and
Late Dr. Ram Chandra*

CERTIFICATE

This is to certify that the thesis entitled "Development of Sustainable Biomaterials for Biomethane and Carbon dioxide Adsorption", being submitted by Mr. Komalkant Adlak to the Indian Institute of Technology Delhi for the award of "Doctor of Philosophy" is a record of bonafide research work carried out by him. He has worked under our guidance and supervision and has fulfilled the requirements for the submission of this thesis. To the best of our knowledge, the results contained in this thesis have not been submitted in part or full to any other university or institute for the award of any degree or diploma.

Prof. Virendra Kumar Vijay
Professor,
Centre for Rural Development
and Technology,
Indian Institute of Technology,
Delhi
Hauz Khas, 110016, New Delhi

Prof. Kamal Kishore Pant
Professor,
Department of Chemical
Engineering,
Indian Institute of Technology,
Delhi
Hauz Khas, 110016, New Delhi

Acknowledgements

The serendipity of life, guided by God's grace, has finally brought me to the end of the PhD journey. The last five and a half years of my Ph.D. journey have been a fantastic roller-coaster ride with extensive learning, intense emotions, countless deadlines, conferences, and extracurricular work. As I reflect, I realize that a Ph.D. is not just a degree to achieve but a journey worth experiencing. It has transformed me into a resilient and passionate individual, ready to take on life's challenges while striving to make a difference in the world.

On this occasion, I want to express my heartfelt gratitude to all the people who played a role, knowingly or unknowingly, in attaining my Ph.D. First and foremost, I remember the **late Dr. Ram Chandra**, who was the reason I could complete this degree. He has been an inspiration, and I've been fortunate to have spent precious years around him; his loss has been irreplaceable for me. Continuous and unwavering support from **Prof. Virendra Kumar Vijay** and **Prof. Kamal Kishore Pant** has been crucial. **Prof. Vijay's** trust and belief in me have provided opportunities to flourish when needed most in a life filled with confidence, making me more fearless in exploring new opportunities. He has been a silent and supportive person throughout the journey.

I also appreciate **Prof. Vivek Kumar** for being an integral part of this journey. I thank **Prof. Ondřej Mašek** for hosting, giving an opportunity and guiding the work at the University of Edinburgh, UK. I sincerely thank the members of my student research committee, Prof. Anushree Malik, and Prof. Sunil Kumar Khare for giving valuable feedbacks. All the faculty members of the centre, Prof. PMV Subbarao, Prof. S.K. Naik, Prof. VM Chariar, Prof. Satyawati Sharma, Prof. Kavya Dassora, Prof. Hariprasad P., Prof. J. K. Sahu, for their valuable time, thoughtful support, and insightful remarks.

I want to acknowledge the Council for Scientific and Industrial Research (CSIR) for funding the PhD study. I would also like to mention The Foreign, Commonwealth and Development Office, United Kingdom, for granting me a one-year fellowship to carry out research work at the UK Biochar Research Centre, the University of Edinburgh. This fellowship has been crucial in supporting my research and academic pursuits. The research infrastructure provided by IIT Delhi and UoE has been key to achieving this feat.

My colleagues and seniors at IIT Delhi have been amazing throughout the journey. I am grateful to research group alumni **Dr. Bhaskar Jha, Dr. Vandit Vijay, Dr. Pushendra, Dr. Abhinav Trivedi**. Thank you to **Biju I.K., Dr. Madan Kumar, Dr. Praveen, Sameer, Subodh, Tinku Casper D'Silva, Sachin Bhujbal, Amit Kumar Sharma, Saptashish Deb and Sharang Shrama**. The laboratory experiment and PhD journey were impossible without the support of **Ravindar, Vinod, Mahesh, Y. P. Singh, Seema, Ramesh** and the whole CRDT staff. Also, I am thankful to **Dr. Christian Wurzner** and **Mr. Thomas Robinson**, UK Biochar Research, for their support.

The Ph.D. journey has been tumultuous and replete with emotional highs and lows, a journey that remains incomplete without the presence of steadfast friends. I sincerely thank **Mr. Pinaki Dasgupta** for his unwavering support and invaluable guidance; his steadfast encouragement has consistently been my pillar of strength. I express my profound gratitude to **Anwita, Yash, Ritesh, Dr. Vinayak, Rahul, Dr. Gaurav, Dr. Lahur, Dhanajay, Pratishtha** for being integral parts of this journey and cherishing every emotion with me. These individuals transformed the journey into a meaningful and worthwhile experience.

Last but not least, my family deserves my utmost gratitude. Starting from being dependent on agriculture, my **grandparents (Sh. Arjun Rao Adlak, Smt. Dwarka Adlak, Late Sh. Rambhau Deshmukh, Smt. Mainabai Deshmukh)** did everything in their power to raise and educate my parents. The continuous perseverance for excellence from my parents **Sh. Prabhu Adlak and Smt. Sunita Adlak** has enabled me to achieve this degree. The persistent support from my parents and brother **Mr. Chetan Adlak** has been key. I am grateful to Mamaji **Dr. Arun Kumar Deshmukh**, for being a continuous source of inspiration and guidance. I am also proud to have support from all my **uncles, aunts, and cousins**, and I look forward to cherishing more moments together in further life.

I am deeply grateful to my Guru **Pt. Shriram Sharma Acharya** and Maa **Gayatri**, thank you for guiding and strengthening my inner wisdom to continuously pursue what is right in life.

With heartfelt appreciation,

Komalkant Adlak

Abstract

The unprecedented challenge of climate change outlines the failure on environmental parameters despite remarkable developments in the previous two and a half centuries. Fossil fuels have been a convenient energy source for long periods; however, their consumption has majorly contributed to mounting levels of carbon emissions. Now, developing alternative energy sources is imperative and anaerobic digestion technology has a crucial role in the transition. Renewable methane is obtained from organic waste digestion and biogas separation thereafter. The key challenge is storing the methane-rich gas where it is currently stored by compressing it at a very high pressure of 20-25 MPa. Alternatively, in adsorption based storage where similar amount of gas (as of compression) is stored under mild pressure. The storage is obtained in a porous material-filled cylinder, where gas is adsorbed under pressure on the microporous surface. The density of gas storage is higher in microporous materials than in simple compression, which reduces the storage pressure requirement. The porous materials obtained from biomass have been another advantage of this technology since they are renewable, and utilising such a source also reduces the environmental burden. The biomass is first carbonised to obtain char and then converted to high surface area activated carbons using activation. In this thesis work, the focus have been on such biomass-based sustainable materials and tailored their production for high-efficiency gas storage and capture.

In the first Objective, biochar's combined physical and chemical activation was explored. Potassium hydroxide has been used for chemical activation and steam as physical activation source to reduce chemical use to obtain high surface area. The combined activated carbons have a surface area of $963 \text{ m}^2\text{g}^{-1}$ compared to only chemically activated $764 \text{ m}^2\text{g}^{-1}$. For the analysis of biomethane adsorption, the pellets were prepared using bentonite and Kaolin. The best results obtained were found with 20% (weight basis) of Kaolin with gravimetric methane uptake of 0.28 g/g and volumetric methane uptake of 231 v/v with 177 v/v of deliverable capacity.

In second work, the various nature biomass has been selected and pore formation pattern has been studied. The activated carbons were synthesized from ten different biomasses under similar activation conditions. The selection has taken place based on the nature (hardness/softness) and their local availability. These prepared carbons were then analysed with different techniques such as BET, X-ray diffraction, Scanning electron

microscope, and FT-infrared. The biomass's nature has an influence on the pores' distribution on the material and hence different methane adsorption capacities. The highest volumetric ratio of ~251 v/v (uptake), ~217 v/v (delivery) has been recorded on coconut shell AC, whereas the highest gravimetric uptake of ~0.32 g/g at 6.5 MPa was recorded on karanja shell AC.

Next, the efforts were made to address the challenge of lower desorption of the gas while system operation since thermodynamics discourages overall technological development. The adsorption was carried out at room temperature and desorption by placing gas-filled cylinders in a water bath at different temperatures maintained at 40, 45, 50 and 55 °C. At room temperature, nearly 54% of adsorbed gas is released whereas, at 55 °C, approximately 80% of the gas is recovered.

Adsorption-based technology was also assessed using Life cycle Assessment as a tool including its comparison with compression-based storage system. The activated carbon-based storage system has been efficient on the environmental parameters however few points to be considered. Lower pressure storage reduces cylinder thickness increases safety as well it also reflects on the midpoint indicators as well. However, the chemical synthesis of activated carbons is still a key concern, and recycling waste during production is crucial in reducing environmental burdens.

Lastly, the activated carbon was prepared with focusing on its heteroatom content and improving sustainability. In this work, the materials have been explored for their application in carbon dioxide capture e.g. from biogas and post-combustion gases. The material synthesised has a considerable presence of nitrogen and sulphur with high oxygen content. They are prepared via the hydrochar route and activated using $\text{KCl} + \text{Na}_2\text{S}_2\text{O}_3$ to activate hydrochar, resulting in high surface area and porosity. The prepared activated carbons achieved high CO_2 uptake of up to 3.5 mmol/g. The influence of heteroatom on CO_2 was also investigated in the study. These materials also shows high regenerable capacity over five cycles.

Further, large-scale development of these materials for successful gas capture and storage technologies application would be vital for sustainable development. The widespread technological application would develop and strengthen new avenues towards rural industrialisation.

सारांश

जलवायु परिवर्तन की अभूतपूर्व चुनौती हमें बताती है कि पिछले दो और आधे सदी में चमत्कारी विकासों के बावजूद, पर्यावरणीय मापदंडों पर नाकामी ही हासिल की है। जीवाश्म ईंधन इतनी समय तक ऊर्जा का सुखद स्रोत रहे हैं, हालांकि, उनका उपभोग प्रमुख रूप से कार्बन उत्सर्जन के स्तरों में योगदान किया है। अब, विकल्पिक ऊर्जा स्रोतों का विकास आवश्यक है, और एनोरोबिक डाइजेशन जैसी प्रौद्योगिकी की इस ऊर्जा परिवर्तन में महत्वपूर्ण भूमिका है। नवीकरणीय मीथेन जैविक अपशिष्ट पाचन और प्राप्त बायोगैस विभाजन से हासिल किया जाता है। मुख्य चुनौती मीथेन युक्त गैस का भंडारण करना है, वर्तमान में बहुत उच्च 20-25 MPa तक दबाकर कहाँ संग्रहित किया जाता है। वैकल्पिक रूप से, अधिशोषण आधारित भंडारण में जहाँ समान मात्रा में गैस (संपीड़न के रूप में) हल्के दबाव में संग्रहीत की जा सकती है। गैस का भंडारण छिद्रपूर्ण सामग्री से भरे सिलेंडर में प्राप्त किया जाता है, जहाँ गैस को सूक्ष्म छिद्रित सतह पर दबाव में सोख लिया जाता है। गैस संग्रहण माइक्रोपोरस सामग्रियों में साधारित कमजोर दब की तुलना में अधिक है, जिससे अंत में भंडारण दबाव की आवश्यकता कम हो जाती है। बायोमास से प्राप्त किए गए माइक्रोपोरस सामग्रियों का लाभ यह है कि वे पुनर्नवीनी हैं और इस तरह की स्रोत का उपयोग करने से पर्यावरणीय बोझ भी कम होता है। बायोमास को कार्बनीकरण प्रक्रिया द्वारा पहले बायोचार में परिवर्तित किया जाता है, और फिर उसे रसायनो का उपयोग करके उच्च सतह क्षेत्र सक्रिय कार्बन्स में रूपांतरित किया जाता है। इस थीसिस में, इस प्रकार के बायोमास आधारित सामग्रियों, और उनके उच्च क्षमता गैस संग्रह और कैद के लिए उनके उत्पादन पर ध्यान केंद्रित किया है।

पहले उद्देश्य में, बायोचार को संयुक्त भौतिक और रासायनिक प्रक्रिया के संयोजन का अन्वेषण किया है। पोटैशियम हाइड्रॉक्साइड रासायनिक सक्रियकरण और भाप भौतिक सक्रियकरण के तरह उपयोग किया गया है। संयुक्त सक्रियकरण कार्बन्स का सतह क्षेत्र 963 मीटर वर्ग/ग्राम जो की, तुलनात्मक रूप से केवल रासायनिक रूप से प्राप्त पदार्थ, 764 मीटर वर्ग/ग्राम से अधिक है। बायोमीथेन अधिशोषण का विश्लेषण के लिए, बेंटोनाइट और कैओलिन का उपयोग करके पेलेट्स भी तैयार की हैं। प्राप्त किए गए सर्वश्रेष्ठ परिणाम में 20% (वजन के आधार पर) कैओलिन के साथ 0.28 ग्राम/ग्राम भार की मीथेन, अधिशोषण क्षमता 231 v/v और प्रदेय क्षमता के 177 v/v के साथ मिला है।

दूसरे कार्य में, विभिन्न प्रकृति के बायोमास का चयन किया और उनके सतही छिद्र निर्माण प्रतिरूप का अध्ययन किया गया है। यहाँ समान सक्रियकरण की शर्तों के तहत दस विभिन्न जैविक स्रोतों से सक्रिय कार्बन उत्पन्न किया है। चयन उनकी भौतिक प्रकृति और उनकी स्थानीय उपलब्धता पर निर्भर है। ये तैयार किए गए कार्बन्स को फिर विभिन्न तकनीकों जैसे कि BET, एक्स-रे डिफ्रेक्शन, स्कैनिंग इलेक्ट्रॉन माइक्रोस्कोप, और एफटी-इन्फ्रारेड के साथ विश्लेषण किया गया है। बायोमास की प्रकृति उनके छिद्र निर्माण को प्रभावित करती है, और इसलिए विभिन्न मीथेन संचयन में भिन्नता प्राप्त होती है। नारियल शैल सक्रिय कार्बन पर लगभग 251 v/v (अधिशोषण), 217 v/v (प्रदेय) का सर्वोच्च आयतन अंश, जबकि करंजा शैल सक्रिय कार्बन पर 6.5 MPa में लगभग 0.32 g/g का सर्वोच्च गैस अधिग्रहण दर्ज हुआ है।

अगले कार्य में, प्रणाली क्रियान्वयन के दौरान गैस के कम निकालने की चुनौती को संबोधित करने का प्रयास किया है। प्रौद्योगिकी की ऊष्मप्रवैगिकी नियम गैस मुक्ती को हतोत्साहित करता है। कमरे के तापमान पर गैस का अधिशोषण करके विभिन्न तापमानों 40, 45, 50 और 55 °C पर पानी के वातवरण में गैस मुक्ती प्रक्रिया का अध्ययन किया। कमरे की तापमान पर, करीब 54% गैस मुक्त हो रही है, जबकि, 55 °C पर करीब 80% गैस पुनर्प्राप्त हो रही है।

जीवन चक्र मूल्यांकन के द्वारा इस अधिशोषण आधारित प्रौद्योगिकी का निरूपण और संपीड़न आधारित भंडारण के साथ तुलनात्मक अध्ययन भी किया है। सक्रिय कार्बन आधारित भंडारण प्रणाली पर्यावरण संबंधित पैरामीटर्स पर यह प्रभावी है, हालांकि कुछ बिंदुओं का विचार किया जाना चाहिए। कम दबाव भंडारण सिलेंडर की मोटाई को कम करता है, सुरक्षा को बढ़ाता है, यह मध्यबिंदु सूचकों पर प्रतिबिम्बित भी होता है। हालांकि, सक्रिय कार्बन के रासायनिक उत्पादन अभी भी मुख्य चिंता है और उत्पादन के दौरान कचरे का पुनःप्रयोग करना पर्यावरण पर बोझ को कम करता है।

अंत में, विषम परमाणु संपन्न पदार्थ पर ध्यान केंद्रित करके सक्रिय कार्बन तैयार किया है और उनके दीर्घकालिक उत्पादन में सुधार किया। इस कार्य में, इन सक्रिय कार्बन का उपयोग बायोगैस और दहन गैसों गैस से प्राप्त कार्बन डाइऑक्साइड कैचर में किया गया है। इन सक्रिय कार्बन में पर्याप्त नाइट्रोजन और सल्फर मौजूद है, जबकि ऑक्सीजन की उपस्थिति प्रचूर मात्रा में हैं। इन्हें हाइड्रोचार मार्ग से तैयार किया जाता है, और हाइड्रोचार को सक्रिय करने के लिए $KCl+Na_2S_2O_3$ का उपयोग किया जाता है, जिससे उच्च सतही क्षेत्र और सरंध्रता प्राप्त होती है। तैयार सक्रिय कार्बन पर 3.5

mmol/g तक का उच्च CO₂ अवशोषण प्राप्त किया गया। अध्ययन में CO₂ पर विषम परमाणु के प्रभाव की भी जांच की गई। ये सामग्रियां पांच चक्रों में उच्च पुनर्योजी क्षमता भी दर्शाती हैं।

इसके अतिरिक्त, इन सामग्रियों के बड़े पैमाने पर विकास के सफल अनुप्रयोग, गैस कैप्चर और स्टोरेज प्रौद्योगिकियों के विकास के लिए महत्वपूर्ण होगा। व्यापक तकनीकी अनुप्रयोग नए ग्रामीण औद्योगिकी की दिशाओं को विकसित और मजबूत करेगा।

Table of Contents

Particulars	Page No.
Certificate	i
Acknowledgements	ii
Abstract	iv
Table of content	ix
List of figures	xii
List of tables	xv
Abbreviations	
Chapter 1: Introduction	1
1.1 General	1
1.2 Biogas upgradation or carbon capture	3
1.3 Methane-rich gas storage	11
1.4 Organization of the thesis	
Chapter 2: Literature review	13
2.1 Adsorbent Materials	13
2.1.1 Metal-organic Frameworks (MOFs)	14
2.1.2 Zeolites and other materials	14
2.2 Activated carbons	16
2.2.1 Activated carbon preparation	17
2.2.1.1 Carbonization	17
2.2.1.2 Physical activation	18
2.2.1.3 Chemical activation	18
2.2.1.4 Materials for methane adsorption	21
2.3 Methane adsorption	21
2.3.1 Surface area and porosity	24
2.3.2 Packing density and palletisation	27
2.4 Process of desorption	28
2.5 Carbon dioxide capture and separation	28
2.5.1 Hydrothermal carbonization and heteroatom doping	30
2.5.2 Parameters affecting CO ₂ uptake	31
2.5.3 Alternative chemicals for activation	32
2.6 Research gaps	33
2.7 Objectives	34
Chapter 3: Materials and Methods	35
3.1 Raw materials	35
3.2 Activated carbon production	36
3.2.1 Physicochemical activation	36
3.2.2 Characterization of activated carbons	37
3.3 Preparation of pellets and strength measurement	38
3.4 Gas uptake and release measurements	38
3.5 Temperature based desorption	40

3.6	Determination of 100% desorption with respect to adsorption	40
3.6.1	Using commercially brought ACs	41
3.7	Life Cycle Assessment	41
3.7.1	Goal and Scope	42
3.7.2	ReCiPe 2016 method	43
3.7.3	IPCC 2013 GWP 100a	45
3.7.4	Functional Units	45
3.7.5	Exclusions and Assumptions	46
3.7.6	Life cycle inventory	46
3.7.6.1	Activated carbons production	46
3.7.6.2	Transportation LCI	48
3.8	Heteroatom-rich activated carbons production	49
3.8.1	Hydrothermal Carbonization	50
3.8.2	Activation of the hydrochar	50
3.8.3	Characterization of materials	51
3.8.4	Experimental CO ₂ uptake	51
3.8.5	Life Cycle Inventory	53
Chapter 4: Activated Carbon Production, Characterization and Methane Storage		55
4.1	Physicochemical activation	55
4.1.1	Yield, ultimate and proximate analysis	56
4.1.2	Surface area and porosity	57
4.1.3	Pellets preparation, bulk density, and strength	59
4.1.4	Methane Uptake and release dynamics	61
4.1.5	Repeatability and Regeneration of Pellets	63
4.2	Porosity development study on ten biomasses	64
4.2.1	Yields, ultimate and proximate analysis	64
4.2.2	Bulk density of AC pallets	66
4.2.3	Surface characteristics	66
4.2.3.1	Adsorption isotherm	68
4.2.3.2	Pore size distribution	70
4.2.3.3	SEM study	72
4.2.3.4	X-Ray diffraction analysis	76
4.2.3.5	FTIR analysis	77
4.2.3.6	Methane adsorption-desorption studies	79
4.2.3.7	Porosity and delivery percentage	81
4.3	Conclusions	82
Chapter 5: Enhanced Methane Desorption and Complete Delivery of Gases		85
5.1	Introduction	85
5.2	Temperature based enhanced desorption	86
5.2.1	Delivery at 40 °C	86
5.2.2	Delivery at 45 °C	87
5.2.3	Delivery at 50 °C	88
5.2.4	Delivery at 55 °C	88
5.3	Achieving full desorption with respect to adsorption capacity	89
5.4	Conclusions	90

Chapter 6: Life Cycle Assessment of Activated Carbons Production and Storage Technology	91
6.1 Introduction	92
6.2 Impact assessment	92
6.2.1 Activated carbon production	
6.2.1.1 Midpoint method	92
6.2.1.2 Endpoint method	96
6.2.2 Transportation impact assessment	97
6.2.2.1 Midpoint indicators	97
6.2.2.2 Endpoint indicators	99
6.2.3 IPCC GWP 100a method	99
6.3 Conclusions	102
Chapter 7: Heteroatom-Rich Activated Carbon Production and Their CO₂ Uptake Capacities	103
7.1 Introduction	103
7.2 Results and Discussion	104
7.2.1 Hydrothermal carbonization	104
7.2.2 Activation of hydrochars	106
7.3 CO ₂ uptake analysis	111
7.4 LCA based comparison of activated mixtures	116
7.5 Conclusions	117
Chapter 8: Perspective Integration of Biochar-based Materials for Rural Areas	119
8.1 Adsorption-based system	119
8.2 Potential entrepreneurial opportunities	120
8.3 Making an economically sustainable model	122
8.3.1 Harnessing maximum efficiency	122
8.3.2 Government support and encouragement	123
8.3.3 Private sector investment	123
8.3.4 Linking with carbon credit schemes	124
8.4 Circular economic model	124
Chapter 9: Conclusions and future perspectives	125
References	129
Curriculum Vitae	143

List of Figures

S. No.	Title	Page No.
Figure 1.1	Biomass-based materials preparation and application for the carbon capture and separations.	5
Figure 1.2	Volumetric and gravimetric energy density of different fuels.	7
Figure 1.3	The overall concept of material preparations and application for the methane storage technology.	9
Figure 1.4	Scope of the work covered in this thesis	10
Figure 2.1	Schematic representation of the adsorption process; Adsorbate- Methane; Adsorbent- Activated carbons porous surface.	22
Figure 3.1	Process flow of Activated carbon production from biomass.	37
Figure 3.2	Schematic diagram of programmable tubular furnace with stainless steel tube.	37
Figure 3.3	Gas adsorption measurement setup with material-filled cylinder.	39
Figure 3.4	Process flow diagram of this life cycle study.	42
Figure 3.5	System boundary for activated carbon production and application for adsorption-based storage.	43
Figure 3.6	Procedure for the heteroatom rich doping and production of activated carbons.	50
Figure 3.7	Overview of the fluidized bed reactor test system for isothermal investigation of CO ₂ uptake.	52
Figure 4.1	(A) Nitrogen adsorption-desorption isotherm and (B) Pore size distribution (C) t-plot of prepared carbons ACC and ACPC.	59
Figure 4.2	SEM images of (A) Physicochemically activated carbons (B) Chemically activated carbons.	59
Figure 4.3	A. Volumetric uptake and deliverable capacity. B. Gravimetric uptake capacity, of different pellets, compared to the target set by Department of Energy, US and compression of the gas at 6.5 MPa.	62
Figure 4.4	Volumetric and gravimetric uptake capacities of Kaolin (20wt%) supported pellets, with variable pressure up to 6.5 MPa.	63
Figure 4.5	Repeatability study of methane uptake and release on Kaolin (20 wt%) supported pellets.	64
Figure 4.6	Adsorption Isotherms of prepared activated carbons with different values of nitrogen adsorption performed at -196 °C on vertical axis. They follow type I a & b type isotherm as per the characteristics of microporous materials.	69
Figure 4.7	Pore size Distribution determined using NLDFT method derived based on nitrogen adsorption and desorption performed at -196 °C. These graphs have been focused in the range from 0 nm to 4 nm which is crucial methane adsorption and storage.	72

Figure 4.8	Scanning electron microscopic (SEM) images at a different scale for understanding developed pore structure on the surface of bio-char.	75
Figure 4.9	X-ray diffraction pattern of all materials of activated carbons in powdered form to understand the staking of the layers.	77
Figure 4.10	Bonding pattern analysis of activated carbons by Fourier transformed infrared spectrum in merged form.	79
Figure 4.11	Volumetric and Gravimetric uptake capacities of activated carbons on prepared pellets with 20 wt% kaolin.	81
Figure 4.12	Delivery percentage of methane by various activated carbons.	82
Figure 5.1	Uptake and volumetric delivery ratio on activated carbons in the cylinder and heated discharge recorded at 40 °C from the second and subsequent cycle.	87
Figure 5.2	Uptake and volumetric delivery ratio on activated carbons in the cylinder and heated discharge recorded at 45 °C from the second and subsequent cycle.	87
Figure 5.3	Uptake and volumetric delivery ratio on activated carbons in the cylinder and heated discharge recorded at 50 °C from the second and subsequent cycle.	88
Figure 5.4	Uptake and volumetric delivery ratio on activated carbons in the cylinder and heated discharge recorded at 55 °C from the second and subsequent cycle.	88
Figure 5.5	Volumetric uptake and release capacity in each number of cycles. Conditions; A- equilibrium time: 12 hrs, B- equilibrium time: 30 min on commercial coconut shell AC.	89
Figure 6.1	Activated carbons production effects on midpoint indicators.	93
Figure 6.2	Individual parameter impacts on ReCiPe midpoint indicators for CAC production.	94
Figure 6.3	Midpoint indicators comparison with and without recycled chemical activation carbon production. Only major impact categories are presented in the figure.	95
Figure 6.4	Damage assessment on ReCiPe 2016 Endpoint indicators. Units of Human health is in loss of life in year, Ecosystem in species loss, and Resources in cost in dollars.	96
Figure 6.5	Transportation options comparison based on midpoint indicators.	98
Figure 6.6	Comparison of high score midpoint indicators with individual contribution of all the inputs in transportation while considering (a) physically and (b) chemically with recovery activated carbon-based storage.	99
Figure 6.7	ReCiPe endpoint indicators scores due to 200 km of transportation. Human health is in loss of life in year, Ecosystem in species loss, and Resources in cost in dollars	99
Figure 6.8	Individual contribution to endpoint indicators while considering PAC based storage.	100

Figure 6.9	Global warming potential posed by all the storage systems. The data has been reported by considering CAC based system as 100% GWP.	101
Figure 6.10	Individual contribution by inputs of the storage system in global warming.	101
Figure 7.1	A&B presents adsorption-desorption isotherm and pore size distribution (using NLDFT method)	109
Figure 7.2	CO ₂ adsorption capacity of high surface area activated carbons.	113
Figure 7.3	Representation of relation between CO ₂ uptake (mmol/g) and surface area (m ² /g).	114
Figure 7.4	Langmuir isotherm model for high surface area activated carbons at 50 °C.	115
Figure 7.5	Cradle-to-gate environmental impact assessment comparison of activating agents required for chemically activated carbon production on some of the significant midpoint categories, characterization method (%).	116
Figure 7.6	NFPA 704 hazard labels of chemicals used as activating agents.	117
Figure 8.1	Biomass utilization for anaerobic digestion and activated carbon production and adsorption based gas storage for vehicular application	121

List of Tables

S. No.	Caption	Page No.
Table 2.1	Recent reported carbon-based adsorbents and its capacities for methane storage	26
Table 3.1	Evaluation categories, indicators and their respective units	43
Table 3.2	Input and output parameters for chemical activation with their amount.	46
Table 3.3	Input and output parameters for physical activation with their amount based on ecoinvent library.	47
Table 3.4	Input parameters for transportation of 200 vkm.	48
Table 4.1	The yield and elemental composition of activated carbons.	57
Table 4.2	Textural properties of activated carbons prepared.	58
Table 4.3	Pellets composition and their bulk density.	60
Table 4.4	Proximate and ultimate analysis of prepared activated carbons.	65
Table 4.5	Bulk density of pellets prepared with 20 wt% of Kaolin	66
Table 4.6	Surface morphological properties of activated carbons.	67
Table 7.1	Hydrochar optimization its elemental analysis and N retention based on varied reaction parameters.	105
Table 7.2	TGA and overall yield comparison of Cellulose and Softwood supported hydrochar.	106
Table 7.3	Activated carbon production from hydrochars their yield and elemental analysis.	107
Table 7.4	Surface properties of Activated carbons.	108
Table 7.5	Morphological properties of activated carbons prepared using KCl+Na ₂ S ₂ O ₃ .	111
Table 7.6	CO ₂ uptake analysis of activated carbons.	112
Table 7.7	Renderability of the ACSWU activated carbons over five cycles.	114

Abbreviations

GHG	Green House gases
CO ₂	Carbon dioxide
CH ₄	Methane
N ₂	Nitrogen
O ₂	Oxygen
MJ	Mega Joule
Kg	Kilo Gram
km	Kilo meter
bio-CO ₂	Biogenic CO ₂
MEA	Monomethylamine
MOF	Metal-organic frameworks
CMS	Carbon molecular sieves
AC	Activated carbons
CCS	Carbon capture and storage
BECCS	Bioenergy with carbon capture and storage
L	Litre
STP	Standard temperature and pressure
NTP	Normal temperature and pressure
MPa	Mega Pascal
CNG	Compressed natural gas
LNG	Liquified natural gas
ANG	Adsorbed natural gas
K	Kelvin
v/v	volume/volume (cm ³ or L)
g/g	gram/gram
DoE	Department of Energy
atm	Atmospheric pressure
IUPAC	International union of pure and applied chemistry
SDG	Sustainable development goals
NNBOMP	New National Biogas and Organic Manure Programme
GOBAR	Galvanizing Organic Bio-Agro Resources
SATAT	Sustainable Alternative Towards Affordable Transportation
CBG	Compressed Biogas
LPG	Liquified petroleum gas
PSA	Pressure swing adsorption
BET	Brunauer–Emmett–Teller
SEM	Scanning electron microscope
FTIR	Fourier transform Infrared
XRD	X-ray diffraction
LCA	Life Cycle Assessment
MS	Molecular sieves
PAFs	Porous aromatic frameworks
NLDFT	Non-local density function theory
ISO	International standard organisation
MPSD	Micropore size distribution
MOVE	Methane opportunities for vehicular energy
wt	Weight

Å	Angstrom
KOH	Potassium Hydroxide
KCl	Potassium Chloride
HCl	Hydrogen chloride
K ₂ C ₂ O ₂	Potassium oxalate
K ₂ CO ₃	Potassium Chloride
NaOH	Sodium hydroxide
NaNH ₂	Sodium amide
H ₃ PO ₄	Phosphoric acid
ZnCl ₂	Zinc Chloride
ACC	Chemically activated carbons
ACPC	Physicochemically activated carbons
GAC	Granular activated carbons
RoW	Rest of the world
CML	Centrum voor Milieukunde Leiden
IPCC	Intergovernmental panel on climate change
CAC	Chemically activated carbon
PAC	Physically activated carbon
CACR	Chemically activated carbon with waste recovery
HTC	Hydrothermal carbonization
TS	Total solid
DC	De-oiled cake (Kanranja)
SW	Softwood
C	Cellulose
TGA	Thermogravimetric analyser
HDC	De-oiled cake hydrochar
ACDC	De-oiled cake activated carbon
K ₂ CO ₃	Potassium carbonate
K ₂ O	Potassium oxide
B	Bentonite
K	Kaolin
S	Sugar
nm	Nanometer
MPD	Mean pore diameter
CCSAC	Coconut shell activated carbons
h	hour