



Cite this: *Green Chem.*, 2024, **26**, 1790

Sustainable valorisation of food waste into engineered biochars for CO₂ capture towards a circular economy

Wenhui Jia,^{†a} Shuangjun Li,^{†b} Junyao Wang,^{†c} Jonathan T. E. Lee,^{id d}
Carol Sze Ki Lin,^{id e} Ondřej Mašek,^f Huiyan Zhang,^{id a} and Xiangzhou Yuan^{id *a}

The large amount of food waste generated worldwide has significant adverse environmental impacts to our entire ecosystem, highlighting the urgent need for a historic resolution to achieve sustainable management of food waste as well as its circular economy. In this regard, preparation of engineered biochars from food waste has garnered significant attention for CO₂ capture, as this upcycling potential could play a significant role in advancing the concept of a negative carbon circular economy. Hence, this review holistically explores the potential of food waste-derived engineered biochars as CO₂ adsorbents, not only from sample-level to process-level CO₂ adsorption, but also from a life-cycle perspective. Sample-level CO₂ adsorption is examined in terms of synthetic methods and procedures, focusing on application and optimisation of carbonisation, activation, and surface modification processes. The application of machine learning for guiding syntheses of high-performance CO₂ adsorbents derived from food waste is also discussed. Process-level CO₂ adsorption is examined in terms of two primary cycling configurations, namely pressure swing adsorption and temperature swing adsorption, whose efficiency is critical for commercialisation. In addition, a comprehensive life-cycle assessment is performed to provide a novel and timely overview of the environmental impacts of CO₂ adsorption using food waste-derived engineered biochars. This review demonstrates the viability and potential of integrating food waste-derived engineered biochars with carbon capture technologies to afford an environmentally friendly innovation for sustainable food waste management and climate change mitigation, which is beneficial to achieving UN Sustainable Development Goals including Goals 11–13.

Received 27th October 2023,
Accepted 29th December 2023

DOI: 10.1039/d3gc04138g

rsc.li/greenchem

Introduction

Approximately one-third of total global food resources, equivalent to 1.3 billion tons annually, is wasted in food production and consumption, resulting in a broad range of adverse environmental impacts.^{1,2} Greenhouse gas (GHG) emissions are one of the most serious environmental impacts,³ with 8–10% of

global GHG emissions attributable to unsustainable management of food waste.⁴ For example, the U.S. Environmental Protection Agency (EPA) highlighted that due to mismanagement of food waste, 170 million metric tons of carbon dioxide (CO₂) equivalent GHGs are emitted annually, an amount that is equal to the annual CO₂ emissions of 42 coal-fired power plants.⁵ Moreover, food waste generation has been increasing globally and it is now included in three of the United Nations' 17 Sustainable Development Goals (UN SDGs), namely goal 11: sustainable cities and communities, goal 12: sustainable consumption and production patterns, and goal 13: climate action.⁶ Food waste, as a typical type of biomass but commonly mismanaged, leads to serious health and environmental issues. It is closely associated with human activities and is the most easily generated, collected, and accumulated in urban environments. It is worth noting that food waste is seasonal and regional properties, which is one key difference from general biomass, *i.e.*, wood biomass and rice husk. Therefore, to attain global environmental sustainability, there is a pressing need to transition from the prevailing linear paradigm of food consumption and production to a circular paradigm,

^aMinistry of Education of Key Laboratory of Energy Thermal Conversion and Control, School of Energy and Environment, Southeast University, Nanjing 210096, China. E-mail: yuanxz@seu.edu.cn

^bDepartment of Chemical & Biological Engineering, Korea University, 145 Anam-ro, Seongbuk-gu, Seoul 02841, Korea

^cSchool of Materials and Energy, Guangdong University of Technology, Guangzhou 510006, China

^dEnvironmental Research Institute, National University of Singapore, Singapore 138602, Singapore

^eSchool of Energy and Environment, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong, China

^fUK Biochar Research Centre, School of GeoSciences, University of Edinburgh, Crew Building, Alexander Crum Brown Road, Edinburgh EH9 3FF, UK

[†]These authors contributed equally as first authors.

where unavoidable residues from the farming and food industry are valorised.

Sustainable valorisation of food waste at a global scale is essential for reducing environmental and economic burdens of food waste and thereby achieving a circular economy.^{7,8} Biochemical approaches (*i.e.*, anaerobic digestion, fermentation, and composting) and thermochemical approaches (*i.e.*, hydrothermal carbonisation (HTC), pyrolysis/co-pyrolysis, and gasification) are widely used to upcycle food waste into value-added bioenergy and solid carbonaceous materials,² and additional pretreatments for size reduction are commonly needed to improve conversion efficiency.⁷ However, there are three major challenges associated with biochemical approaches, namely that (1) they are inherently slow (*i.e.*, can take up to several months) and typically require large reactors, and their conversion efficiency is low owing to the diversity and complexity of food waste; (2) they rely on microorganisms, which are susceptible to variations in operating conditions, such that precise control is required to optimise product yields, energy exergy, and economic feasibility, and (3) they require additional rebuilding treatments to transform main products with low-molecular weight into high-molecular weight variants, resulting in energy-inefficient conversion.²

The aforementioned challenges have driven investigations into thermochemical approaches, which offer fast reaction rates, mild operating conditions, controllable product compositions, and high feasibility for commercial applications.⁹ Gasification is a thermochemical approach that efficiently converts solid carbonaceous materials into syngas *i.e.*, carbon monoxide (CO) and hydrogen (H₂) in a gasifier under high-temperature and low-oxygen conditions.^{10,11} Owing to the high volatile matter content (<80%) of food waste, it is regarded as a promising alternative for energy production, as it solidifies energy security *via* a waste-to-energy strategy.^{9,12} Pyrolysis of food waste occurs at relatively low temperatures (300–600 °C) in an oxygen-free atmosphere and results in upcycling of food waste into highly heterogeneous solid, liquid, and gaseous products.¹³ Pyrolysis is classified as either slow pyrolysis (which mainly generates biochar) or fast (flash) pyrolysis (which mainly generates bio-oil) and has wide applications in research and industry. HTC is widely considered to be a practical and suitable approach for valorising high-moisture content food waste to produce value-added carbonaceous materials that are termed 'hydrochar'.¹⁰ However, it remains difficult to upcycle food waste into biofuels in a profitable manner with zero carbon emissions. This has led to numerous research on valorisation of food waste into high-performance carbonaceous materials for environmental protections and various energy conversion and storage applications.^{10,14}

Current research on organic waste-derived carbonaceous materials for energy and environmental applications have been reviewed.^{10,26,27} These reviews have highlighted that conversion of organic waste into carbonaceous materials for CO₂ capture is sustainable and practical, especially in the context of carbon neutrality.^{28,29} Moreover, food waste-derived high-performance engineered biochars have been researched as materials for CO₂

capture (Table 1), which is a feasible approach for simultaneously mitigating climate change and achieving sustainable waste management.^{30,31} Therefore, there is an urgent need for a comprehensive review of current research on food waste-derived CO₂ adsorbents to cover recent advances, existing challenges, and future perspectives. As summarized in Fig. 1, this review (1) examines the synthesis routes from food waste to high-performance CO₂ adsorbents (including machine learning-aided optimization of these routes), (2) evaluates capture performance of CO₂ adsorbents from both sample- and process-level perspectives, and (3) assesses their environmental impacts from a life-cycle perspective. Thus, this review reveals key aspects of design and optimization of food waste-derived CO₂ adsorbents and provides guidelines for large-scale CO₂ capture deployment. It is hoped that this will assist researcher from academic and industrial communities and/or even policymakers from governmental agencies, who are striving to achieve goals such as carbon neutrality, sustainable waste management, circular carbon economy, and even UN SDGs.

Advances in food waste-derived CO₂ adsorbents

Conventional synthetic procedures

Carbonisation and activation have been widely used to synthesise high-performance CO₂ adsorbents from food waste.^{30–32} Slow pyrolysis has been considered one of the most promising and practical routes for carbonization of biomass and its waste to produce biochars.^{10,33} However, direct pyrolysis is not practically applicable to food waste because its high moisture content requires an energy-intensive pre-drying process for upcycling food waste into biochars (as shown in Fig. 1). Therefore, HTC, which is a thermochemical conversion performed in the presence of water at 180–265 °C and 2–6 MPa for 5–240 min,¹⁰ has attracted much attention to produce food waste-derived hydrochar,^{34,35} due to that HTC does not require pre-drying treatment of food waste and requires the addition of little or no water. It suggests that HTC reduces both energy and water consumptions, simultaneously.

Biochar and hydrochar have poor textural properties, and thus have been subjected to physical or chemical activation to enlarge their surface areas and generate microporous structures, which facilitate high-performance CO₂ adsorption.^{30,31} Physical activation is typically performed using CO₂, steam, and air as activation agents, and increases the porosity and functional groups of biochars or/hydrochars in an eco-friendly and cost-effective manner. CO₂ is the most widely used in physical activation, due to its relatively green characteristics and low reactivity at high activation temperatures (*i.e.*, >700 °C).³⁶ Moreover, the off-gas produced by physical activation with CO₂ can be reused as an activation agent after simple combustion,³⁷ minimizing additional CO₂ emissions and achieving a closed carbon loop. Compared with physical activation of food waste, chemical activation is typically conducted at lower temperatures and with shorter residence time,

Table 1 Research and development on food waste-derived engineered biochars for CO₂ capture

Feedstock	Synthesis approach		Textural properties			CO ₂ adsorption capacity		CO ₂ /N ₂ selectivity		Ref.
	Carbonisation	Activation	Surface functionalisation	Specific surface area (m ² g ⁻¹)	Total pore volume (cm ³ g ⁻¹)	N content (%)	25 °C & 1 bar	0 °C & 0.1 bar	ISAT method	Henry's law
Crab shells	Pyrolysis at 400 °C for 1.5 h	KOH activation at 600 °C for 1.5 h (2)	—	1004	0.46	6.2	4.37	—	19 (15/85)	—
Fish scales	Pyrolysis at 330 °C for 3 h	KOH activation at 600 °C for 1 h (1)	—	1479	0.88	—	2.80	—	—	16
Rotten strawberries	Hydrothermal carbonization at 180 °C for 12 h	KOH activation at 600 °C (2)	—	935	0.42	3.68	3.63	5.27	16 (10/90)	—
Shrimp shells	Pyrolysis at 400 °C for 1.5 h	KOH activation at 700 °C for 1 h (1)	—	1406	0.72	2.16	4.20	6.69	23 (15/85)	—
Coconut shells	Pyrolysis at 500 °C for 2 h	K ₂ CO ₃ activation at 600 °C for 1 h (3)	Urea modification at 350 °C for 2 h in air	1082	0.39	2.74	3.71	5.12	13 (10/90)	19
Pineapple waste	Hydrothermal carbonisation at 210 °C for 10 h with alkali metal oxalate	K ₂ C ₂ O ₄ activation at 700 °C for 2 h	—	1076	0.08	0.33	4.25	5.32	27 (10/90)	20
Garlic peels	Hydrothermal carbonisation at 220 °C for 24 h followed by pyrolysis at 400 °C for 2 h	KOH activation at 600 °C for 1 h (2)	—	947	0.51	—	4.22	6.25	—	21
Sugarcane bagasse	Pyrolysis at 600 °C for 0.5 h	KOH activation at 600 °C for 1 h (2)	Urea solution	1113	0.57	1.98	4.80	—	—	22
Coffee grounds	—	K ₂ CO ₃ activation at 700 °C for 5 h (1)	—	1476	0.61	1.67	4.54	7.02	16 (20/80)	23
		K ₂ CO ₃ activation at 800 °C for 1 h (1)		1692	0.71	1.51	4.46	7.18	14 (20/80)	
Food waste and wood waste	10 kW fixed-bed downdraft gasifier	KOH activation at 850 °C for 2 h (1)	—	841	0.85	—	3.23	—	—	24
Mixed food waste	pyrolysis at 400 °C for 1 h	KOH activation at 600 °C for 1 h (2)	—	807	0.41	1.6	2.54	—	—	25

Notes. KOH = potassium hydroxide; K₂CO₃ = potassium carbonate; K₂C₂O₄ = potassium oxalate; ISAT = initial surface absorption test; CO₂ = carbon dioxide; N₂ = nitrogen.

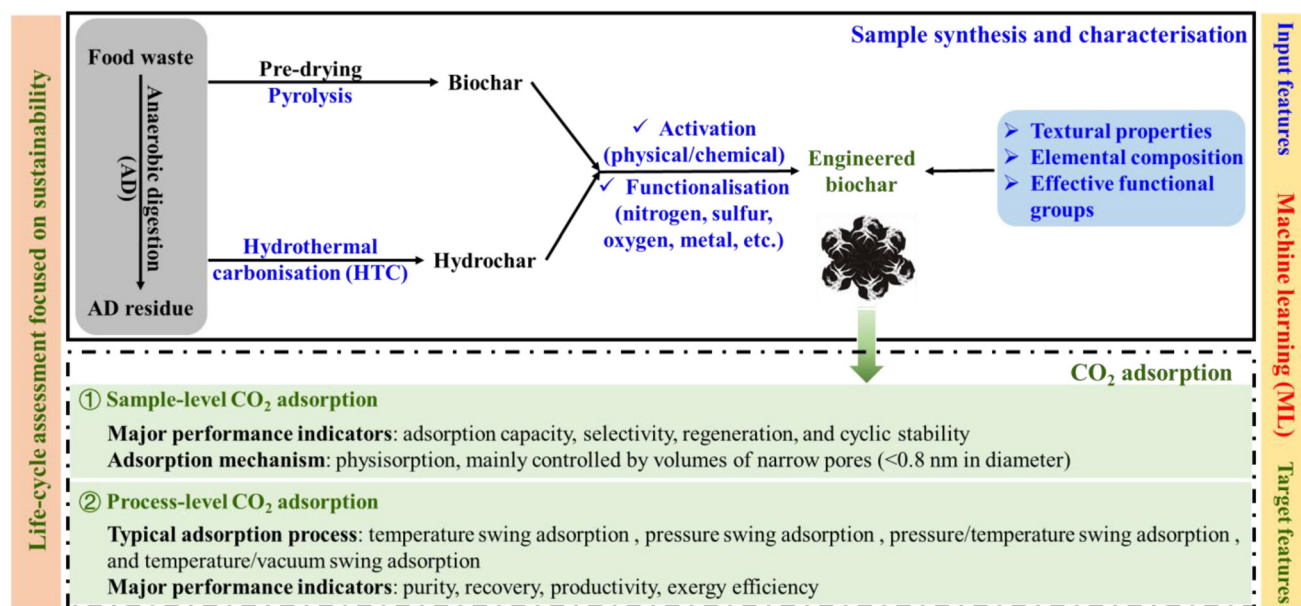


Fig. 1 Sustainable valorisation of food waste into engineered biochars for carbon dioxide (CO₂) capture, as part of a circular economy.

and produces engineered biochar with more well-developed pore structures and high yields of final products. Potassium hydroxide (KOH), phosphoric acid (H₃PO₄), and zinc chloride (ZnCl₂) are the chemical agents that are most commonly used for chemical activation.³¹ For example, Ma *et al.*³⁸ treated hazelnut shells with KOH at 650 °C for 1 h to form an engineered biochar with a surface area of 2134 m² g⁻¹ and total pore volume of 0.96 cm³ g⁻¹. However, the aforementioned chemical agents are toxic, hazardous, and corrosive, and thus damage equipment and can pollute the environment if not subjected to secondary treatments. This indicates that additional investigations on environmental impacts of chemical activation is warranted. In addition, from a life-cycle perspective, the use of chemical agents significantly contributes to environmental impacts, and the trade-off between performance enhancement and synthesis methods needs to be well considered for practical applications.

Another effective process to increase adsorption performance of CO₂ adsorbents, especially in terms of their adsorption capacity for CO₂ and selectivity for CO₂ over other gases, is functionalisation, which generates active sites for efficient adsorption of CO₂ molecules.^{39–41} Nitrogen (N), sulfur (S), and metal oxides (*e.g.*, MgO, CuO), are the main sources of heteroatoms that are added to CO₂ adsorbents to increase the number and basicity of active sites on their surfaces. Compared with single-doping treatment, dual-doping and triple-doping treatment provides more active sites on CO₂ adsorbents, resulting in them exhibiting excellent CO₂ adsorption.⁴² Furthermore, several typical biomass waste-derived engineered biochars have a high content of functional groups as they are naturally rich in heteroatoms. This means that the prepared biochar could have naturally added heteroatoms, thereby exhibiting the characteristic of enhanced adsorption

performance as well. For example, shrimp shell-derived engineered biochar contains 2.86 wt% N,⁴³ and willow catkin-derived engineered biochar contains 2.56 wt% S and 4.62 wt% N.⁴⁴ Jellyfish-based engineered biochar was proved rich in heteroatoms (Na, P, N, Ca and Mg), the functional groups existed were confirmed advantageous for CO₂ capture.⁴⁵ And nitrogen-rich seaweed-based engineered biochar has demonstrated excellent CO₂ adsorption capacity, and its mechanism has been studied using density functional theory. This suggests that surface functionalisation is a useful and practical additional treatment for increasing the CO₂ capture performance of a food-waste-derived engineered biochar that contains insufficient functional groups on its surface.

Conventional synthetic approaches

Optimisation of synthesis conditions (in terms of operating temperature, residence time, and heating rate) for carbonisation and activation (or surface functionalisation) of food waste is critical for producing engineered biochar with excellent CO₂ capture performance. Current synthesis approaches are summarized as (1) intuition-based approaches, (2) one-factor-at-a-time approaches, and (3) design of experiment *via* response surface methodology (RSM).^{46,47} The first two approaches are frequently integrated to develop engineered biochar for CO₂ capture, but (3) is widely considered as a more efficient way to synthesise engineered biochar samples for CO₂ capture. However, (1), (2), and (3) are all time- and labour-intensive, and it is difficult to determine the underlying relationships between synthetic conditions and CO₂ capture performance of the resulting engineered biochar materials. Thus, it remains highly challenging to directly optimize the production of engineered biochars such that they exhibit high-performance CO₂ capture.

Machine learning-aided synthesis approaches

Machine learning (ML) is one of the most widespread data-driven approaches in many fields. For example, ML approaches have been applied in many studies on material discovery, process optimization, and environmental protection^{48–50} as they provide valuable insights that aid the understanding, interpretation, and inverse design of complicated systems. Moreover, as shown in Fig. 2, recent studies have verified that ML is an efficient and practical tool for developing engineered biochars for CO₂ capture.^{50–52} Data collection, formatting, and pre-processing are the essential steps in the application of ML for predicting CO₂ capture performance of engineered biochars. Collected and formatted dataset are invariably lacking some data, meaning that data imputation and/or discarding is a critical step that must be conducted prior to ML investigations.^{50,52} For example, Yuan *et al.* collected 632 data-points from 76 peer-reviewed publications and ultimately used 527 of them without missing data values for predicting the CO₂ adsorption capacities of biomass waste-derived porous carbon materials.⁵⁰ In addition, tree-based ML algorithms (*i.e.*, random, forest, decision trees, gradient boosting decision trees, and light gradient boosting tree) are widely used to develop prediction models for engineered biochar-based CO₂ capture. First, pre-processed data are randomly divided into a training dataset (70%–90%) and a test dataset (30%–10%), which are then used for training and testing ML models. The current major findings are that (1) different models, such as random forest and gradient boosting decision trees, exhibit high accuracy and predictive performance, and (2) textural properties play a more critical role than chemical composition in engineered biochar-based CO₂ adsorption. It is the basic and plain application of ML algorithms to develop predictive ML models for CO₂ capture performance, the more interesting and critical thing is to apply ML modeling approaches to design specific CO₂ adsorbents and adsorption parameters with the aim of practical CO₂ capture applications.⁵³ However, it remains challenging to directly design engineered biochars

with high CO₂ capture performance. This is because recent research has focused on forward prediction of CO₂ adsorption performance but not backward design of high-performance engineered biochar-based CO₂ adsorbents.

Cyclic performance evaluation for post-combustion CO₂ capture

Sample-level characterisations of the aforementioned textural properties and gas adsorption performances of food waste-derived engineered biochars suggest that they are effective CO₂ adsorbents.³¹ However, process-level characterization *via* experimental and/or simulation approaches are needed to determine the practicality of food waste-derived CO₂ adsorbents. For example, cyclic performances of food waste-derived engineered biochars-based post-combustion CO₂ capture must be quantitatively assessed. Shen⁵⁴ and Yuan *et al.*^{30,31} have reported that the CO₂ adsorption performance of an engineered biochar is primarily affected by its characteristics (*i.e.*, textural properties and surface functionality) and the adsorption conditions (*i.e.*, temperature, pressure, humidity, and CO₂ concentration). In addition, Bernardo *et al.*⁵⁵ highlighted the cyclic performances and dynamic behaviours of newly developed CO₂ adsorbents as emerging research trends.

Research on cyclic performance has been extensively explored in recent studies, various factor-driven processes were discussed separately. Álvarez-Gutiérrez *et al.*⁵⁶ valorised cherry stones into engineered biochars to separate CO₂ and methane (CH₄) gas mixtures *via* a pressure swing adsorption (PSA) process, thereby affording fuel-grade methane. They used three cyclic performance indicators, CO₂ working capacity, CH₄ productivity, and CH₄ purity as the target parameters, and found that an adsorption pressure of 5 bar was effective for PSA using cherry stone-derived adsorbents. Surra *et al.*⁵⁷ developed a maize cob waste-derived engineered biochar that exhibited great potentials for CO₂ separation and biogas upgrading. In addition, Liang *et al.*⁵⁸ noted that their prepared

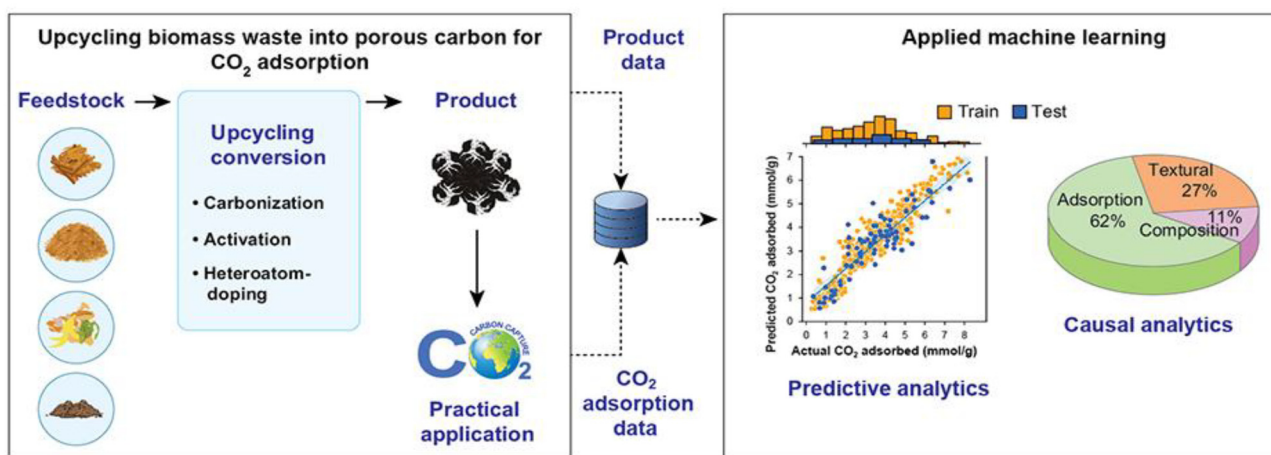


Fig. 2 Application of machine learning (ML) for upcycling food waste into engineered biochars for carbon dioxide (CO₂) capture.

popcorn-derived porous carbon reached a CO_2 adsorption capacity of 4.60 mmol g^{-1} at 1066 mbar and 25°C . Du *et al.*⁵⁹ prepared engineered biochar with high CO_2 adsorption properties using cauliflower and demonstrated 3.1 mmol g^{-1} CO_2 adsorption capacity at 1 bar and 25°C . The breakthrough curves and adsorption isotherms of cherry stone-derived engineered biochar filled gaps between the sample- and process-level investigations, providing critical knowledge to design, summarize, and validate the PSA process.^{56,57} The addition of a vacuum step into the PSA process creates the vacuum-pressure swing adsorption (VPSA) process, which uses food waste-derived engineered biochars as CO_2 adsorbents. Majchrzak-Kucęba *et al.*⁶⁰ upcycled coconut shells into an engineered biochar for a bench-scale VPSA process, verifying as a practical and feasible way for adsorbing CO_2 emitted from cement plants. They also reported that CO_2 purity increased but CO_2 recovery decreased as the flow rate of feeding gas and working cycle time increased. In addition, Majchrzak-Kucęba *et al.*⁶¹ developed the dual-reflux vacuum-pressure adsorption (DR-VPSA) process, aiming to improve both CO_2 recovery and purity. In a pilot-scale application, they divided a single adsorber into a two-stage reactor that completely separated CO_2 from inlet gas using various engineered biochar-based CO_2 adsorbents. In addition, they determined that sample-level characterisations, such as isotherm and working capacity, selectivity, renderability, and cyclic stability, are essential for accelerating process-level investigations.

Bahamon *et al.*⁶² applied the grand canonical Monte Carlo (GCMC) simulation method to obtain pure and multi-component gas adsorption data, and then used working capacity, purity, and energy consumption as major indicators to evaluate their performance of various CO_2 adsorption processes (*i.e.*, PSA, temperature swing adsorption (TSA), and vacuum swing adsorption (VSA)) using date seed-derived engineered biochars. Bahamon *et al.*⁶² concluded that two of the major indicators, *i.e.* working capacity and purity, were not significantly affected by pre-treatment of pre-adsorbed water. Food waste-derived engineered biochars have been widely investigated in laboratory-scale CO_2 adsorption studies. These engineered biochars have exhibited favourable textural properties and demonstrated excellent equilibrium and dynamic adsorption performance, as discussed in section 2. However, it remains challenging to directly use food waste-derived engineered biochar in commercial-scale CO_2 capture applications. Therefore, to facilitate practical applications, process design and optimization for CO_2 adsorption is urgently needed to determine optimal cycle configuration and operation conditions while using food waste-derived engineered biochars as CO_2 adsorbents. Fig. 3 depicts the most commonly used cycle configuration. To the best of our knowledge, few investigations of biochar have been performed to examine process-level CO_2 adsorption,⁶² and these provided useful guidelines and valuable insights on food waste-derived engineered biochars for CO_2 adsorption. In addition, this review examines several process configurations to show the CO_2 capture applications of food waste-derived engineered biochars. To bridge the gaps

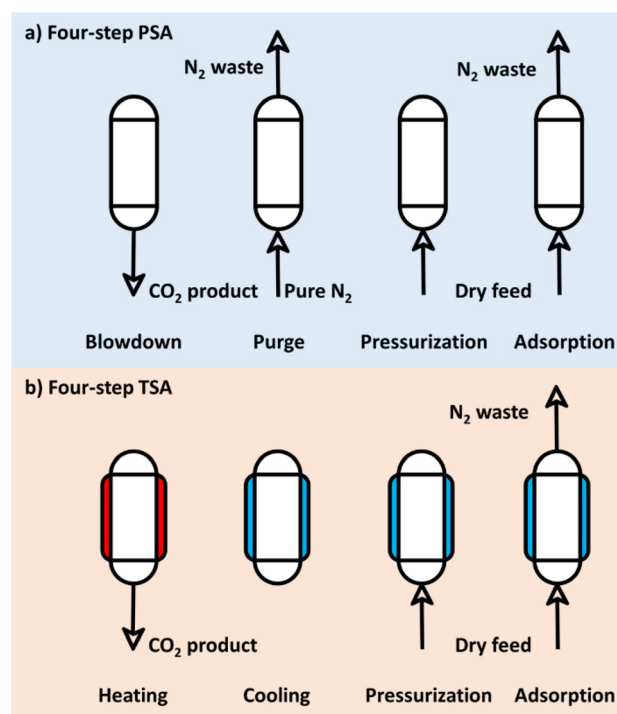


Fig. 3 Typical pressure and temperature difference driven carbon dioxide (CO_2) adsorption cycle configurations (PSA = pressure swing adsorption; TSA = temperature swing adsorption; N_2 = nitrogen).

between sample synthesis and practical CO_2 capture, there is a need for focused collection of essential data on the sample-level CO_2 adsorption performance of food waste-derived engineered biochars to facilitate the evaluation and optimization of the process-level CO_2 adsorption performance of such engineered biochars.⁶³

Pressure-driven CO_2 adsorption

The PSA process configuration was pioneered by Skarstrom in 1960. It exploits a pressure difference to achieve CO_2 separation; that is, a high-pressure condition is used to adsorb CO_2 gas, and then a low-pressure condition is used to desorb CO_2 gas.⁶⁴ Dual-step PSA is the basic PSA process, whereas four-step PSA is the most commonly used PSA process, with the four-step being adsorption, blow-down, purging, and pressurization. A high working capacity of the CO_2 adsorbent is obtained *via* the pressurization step, and extra energy is consumed by compression and vacuum generation.⁶⁵ Commercial deployment of the four-step PSA process employs physical adsorbents (especially engineered biochars) in packed-bed reactors, owing to their ease of handling, high stability and safety, low energy consumptions, low capital investment cost, and high deployment feasibility.⁶⁶

Pressure-driven CO_2 adsorption using engineered biochars as adsorbents is summarized in Table 2. For example, Drage *et al.*⁶⁷ investigated the necessity of adsorbent pretreatment for achieving high CO_2 uptake. Specifically, they examined the CO_2 working capacity of engineered biochars at atmospheric

Table 2 Summary of research on pressure-driven processes using engineered biochars as CO₂ adsorbents

Mixture treated	Process configuration	Research method	Pressure range (MPa)	Sample-level characterisation	Process-level evaluation	Results and discussion	Ref.
CO ₂ /N ₂	PSA	Simulation	0.1–4.1	CO ₂ and N ₂ isotherms measurement	CO ₂ adsorption capacity	Cyclic adsorption capacity depends on adsorption capacity and isotherm shape.	67
CO ₂ /N ₂	Four-step PSA	Experiment and simulation	0.6, 1.2, 2.0	Single and binary isotherms measurement	CO ₂ purity, recovery, and productivity	Commercial activated carbon showed high CO ₂ adsorption capacity. The mathematical model devised to describe the PSA process validated by experimental data.	68
CO ₂ /CH ₄	Four-step PSA	Experiment	0.3, 0.5, 1.0	CO ₂ and CH ₄ isotherms, heat capacity measurement	CO ₂ purity, recovery, and productivity	3D-printed adsorbent monoliths displayed PSA performance competitive with that of conventionally manufactured structured adsorbents.	70
CO ₂ /CH ₄	Two-step PSA	Simulation	0.1–0.5	CO ₂ and CH ₄ isotherms, and binary gas breakthrough curves	CO ₂ purity, recovery	Devised short-cut model that simply, but accurately clarifies the physical phenomenon in the PSA process, and screened performance of CO ₂ adsorption by engineered biochars	71
CO ₂ /N ₂ /O ₂	Five-step VSA	Experiment	—	Binary and ternary isotherms, and breakthrough curves	CO ₂ purity, recovery	Vacuum level found to play an important role in the cyclic performance.	73
CO ₂ /N ₂	Four-step PSA	Simulation	Not mentioned	CO ₂ and N ₂ isotherms	CO ₂ purity, recovery, productivity, energy consumption	It including process optimisation tools early in the adsorbent development workflow can be beneficial	75
CO ₂ /N ₂	Six-step VSA	Simulation	Low to 0.005	CO ₂ and N ₂ isotherms, uptake rates	Measurement of CO ₂ purity, recovery, productivity, and energy consumption	Detailed process modelling, simulation and optimisation strategies are the most reliable way to qualitatively and quantitatively evaluate potential adsorbents for CO ₂ capture.	76
CO ₂ /N ₂	VP SA	Experiment	Low to 0.030	CO ₂ and N ₂ isotherms measurement	CO ₂ working capacity	Porous structure and unique surface properties of biochar make it an effective CO ₂ adsorbent, including in the VPSA process.	77
H ₂ /CO ₂ /CH ₄ /CO/N ₂	10-step VPSA	Simulation	0.05–0.50	Adsorption isotherms and kinetic measurement	H ₂ purity and recovery	Multi-objective optimization methods can be used to design the VPSA process using engineered biochars as adsorbents.	78
CO ₂ /N ₂	Five-step VPSA	Simulation	0.03–0.40	CO ₂ and N ₂ adsorption isotherms measurement	CO ₂ working capacity, CO ₂ recovery, actual work consumption, second-law efficiency	Effect of adsorbent isotherm type on energy efficiency of the VPSA process clarified.	79

Notes. PSA = pressure swing adsorption; VSA = vacuum swing adsorption; VPSA = vacuum pressure swing adsorption; CO = carbon monoxide; CO₂ = carbon dioxide; H₂ = hydrogen; N₂ = nitrogen; CH₄ = methane; O₂ = oxygen; 3D = three-dimensional.

and elevated pressures (up to 4.1 MPa), using a single PSA process as the cycle configuration. Their major findings highlighted that cyclic adsorption capacity was affected by adsorption capacity and isotherm shape. Siqueira *et al.*⁶⁸ devised a mathematical model to describe the PSA process and then validated this model with a pilot-scale PSA experimental data using commercial activated carbon in a packed-bed reactor. Only tests of equilibrium CO₂ uptake measurements and adsorption kinetics were required for determining process-

level investigations, and the major indicators for evaluating cyclic performance, namely product purity, recovery, and productivity, were obtained after performing a process-level numerical simulation. Decreases in pressure in the packed-bed reactor in the PSA process were considered; these occur as the pressure-driven process is negatively affected by the unreasonable flow resistance. In addition, the application of adsorbent monoliths was found to increase mass transfer rates and reduce pressure decreases in the packed-bed reactor.⁶⁹ A three-

dimensional (3D)-printed activated carbon monolith was used for CO₂ adsorption,⁷⁰ thereby demonstrating a novel synthetic route for enhancing cyclic gas separation performance.

However, the above-mentioned mathematical models are too complex to be used for describing the physical processes occurring in packed-bed reactors, as it requires time-consuming and labour-intensive simulations, especially when comparing cyclic performances of PSA processes using various carbonaceous materials as CO₂ adsorbents. Fortunately, Álvarez-Gutiérrez *et al.*⁷¹ presented a straightforward model of engineered biochar-based PSA processes. They formed two groups to classify four-step PSA processes: one group is comprising the pressurisation and adsorption steps, and the other group is comprising the depressurisation and purging steps. Next, with reference to a series of basic assumptions, they determined the mass, momentum, and energy conservation in the packed-bed reactor. In addition, only adsorption isotherms of pure gas component were required to characterize the sample-level process in this simplified model. However, some other process-level indicators, including bed density and porosity, are required for studying PSA processes. A pressure-driven cycle configuration that employs a step involving adsorbent regeneration under vacuum conditions is referred to as a VSA process. A VSA process is simple because CO₂ desorption is readily conducted under vacuum, whereas CO₂ adsorption occurs at atmospheric pressure.⁷² A VSA process can also be used to treat flue gas emitted from power plants, even though its pressure is slightly above atmospheric pressure and adsorbent regeneration occurs under low-pressure conditions. The level of vacuum required in the regeneration step is affected by the rectangular shape of CO₂ adsorption isotherms, and engineered biochars exhibited better CO₂ recoverability under a moderate vacuum level.⁷³ Zhang and Webley⁷² and Haghpanah *et al.*⁷⁴ both determined that a VSA process results in a better CO₂ separation process than a PSA process. They devised a simplified model for rapid process simulation to evaluate the cyclic performance of solid CO₂ adsorbents.⁷⁵ They obtained basic measurements of pure-component isotherms of CO₂ adsorbents, which was necessary for the process simulation. Sample-level measurements revealed the adsorbent metrics, namely CO₂ adsorption capacity, gas selectivity, and gas working capacity (all of which can be obtained from the adsorption isotherms). The performance indicators, namely purity, recovery, energy consumption, and productivity, were considered as optimisation objectives for cyclic process-level CO₂ adsorption. The adsorbent metrics were used to screen promising and practical adsorbents for use in designing process-level CO₂ capture using food waste-derived engineered biochars. Moreover, Nikolaidis *et al.*⁷⁶ established mathematical models to describe the physical phenomena occurring in the PSA/VSA process, and concluded that product purity, recovery, productivity, and energy consumption were the optimisation objectives for process-level CO₂ capture. Furthermore, they designed a dual-bed VSA cycle consisting of six steps, including a counter-current pressurisation with the light product, adsorption, pressure equalization,

blowdown, evacuation, and counter-current re-pressurisation, for investigating CO₂ separation performance. They found that this novel cyclic process is potentially feasible for engineered biochar-based CO₂ adsorption.

As mentioned above, the PSA process adsorbs CO₂ at super-ambient pressure and desorbs CO₂ at near-ambient pressure, whereas the VSA process adsorbs CO₂ at nearly atmospheric pressure and desorbs CO₂ under a vacuum conditions. A combination of the PSA and VSA processes, denoted as the VPSA process, has attracted much attention; its adsorption step is performed at above atmospheric pressure, and its desorption step is performed under vacuum. Izabela and Marcelina⁷⁷ reported that engineered biochar-based adsorbents, even with poor CO₂ uptake in sample-level CO₂ adsorption, achieved high CO₂ working capacities in a VPSA process. They also tested biomass-based carbonaceous materials in this VPSA process, highlighting their potential as practical CO₂ adsorbents for commercial CO₂ capture. Crucially, only CO₂ uptake obtained from sample-level CO₂ adsorption is necessary to enable preliminary design of a VPSA process. Moreover, more complex VPSA configurations have been devised to solve specific problems. For example, Xiao *et al.*⁷⁸ developed a 10-step VPSA cycle for H₂ purification, which contains several additional steps of pressure equalisation compared with the basic VPSA. They employed commercial engineered biochars as CO₂ adsorbents for evaluating H₂ separation performance, and also they compared the performance of various engineered biochars in different process configurations (*i.e.*, PSA and VPSA). In addition, they used sample-level experimental data to determine multi-component gas adsorption and CO₂ adsorption kinetics to establish heat and mass transfer models, and then identified engineered biochars' solid thermal conductivities and heat transfer coefficients. Zhou *et al.*⁸⁰ developed a new VPSA cyclic process for nitrogen (N₂)/CH₄/CO₂ mixture separation using engineered biochars as CO₂ adsorbents in a packed-bed reactor. They showed that engineered biochars are viable as gas adsorbents in a VPSA process, demonstrating good potential for further development.

As vacuum regeneration is a time-saving process, VPSA processes are especially suitable for industrial-scale CO₂ capture applications. A high CO₂ working capacity can be obtained in the CO₂ adsorption (*i.e.*, physisorption) step at higher than atmospheric pressures, and the vacuum-level desorption step is suitable for the regeneration of engineered biochars. However, the compressor and vacuum pump used for CO₂ adsorption and desorption are energy-intensive equipments, suggesting that energy consumption must be reduced or energy efficiency improved in VPSA processes. Zhao *et al.*⁷⁹ designed a research framework to evaluate the energy efficiency of a VPSA process for CO₂ capture. They obtained the isotherm parameters and physical properties of CO₂ adsorbents (including commercial engineered biochars) and applied these in a process-level CO₂ adsorption simulation. They employed energy efficiency indicators, namely minimum separation work, consumption of actual work, and second-law efficiency, as cyclic performance indicators together with other

classic indicators, *e.g.*, product purity, recovery, and productivity. They concluded that the second-law efficiency of the VPSA cycle was lower than 10%, indicating that it had large energy-saving potential and isotherm shape profoundly affected energy efficiency indicators. For example, they found that the proportionality factor of CO₂ working capacity – a parameter they devised that represents the steepness of an isotherm – needed to be low to obtain high second-law efficiency. This may indicate that this is an important parameter for consideration when designing high-performance food waste derived-engineered biochars for CO₂ capture.

Temperature-driven CO₂ adsorption

A short regeneration time is essential for pressure-driven processes and facilitates industrial-scale CO₂ adsorption applications. However, high-grade energy (*i.e.*, electrical energy) is required to drive both a compressor and a vacuum pump, hence it remains challenging to achieve energy-saving targets for these pressure-driven processes *via* re-design and optimisation. Therefore, temperature-driven CO₂ adsorption has attracted much attention, given the temperature-dependence of the working capacities of engineered biochar-based CO₂ adsorbents. Compared with PSA processes, TSA process consume much less energy^{81,82} and have lower CO₂ emissions when using renewable energy (*i.e.*, low-grade thermal energy). Typical investigations of temperature-driven CO₂ capture process are summarized in Table 3, and the data needed from sample-level CO₂ adsorption processes for evaluating process-level CO₂ capture performance are also indicated.

Zhao *et al.*⁸³ investigated the basic four-step TSA process consisting of pressurization, adsorption, heating, and cooling steps. They established a shortcut model to describe the physical process occurring in a packed-bed reactor and simulated in detail the steps of the TSA process. Several typical CO₂ adsorbents (*i.e.*, commercial activated carbon adsorbents) were compared, with their CO₂ and N₂ isotherms and physical properties (*i.e.*, specific heat capacity, density, and isosteric heat of adsorption) regarded as the input features (the same scenario evidently applies to biochar as well). The CO₂ selectivity, recovery, purity, productivity, and other energy-efficiency indicators (*i.e.*, specific thermal energy consumption, minimum separation work, and the second-law efficiency) were calculated as the target features. Following this research, Jiang *et al.*⁸⁴ further developed a four-step TSA process equipped with a heat recovery step, and calculated its energy-efficiency indicators (*i.e.*, minimum separation work, and exergy efficiency), which they compared in detailed with those of the basic four-step TSA process. They concluded that the heat recovery step was an effective and practical route for heat re-utilisation, as it increased the theoretical exergy efficiency by approximately 20%–30% in the four-step TSA CO₂ capture process. In addition, Jiang *et al.*⁸⁵ studied the four-step TSA CO₂ adsorption process incorporated an internal heat recovery, internal mass recovery, and internal heat/mass recovery step, respectively, and showed that these processes exhibited greater energy efficiency than the basic four-step TSA CO₂ adsorption process.

Raganati *et al.*⁸⁶ determined that the main limitation of the conventional TSA process is the dilution of CO₂ by the purging gas, and also used engineered biochars as CO₂ adsorbents in a laboratory-scale experimental study. The CO₂ purity decreased with the introduction of N₂ purging, and thus, the regeneration process that separated the heating and purging steps was studied. The main focus was the trade-off between an increase in CO₂ recovery and a decrease in CO₂ purity with the introduction of N₂ purging. They employed an indirect heating method integrated with additional force enhancement, *i.e.*, a sound-assisted TSA process. Raganati *et al.*⁸⁷ also experimentally investigated a laboratory-scale TSA process in a sound-assisted fluidised-bed reactor, and tested the CO₂ working capacity, dynamic breakthrough, and regeneration performance of CO₂ adsorbents. Moreover, they examined effects of adsorption/desorption temperatures and CO₂ partial pressure on cyclic performance. The abovementioned comprehensive research framework is suitable for the design and optimisation of cyclic configurations using food waste-derived engineered biochars as CO₂ adsorbents.

The heating and cooling steps used in the CO₂ adsorption-desorption processes of the conventional TSA process render it unsuitable for used in a rapid cyclic fashion. Therefore, the vacuum step is combined with a heating regeneration method to give the vacuum-temperature swing adsorption (VTSA) process. This process reduces the temperature and pressure differences required for process-level CO₂ adsorption, thereby shortening residence time for cyclic CO₂ adsorption-desorption processes, and increasing CO₂ productivity and recovery. Plaza *et al.*⁸⁸ used a mathematical model to simulate a VTSA process and then used laboratory-scale experimental data to validate the model. Specifically, they studied a four-step VTSA process, consisting of adsorption, heating and evacuation, cooling, and pressurisation and purging, in a laboratory-scale fixed bed adsorption unit. The physical aspects (mass, energy, and momentum conservation) in the packed-bed reactor were examined by establishing non-isothermal non-adiabatic dynamic model. In addition, adsorption isotherms of pure component were collected to quantify the gas adsorption amounts, and breakthrough experiments were conducted to evaluate overall mass transfer resistance. Durán *et al.*⁸⁹ explored a five-step VTSA process based on a conventional VSA process and developed three columns to decrease its energy requirements. They assessed detailed process configurations in a laboratory-scale fixed-bed reactor, using commercial activated carbon as a CO₂ adsorbent. Li *et al.*⁶³ proved that a VTSA process was superior to both TSA and pressure-temperature swing adsorption processes. The methodology devised for evaluating the cyclic CO₂ adsorption performance of engineered biochars could guide food waste-derived engineered biochar-based CO₂ capture well.

As mentioned, a possible energy-saving route involves the use of low-grade thermal energy in temperature-driven CO₂ adsorption, *i.e.*, integrating renewable energy (*i.e.*, solar thermal energy or geothermal energy) with a TSA process. Dang *et al.*⁹⁰ investigated the integration of a TSA

Table 3 Research summary of the temperature-driven process using engineered biochars as CO₂ adsorbents

Mixture treated	Process configuration	Research approach	Temperature range (°C)	Sample-level characterisation	Process-level evaluation	Results and discussion	Ref.
CO ₂ /N ₂	Four-step TSA	Simulation	100–120	CO ₂ and N ₂ isotherms, adsorbent heat capacity measurement	CO ₂ purity, recovery, productivity, energy consumption, second-law efficiency, capture cost	Comprehensive analysis conducted using cyclic performance indicators to investigate performance in separating CO ₂ from a post-combustion stream; adsorbent selection found to require multi-index evaluation.	83
CO ₂ /N ₂	Four-step TSA (with heat recovery)	Simulation	55, 65, 75, 85	CO ₂ and N ₂ isotherms, adsorbent heat capacity measurement	CO ₂ working capacity, recovery, exergy efficiency	Cyclic performance evaluation results demonstrate that heat recovery has a positive influence on the TSA process	84
CO ₂ /N ₂	Four-step TSA (with internal heat and mass recovery)	Simulation	55, 65, 75, 85	CO ₂ and N ₂ isotherms, adsorbent heat capacity measurement	CO ₂ working capacity, recovery, exergy efficiency	The main parameters, <i>e.g.</i> regeneration heat and exergy efficiency, are determined based on the characteristics of activated carbon, and the internal heat and mass recovery steps are effective.	85
CO ₂ /N ₂	Four-step TSA (with sound assisted)	Experiment	40, 70, 100, 130, 150	None	CO ₂ working capacity, purity, recovery	Heating very efficient for desorbing carbon dioxide, and sound-assisted strategy is effective.	86
CO ₂ /N ₂	Four-step TSA (sound assisted)	Experiment	40, 70, 100, 130, 150	Adsorption dynamic measurement	CO ₂ productivity, working capacity, capture energy cost	Preliminary energy and feasibility estimations suggest that the sound-assisted TSA configuration may be industrially competitive.	87
CO ₂ /N ₂ /H ₂ O	Four-step VTSA	Experiment	70	Adsorption dynamic measurement	Working capacity	Devised model is a good approximation of the physical phenomenon in fixed bed filling with engineered biochars.	88
CO ₂ /N ₂	Five-step VTSA	Experiment and simulation	70	CO ₂ and N ₂ isotherms, breakthrough curves, adsorbent heat capacity, thermal conductivity measurement	CO ₂ purity, recovery	Prediction of the characteristic parameters of cyclic configurations analysed was accurate, different process configurations can be compared fairly.	89
CO ₂ /N ₂	Two-step TSA	Experiment	100–120	Static gas adsorption, dynamic adsorption tests	Working capacity, energy consumption	Adsorption/desorption system integrated with radiative cooling and solar heating devised, thus avoiding use of energy for heating and cooling processes; represents proof-of-concept work that may pave an alternative way to for developing energy-saving TSA techniques.	90

process with passive radiative cooling and solar heating routes in a packed-bed reactor using commercial activated carbon as CO₂ adsorbent. Their results demonstrated that they were viable as energy-saving TSA processes. They measured CO₂ adsorption isotherms and breakthrough from sample-level CO₂ adsorption data, and examined a

packed bed's radiative cooling properties and solar heating effects. Their findings suggested that the integration of renewable energy with food waste-derived engineered biochar-based CO₂ capture is a promising and practical strategy to achieve both carbon neutrality and sustainable waste management.

Other CO₂ adsorption processes

In addition to temperature- and pressure-driven CO₂ adsorption processes, other CO₂ adsorption processes have been developed to evaluate CO₂ working capacities using engineered biochars as CO₂ adsorbents. For example, electric swing adsorption (ESA) has been considered as a promising route to accelerate the heating step by using a low-voltage electric current *via* establishment of the Joule effect. Lillia *et al.*⁹¹ developed a temperature/electric swing adsorption (T/ESA) process for CO₂ adsorption that comprised eight steps, including adsorption, recuperative pre-heating, steam heating, electric heating, purge-to-capture, purge-to-recycle, thermal recovery, and cooling. As both steam heating and electric heating were energy-consuming steps, the sensible heat was recycled in the thermal recovery step and then used for the recuperative pre-heating step. The T/ESA process was developed based on CO₂ gas adsorption isotherms, adsorbent density and porosity, heat capacity, mass transfer coefficient, and electric resistance, which were obtained from sample-level CO₂ adsorption experiments. This T/ESA process was thus found to be applicable to the conventional natural gas combined cycle with an exhaust gas recycle. Zhao *et al.*⁹² experimentally studied a combined electrical and vacuum swing CO₂ adsorption (VESA) process to evaluate the cyclic performance of a novel hybrid monolith. Their findings revealed that the CO₂ purity obtained from the VESA process was up to 33% higher than the CO₂ purity (17%–23%) obtained from a simple VSA process, validating the VESA process as more promising than a VSA process for CO₂ adsorption.

The microwave swing adsorption (MSA) process was devised as an alternative to the TSA process; the MSA involves a fast, contactless, and direct heating route, which is beneficial to increasing CO₂ working capacity and accelerating CO₂ desorption.⁹³ It was found that rapid adsorbent regeneration can be achieved using the MSA process, and also that energy consumption can be reduced by integrating renewable energy with electrified CO₂ capture. Yassin *et al.*⁹⁴ performed detailed comparisons between an MSA process and a conventional TSA process using a rotatory fixed-bed reactor filled with CO₂ adsorbents. They evaluated CO₂ adsorption performance in terms of CO₂ uptake capacity, regeneration efficiency, and rate of regeneration. And they also considered power consumption per adsorbent mass as a CO₂-adsorption performance indicator. They concluded that MSA was an energy-saving process, because the power consumed by microwave regeneration was 18.69% less than conventional TSA process.

In addition, other environmental factors have been investigated. For example, Querejeta *et al.*⁹⁵ studied the effect of humidity on CO₂ adsorption capacity using K₂CO₃-doped engineered biochar samples in a packed-bed reactor. The results revealed that carbonisation occurred in the packed-bed reactor and improved the CO₂ adsorption capacity. In another typical example, Cuesta and Song⁹⁶ devised a novel pH-based swing CO₂ adsorption process by using activated carbon black adsorbents and immobilised carbonic anhydrase biocatalysts for ambient CO₂ capture. This proof-of-concept study revealed

a new way to drive CO₂ enriched from a dilute source to a sink for storage or usage.

Life-cycle assessment of food-waste derived CO₂ adsorbents

Use of food waste-derived engineered biochars for CO₂ capture contributes to sustainable management of food waste and carbon neutrality,⁹⁷ thereby mitigating two critical environmental problems. Life-cycle assessment (LCA) is widely considered to be a powerful and practical tool for fully investigating environmental impacts, thereby affording guidelines for researchers and policymakers.

Life cycle assessment of CO₂ adsorbents

The synthesis of CO₂ adsorbents, particularly green CO₂ adsorbents from waste materials, is considered an emerging technology. Thus, LCA of this emerging technology has received attention in recent years. Table 4 provides a detailed summary of LCAs of various types of CO₂ adsorbents. Researchers have studied biomass, particularly food waste, which is transformed into biochars for use as CO₂ adsorbents. These studies have evaluated the environmental sustainability of this approach using LCA. The food waste that has been used has included waste oyster shells,⁹⁸ coconut shells,^{99,100} wheat flour and glucose,¹⁰¹ and mixed food waste.²⁵ Wang *et al.*⁹⁸ conducted a preliminary and simplified LCA to compare the CO₂ emissions of two calcium oxide (CaO)-based adsorbents derived from waste oyster shells and limestone. Their results indicated that net negative emissions were more easily achieved using oyster shells than limestone. Zhu *et al.*¹⁰¹ conducted a preliminary LCA of five kinds of CO₂ adsorbents synthesised from glucose or wheat flour to provide optimisation information, from an environmental perspective, at the early development stage. Yuan *et al.*²⁵ assessed the environmental impacts of an engineered biochar derived from general mixed food waste and its application for industrial CO₂ capture. They found that their technical route had high potential to achieve a closed carbon loop.

Environmental impacts of food waste-derived CO₂ adsorbents

LCA results from different studies cannot be compared due to the studies having used different system boundaries and functional units (FUs). As shown in Fig. 4, FUs that have been commonly used are 1 kg of adsorbent and 1 kg of CO₂ adsorbed/captured, with cradle-to-gate and cradle-to-grave, respectively, as system boundaries. Specifically, the cradle-to-gate system boundary assesses the impacts of biochar until it leaves the factory gate while the cradle-to-grave system boundary also includes the use and end life of biochar.

We highlight that in both studies, a thorough consideration of all potential carbon emissions during the processes is required, including the additional emissions as by-product. The life cycle equivalent CO₂ emissions of the aforementioned CaO-based adsorbent derived from oyster shells achieved negative CO₂ emissions when coupled with 10 cycles of CO₂ carbo-

Table 4 Life-cycle assessment summary of engineered biochar-based CO₂ adsorption

Type of CO ₂ adsorbent	Framework/origin	Functional unit	System boundary	Inventory data	Environmental impact category	GWP performance (kg CO ₂ eq.)	Impact assessment	Ref.
CaO-based sorbent	Oyster shell	1 kg sorbent	Raw material acquisition, CO ₂ production, recycle and disposal	Laboratory-scale experiment	1	-4.4-0	Midpoint	98
CaO-based sorbent	Limestone	1 kg sorbent				-3.7-0.7	and endpoint	
Physiosorbed carbon nanotube (CNT) supported polyethylenimine (PEI) sorbent	CNT	1 kg sorbent (1 kg CO ₂ adsorbed)	Production, CO ₂ adsorption	Laboratory-scale experiment	1-5, 10, 11, 17, 18	209	Midpoint	102
Covalently bound CNT supported PEI sorbent	CNT					326		
Amine on alumina sorbent	Alumina	1 kg CO ₂ adsorbed	Production and disposal	Scientific paper and database	1-16	12.76 E10-3	Midpoint	99
Amine on silica sorbent	Silica					23.53 E10-3		
Amine on cellulose sorbent	Cellulose					46 E10-3		
Potassium carbonate on silica sorbent	Silica					9.38 E10-3		
Potassium carbonate on activated carbon sorbent	Coconut shell					11.54 E10-3		
Anionic exchange Resin sorbent	Resin					36.30 E10-3		
Activated carbon	Persian ironwood biomass	1 kg CO ₂ adsorption	Production	Laboratory-scale experiment and simulation for scale up	1, 2, 4, 5, 9, 12, 18-22	9.501	Midpoint	103
Copper oxide-activated carbon	Persian ironwood biomass					26.367		
Zeolite (13X-APC)	FAU zeolite	1 kg CO ₂ adsorbed	Production, CO ₂ adsorption and disposal	Laboratory-scale experiment	1, 2, 4, 5, 12, 16, 18-22	4.2	Midpoint	100
Carbon molecular sieve (CMS) (CMS-330)	Coconut shell					7.73		
Biochar	Glucose and sodium	1 kg CO ₂ uptake	Production, CO ₂ uptake	Laboratory-scale experiment	1, 2, 4-6, 8-14, 16, 18, 19, 21, 23	666.26	Midpoint	101
Biochar treated with urea	Glucose and sodium					679.89		
Biochar treated with cobalt acetate	Glucose and sodium					586.41		
Biochar	Wheat flour, Glucose and sodium					138.93-309.43		
Biochar treated with dicyandiamide	Wheat flour, Glucose and sodium					108.36-459.11/		
Biochar	Mixed food waste obtained from the Korea Institute of Civil Engineering and Building Technology	1 kg adsorbent	Production Raw material acquisition, CO ₂ adsorption, disposal	Laboratory-scale experiment	1, 2, 4, 5, 8, 9, 14, 23, 24, 25	12.87	Midpoint	25
Biochar						-5.4	Midpoint	

1. Climate change, 2. Ozone depletion, 3. Respiratory inorganics, 4. Acidification terrestrial and freshwater, 5. Eutrophication freshwater, 6. Eutrophication marine, 7. Eutrophication terrestrial, 8. Ionizing radiation, 9. Photochemical ozone formation, 10. Cancer human health effects, 11. Non-cancer human health effects, 12. Ecotoxicity freshwater, 13. Land use, 14. Water scarcity, 15. Resource use, energy carriers, 16. Resource use, mineral and metals, 17. Smog, 18. Fossil fuel depletion, 19. Marine aquatic ecotoxicity, 20. Human toxicity, 21. Terrestrial ecotoxicity, 22. Photochemical oxidation, 23. Fine particulate matter formation, 24. Abiotic depletion potential, and 25. Primary energy demand.

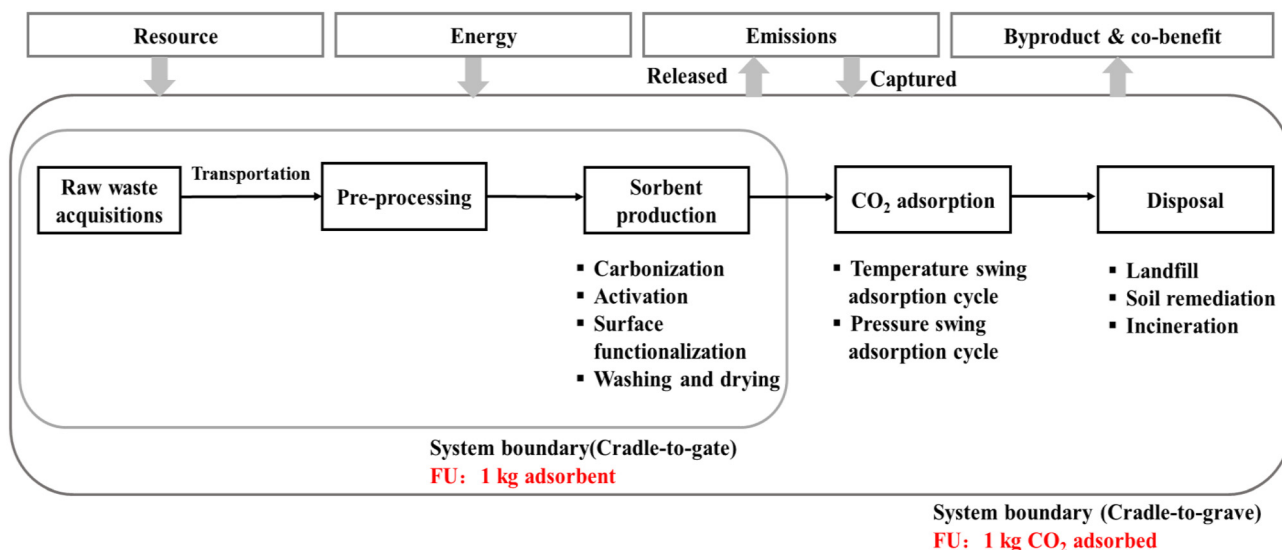


Fig. 4 A general system boundary for food waste derived carbon dioxide (CO₂) adsorbent (FU = functional unit).

nation/calcinations.⁹⁸ Moreover, it was assumed that no CO₂ emissions occurred during the CO₂ adsorption cycle due to waste heat being utilised for CO₂ adsorbent regeneration. Deutz and Bardow⁹⁹ showed that the carbon footprint (11.54 10⁻³ kgCO₂-equivalents per kg CO₂ captured) of a coconut-derived adsorbent performed well compared with five other types of CO₂ adsorbents. Yuan *et al.*²⁵ estimated that the life-cycle CO₂ equivalent emissions of food waste-derived engineered biochar was 12.87 kg within a cradle-to-gate system boundary, which is on the same order of magnitude as engineered biochars prepared from other types of biomass. However, this process may lead to non-negligible impacts on primary energy consumption and water depletion. In addition, in the cradle-to-grave assessment, the CO₂ equivalent emissions of engineered biochar in carbon capture applications play a larger proportion. This implies that a one-sided pursuit of reducing CO₂ equivalent carbon emissions caused by energy consumption during sample preparation may have negative effects, and optimizing the process to improve sample performance may yield great benefits.

Challenges and future prospects

Currently, LCA is widely regarded as an important and powerful tool for assessing the overall performance of emerging CO₂ adsorbents derived from food waste. However, there have been insufficient LCA studies in this field and they lack a consensus framework. Firstly, the technical route of upcycling food waste into engineered biochars for CO₂ adsorption involves multiple functions, including waste management, production of CO₂ adsorbents, and CO₂ mitigation. Thus, inconsistent FUs have been used to describe this process, which makes it difficult to compare the use of different carbon precursors and adsorbent preparation pathways. Secondly, as shown in Table 4, the system boundaries considered vary significantly, and as shown in Fig. 4, few studies have provided a complete cradle-to-gate

or cradle-to-grave assessment. Penultimately, the CO₂ adsorption process has been oversimplified in most studies, and few studies have considered the actual life span of CO₂ adsorbents. In actual industrial scenarios, the condition of the CO₂ sources, the types of CO₂ adsorption cycles, the energy consumed by adsorbent regenerations, and the total cyclic number of CO₂ adsorbents vary significantly and have a considerable influence on LCA results. Finally, laboratory-scale experiment data have been used as the core inventory data in almost all studies, because this technology remains in its early stages. Thus, these data may not necessarily parallel industrial scale data, resulting in rather high potential uncertainty. Moreover, few studies have conducted uncertainty and sensitivity assessments of key factors.

Conclusions and perspectives

This review extensively explored whether food waste-derived engineered biochars have been found to be effective as CO₂ adsorbents, by examining studies on sample preparation with different capture processes (including both sample- and process-level) and on environmental impacts with comprehensive LCAs. This review revealed the viability of food waste-derived engineered biochars as promising alternative materials for CO₂ capture. Moreover, this review addressed challenges in sample preparation process, harnessing ML for optimisation, tailoring cyclic process, and embracing holistic LCA, thereby significantly contributing to the understanding and advancement of sustainable food waste upcycling and carbon capture technologies within the context of a circular economy. It's worth mentioning that wasting food itself is not considered a sustainable practice, by utilizing the wastes generated during the grain production process, such as banana peels, potato peels, these can still be regarded as effective sources of engineered biochars.

Owing to the high moisture content of food waste, anaerobic digestion followed by HTC is highly preferred for upcycling food waste into value-added CH_4 (or H_2) and carbonaceous materials. In particular, the hydrochar obtained in this approach can be further converted into effective catalysts *via* anaerobic digestion, which enhancing CH_4 or H_2 yields.¹⁰⁴ Only a simple batch reactor is required for valorising food waste into valuable gaseous and solid products, making it practical and feasible for commercialisation. Activation and surface functionalisation have been widely used to develop engineered biochars with significant enhancement of their potential applications, especially for CO_2 adsorption. From a life-cycle perspective, HTC, avoiding energy consumption for pre-drying treatment of food waste, is classified as a green technical route to upcycle food waste into solid carbon materials. The environmental impacts potentially introduced by the chemical agents during activation and surface functionalisation also need to be assessed in the context of the trade-off with performance enhancement.

Within the context of carbon neutrality, this review comprehensively addressed food waste-derived engineered biochars for CO_2 capture at both the sample-level and process-level processes. Conventional synthetic routes for high-performance CO_2 adsorbents are both time- and labour-intensive, therefore, ML is considered as an emerging technology for accelerating synthesis of biomass-based CO_2 adsorbents and effectively providing valuable guidelines for inverse design of engineered biochars with high-performance CO_2 capture. These results promote commercial applications of engineered biochar-based CO_2 adsorption approaches. As ML is a data-driven approach, data collection and pre-processing treatment need to be improved, which are critical for providing accurate performance prediction and valuable guidelines for CO_2 adsorbent synthesis. Moreover, various factor-driven CO_2 adsorption processes, including TSA and PSA processes were reviewed and compared in detail. Research on a wide range of cyclic processes has underscored the necessity of tailoring cycle configurations to match the characteristics of food waste-derived engineered biochars. The judicious selection of cyclic processes, with a particular focus on adsorption isotherm profiles, plays a pivotal role in enhancing cyclic performance.

When targeting the UN SDGs and striving towards achieving a circular economy, the environmental impacts of emerging technical routes need to be comprehensively assessed from a life-cycle perspective. This review evaluated studies on CO_2 adsorption using food waste-derived engineered biochars with the consideration of various system boundaries (*i.e.*, cradle-to-gate and cradle-to-grave). These studies suggested that food waste-derived engineered biochars for CO_2 capture has great potential to achieve a closed carbon loop and is thus a promising alternative to conventional CO_2 adsorption methods. However, these studies highlighted that there is currently no standardized research approach to provide a unified assessment for the selection of different boundaries, FUs, and environmental impact parameters. The LCA-based conclusions have limited applicabilities and cannot be compared across

different studies horizontally. Considering the uncertainties of laboratory-scale investigations, more comprehensive assessments need to be performed to ensure that food waste-derived engineered biochars makes substantial contributions to a circular economy and environmental sustainability.

Author contributions

J. Wen, J. Wang, and S. Li contributed equally to this work. J. Wen – conceptualization, investigation, visualization, writing – original draft, review & editing; J. Wang and S. Li – investigation, data curation, formal analysis, visualization, writing – original draft, review & editing; J.T.E. Lee – writing – review & editing; C.S.K. Lin, O. Mašek, and H. Zhang – supervision, writing – review & editing; X. Yuan – supervision, visualization, validation, funding acquisition, writing – original draft, review & editing.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This research was supported by the Start-up Research Fund of Southeast University (RF1028623274) and Youth Program of National Natural Science Foundation of China (Grant No. 72104257). J. T. E. Lee was supported by the National Research Foundation, Prime Minister's Office, Singapore under its Campus for Research Excellence and Technological Enterprise (CREATE) programme.

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