1	Thermochemical upcycling of food waste into engineered biochar for energy and
2	environmental applications: A critical review
3	Xiangzhou Yuan <sup>a,b</sup> , Yafei Shen <sup>c,d,*</sup> , Piumi Amasha Withana <sup>b</sup> , Ondřej Mašek <sup>e</sup> , Carol S.K. Lin <sup>f</sup> ,
4	Siming You <sup>g</sup> , Filip M.G. Tack <sup>h</sup> , Yong Sik Ok <sup>b,*</sup>
5	<sup>a</sup> Ministry of Education of Key Laboratory of Energy Thermal Conversion and Control,
6	School of Energy and Environment, Southeast University, Nanjing 210096, China
7	<sup>b</sup> Korea Biochar Research Center, Association of Pacific Rim Universities (APRU)
8	Sustainable Waste Management Program & Division of Environmental Science and
9	Ecological Engineering, Korea University, Seoul 02841, Republic of Korea
10	<sup>c</sup> Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control
11	(AEMPC), Collaborative Innovation Center of Atmospheric Environment and Equipment
12	Technology, School of Environmental Science and Engineering, Nanjing University of
13	Information Science and Technology, Nanjing 210044, China
14	<sup>d</sup> Residues and Resource Reclamation Centre (R3C), Nanyang Environment and Water
15	Research Institute, Nanyang Technological University, Singapore 637141, Singapore
16	<sup>e</sup> UK Biochar Research Centre, School of GeoSciences, University of Edinburgh, Alexander
17	Crum Brown Road, Crew Building, EH9 3JN Edinburgh, UK
18	<sup>f</sup> School of Energy and Environment, City University of Hong Kong, Tat Chee Avenue,
19	Kowloon, Hong Kong, China
20	<sup>g</sup> James Watt School of Engineering, University of Glasgow, Glasgow G12 8QQ, United
21	Kingdom
22	<sup>h</sup> Department Green chemistry and Technology, Ghent University, Coupure Links 653, 9000
23	Ghent, Belgium
24	* Corresponding authors:
25	yongsikok@korea.ac.kr (Y.S. Ok); shen1225@nuist.edu.cn (Y. Shen)

#### 26 Abstract

Environmental issues caused by food waste are important concerns for human well-27 being and ecosystem health. Valorization of food waste into energy and carbon 28 materials has been extensively investigated. Here, we reviewed the most recent 29 advancements in the thermochemical conversion of food waste into engineered biochar. 30 Synthesis routes and practical applications of the food waste-derived biochar was 31 32 succinctly reviewed. Engineered biochar is a promising alternative for mitigating environmental pollution and alleviating energy crisis. The underlying relationships 33 between engineered biochar properties and specific applications are still unclear, 34 therefore, machine learning-aided engineered biochar design and process optimization 35 was proposed. Moreover, before any industrial scale implementation, detailed 36 assessments of the environmental benefits and economic feasibility must be conducted. 37 38 In the context of carbon neutrality, thermochemical upcycling of food waste into engineered biochar for energy and environmental applications can significantly 39 contribute to attaining sustainable food waste management, mitigating environmental 40 pollution, and addressing the energy shortage crisis and thus will eventually facilitate 41 the fulfillment of United Nations Sustainable Development Goals (SDGs). Furthermore, 42 the existing challenges in the practical valorization of food waste into engineered 43 44 biochar are comprehensively discussed, and outlooks are proposed.

Keywords: Biochar catalyst; Pyrolysis; Carbon capture and storage; Machine learning;
Sustainable Development Goals

#### **1. Introduction** 48

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Food waste is a global issue that brings serious environmental, economic and social 49 challenges [1-3]. Many countries have taken measures for reduction of food waste 50 51 generation and sustainable management of unavoidable waste via controlling the sources and collecting the food waste separately from other wastes. The collected 52 food waste could be further processed using various technical approaches to minimize 53 its environmental impact and to maximize its benefits within a circular economy. 54 Food waste, which is mainly composed of organics (i.e., carbohydrates, proteins, fats, 55 and lipids), is hereby converted into high value-added products (e.g., methane, 56 bioethanol, syngas, chemicals, and carbon materials) [4]. Recently, the upcycling of 57 food waste into advanced carbon materials has been considered as an emerging 58 59 paradigm shift from landfilling and incineration. The major upcycling approaches include thermochemical conversions (e.g., hydrothermal carbonization (HTC), 60 pyrolysis, and gasification) and biological conversions (e.g., anaerobic digestion 61 62 (AD)) [4, 5]. AD is extensively investigated for transforming food waste into valueadded products, owing to its considerable advantages of cost-effectiveness, mild 63 operating conditions, and relatively low energy consumption [6-9]. However, AD 64 generally requires a long residence time and a high-quality feedstock [10]. 65 Compared with the biological conversions discussed above, thermochemical 66 conversion is fast and achieves high yields of value-added products [11]. Pyrolysis, a 67 conventional thermochemical conversion, requires large amount of energy to recover

69	value-added products and thermal energy from food waste directly, owing to the food
70	waste's actual characteristic of high moisture content. To effectively upcycle food
71	waste with high moisture content, dewatering (or pre-drying) is an essential pre-
72	treatment followed by pyrolysis. However, without pre-drying treatment, HTC is
73	considered one promising alternative for food waste valorization, reducing extra water
74	requirement in hydrothermal reactors and increasing conversion efficiency [11, 12].
75	The main products from slow pyrolysis and HTC are carbon-rich materials termed as
76	biochar and hydrochar, respectively, which can be further activated and/or modified
77	into advanced engineered biochar for various applications. Researchers are currently
78	focusing on developing green, sustainable, and cost-effective thermochemical
79	processes or integrated biological and thermochemical processes to achieve
80	sustainable management of food waste towards a circular economy (as given in
81	Figure 1). Due to the diversity and complexity of food waste, more feedstock-specific
82	process technologies are required for food waste treatment in biorefineries and
83	environmental protections from perspectives of environmental impact and economic
84	feasibility [13, 14]. Solid carbon residue that cannot be further biologically
85	decomposed or chemically extracted normally accounts for a large proportion of
86	processed food waste. These residues can be converted into solid biochar, liquid bio-
87	oil, and gaseous products via hydrothermal conversion or pyrolysis. So far, only a few
88	papers have reviewed the thermochemical conversion of food waste, focusing on the
89	operating conditions, reaction mechanisms, product distributions, and evolution
90	pathways. In particular, the gaseous and liquid products derived from various food

91	wastes have been extensively investigated [15-18] in comparison to the solid biochar
92	products. For example, Pham et al. [15] reviewed food waste-to-energy technologies
93	involving biological conversion using AD or fermentation, and thermochemical
94	processes including pyrolysis, incineration, hydrothermal oxidation, and gasification
95	Current state-of-the-art in the pyrolysis and gasification of food waste was reviewed
96	by Su et al. [17], Murugesan et al. [18], and Makkawi et al. [19].

97 Therefore, it is necessary to provide a timely and comprehensive review of food waste-derived engineered biochar for environmental and energy applications, 98 including major and emerging production routes of engineered biochar, machine 99 100 learning (ML)-aided guidelines for application-targeted engineered biochar, practical applications in environmental and energy fields, and conclusions and outlook. 101 102 Physical, chemical, and integrated methods are used to activate (i.e., developing new 103 pores and controlling pore size distributions) and/or modify (i.e., doping surface functional groups) biochar into engineered biochar with wide applications, such as 104 solid fuels, adsorbents (e.g., for CO<sub>2</sub> capture), and advanced catalysts (e.g., for 105 106 electrochemical energy storage and conversion). The review sheds light on the sustainable upcycling of food waste into engineered biochar with high-performance 107 applications and ultimately promotes their large-scale deployment, significantly 108 109 providing benefits and guidelines to researchers from both academies and industries working in the areas of sustainable valorization of solid waste, climate change 110 111 mitigation, renewable fuels, energy conversion and storage, and circular carbon economy. In the context of carbon neutrality, sustainable management of food waste, 112 5

- 113 climate change mitigation, and renewable alternative fuels need to be performed in a
- sustainable route to achieve United Nations Sustainable Development Goals, based on
- 115 the takeaways from this comprehensive review.



Figure 1. Schematic diagram of thermochemical processing of food waste to produce
biochar and engineered biochar materials for various applications.

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### 120 2. Thermochemical upcycling of food waste

- 121 2.1. Hydrothermal conversion
- 122 Hydrothermal conversion (HTC) allows for the synthesis of value-added carbon
- 123 materials and the recovery of energy from food waste with high moisture content [20,
- 124 21]. HTC at relatively low temperature (180-265 °C) and low pressure (subcritical
- 125 conditions, up to 6.0 MPa) is favorable for hydrochar production [12, 22]. Compared
- 126 with other thermochemical conversion techniques, HTC for converting food waste
- 127 into hydrochar as the primary product avoids the requirement for a pre-drying
- treatment, involves rather mild operating conditions, and features high operational

129	feasibility and relatively low energy consumption [4, 11, 23, 24]. HTC can recover
130	nutrients and fatty acids in liquid food waste, which can be intensively recirculated
131	during processing [25-27]. The range of processes and mechanisms involved in
132	hydrochar formation from food waste via HTC are schematized in Figure 2. The
133	chemical composition of food waste is complex. Lipids, proteins, and carbohydrates
134	are main component groups. During HTC, subcritical water promotes the hydrolysis
135	of lipids, polysaccharides, and proteins into oligomer and monomer units [28, 29].
136	The decomposition of proteins usually requires a minimum operating temperature of
137	150 °C but needs to be at least 220 °C if the protein content is high, above 30 wt.%
138	[28]. Protein hydrolysis is typically slower than carbohydrate hydrolysis due to the
139	higher stability of peptide bonds than glycosidic bonds.
140	Researchers widely investigated specific mechanisms involved in the HTC of food
141	waste [29, 30]. Yang et al. [29] suggested that the HTC of complex food waste
142	involved three key reactions: carbohydrates carbonization, Maillard reaction, and
143	amidation reaction. The components of food waste play different roles in hydrochar
144	formation. Lipids play a crucial role in hydrochar formation by inhibiting interactions
145	between carbohydrates and proteins, and carbohydrates play a key role when proteins
146	boost their N-heterocyclic functional groups. As hydrochar surface can adsorb the
147	lipid hydrolysate, the lipids present in the food-waste do not affect the carbonization

148 process [31].



Figure 2. Major reactions involved in hydrochar formation via the hydrothermal conversion of food waste [30]. Reproduced with the permission from Elsevier

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153 2.2. Pyrolysis

154 Compared with HTL, pyrolysis is the thermal degradation of organic materials into

- carbon-rich by products at a moderate temperature (300-800 °C) and low pressure
- 156 with little or no available air. A range of atmospheres can be used to ensure anaerobic
- 157 conditions, including inert gases such as nitrogen, argon, or helium; reactive gases
- such as carbon dioxide or steam; or vacuum. The properties of the products based on
- 159 pyrolysis are mainly influenced by the operating conditions, such as the type of inert

160 gas, flow rate of carrier gas, temperature, heating rate and residence time [32-35].

161	In general, pyrolysis has several advantages due to diverse output products (as liquid,
162	solid, and gas), environmental friendliness and the maturity of the technology. The
163	yields and properties of the target products can be optimized by adjusting the
164	operating conditions of pyrolysis [36]. The valorization of food waste into biochar
165	and bio-oil like products is a highly viable choice in accordance with circular
166	economy principles [37]. State-of-the-art biofuel production from conventional and
167	advanced pyrolysis and the co-pyrolysis of food waste were reviewed recently [38].
168	Various mechanisms are involved in the pyrolysis of food waste [39]. Glucose is one
169	of the most abundant monosaccharides in food waste. During pyrolysis, a dehydration
170	reaction converts glucose into anhydrosugars, which are converted to sorbitol and 1,4-
171	sorbitan through a reduction reaction and a dehydration reaction, respectively,
172	subsequently converted into isosorbide [40]. During the pyrolysis of triglycerides, the
173	major component is lipids. Via decarbonylation, decarboxylation, and fragmentation
174	reactions, glycerol can be converted into acids, esters, alcohols, hydrocarbons, and
175	alicyclic compounds. The decomposition of steroids could generate hydrocarbons
176	[41]. Amine and diketopiperazine species are produced during the pyrolysis of
177	proteins. In the process, amino acids are connected by peptide bonds. Amine species
178	are produced via the breakage of peptide bonds, while diketopiperazine species are
179	generated because of the cyclization of amino acids [42]. To date, investigations of the
180	food waste pyrolysis mechanisms have focused on the individual components.

However, the complex interactions between the different components in food wasteduring pyrolysis remain unclear and should be investigated further.

183	Direct pyrolysis of food waste with high water content would, while technically
184	feasible, require considerable energy input for the drying pretreatment, and
185	evaporation of remaining water. Therefore, integrated conversion systems, e.g., AD +
186	pyrolysis; HTC + pyrolysis, have been developed to efficiently achieve the circular
187	economy of food waste [43-45]. Both AD and HTC readily reduce the water content
188	(especially bound water) from food waste by destroying the cell walls [46]. More
189	importantly, AD recovers valuable biogas and nutrients (digestate) from food waste
190	with high water content. The solid AD residue can then be pyrolyzed for the co-
191	production of biochar and biofuels (e.g., syngas), as illustrated in Figure 3. The
192	pyrolysis of solid AD residue from food waste has increasingly attracted the interest
193	of researchers [47-49]. Zhao et al. [47] comparatively studied the pyrolysis of food
194	waste and its solid digestate. Organic matter of food waste was partially consumed
195	during AD, influencing the pyrolysis performance of the resulting solid digestate. The
196	pyrolytic gas from food waste was of better quality because its lower heating value
197	was high. In contrast, the bio-oil and biochar derived from the solid digestate showed
198	better qualities as liquid fuel and carbon sorbents. Pyrolysis of the solid digestate
199	generated fewer N-containing pollutants, indicating that AD coupled with pyrolysis is
200	a more environmentally friendly approach for treating food waste [50].



201

Figure 3. Integrated anaerobic digestion and pyrolysis process for efficiently converting food waste into value-added products toward sustainable food waste management [43]. Copyright permission from Elsevier (License Number: 5437490493245).

#### 207 **3. Development of engineered biochar derived from food waste**

- 208 3.1. Activation to engineered biochar
- 209 Biochar and hydrochar have a limited surface area and pore structure, and unspecified
- 210 functional groups [34, 51]. Activation treatment aims to improve the textural
- 211 properties of biochar and hydrochar by creating and/or digging out the internal porous
- structure in order to broaden the practical applications in environmental protection
- and energy storage/conversion [51-53].
- 214 Generally, activation treatments are categorized as physical and chemical. In physical
- 215 activation, CO<sub>2</sub>, steam, and air at high operating temperatures (even up to 1,000 °C)

216	are widely used [11, 54]. Physical activation is a relatively eco-friendly and cost-
217	effective approach for increasing textural properties (i.e., porosity). Among these
218	post-modification treatments, physical activation exhibits lower global warming
219	potential than chemical activation [55]. Steam and CO <sub>2</sub> , both endothermically react
220	with carbon via two major reactions: $C + CO_2 \rightarrow 2CO, \Delta H = 159 k Jmol^{-1}; C +$
221	$H_2 O \rightarrow CO + H_2, \Delta H = 119 k Jmol^{-1}$ [54]. Steam activation effectively improves the
222	porosity and increases the number of O-functional groups on the surface of
223	engineered biochar [56]. Due to its easy handling and low reactivity even at operating
224	temperatures higher than 700 °C, CO <sub>2</sub> gas has become the preferred activating agent,
225	making activation easier to control [57, 58]. However, in the context of carbon
226	neutrality, CO <sub>2</sub> emissions from CO <sub>2</sub> activation are an issue. Although the surface area,
227	total pore volume, and yield of engineered biochar are generally less than 1,000 m <sup>2</sup> /g,
228	$0.7 \text{ cm}^3/\text{g}$ , and 30%, respectively, after physical activation [51, 59], the approach has
229	several key advantages, including minimal corrosion to the activating reactor, high
230	deployment feasibility and low production and operating costs, showing a broad
231	commercial potential of engineered biochar-based practical applications.



238 energy consumption due to the low activation temperature and short activation time, and high porosity and yield of the produced engineered biochar. However, issues 239 240 include intensive chemical corrosion of the reactor during activation, the need for 241 wastewater treatment due to the production of chemically contaminated wastewater, and the extra energy required for subsequent drying of the produced carbon materials. 242 243 KOH activation is one of the most common and promising routes to produce well-244 developed porous engineered biochar. In a study engineered biochar was synthesized using KOH activation from water chestnut shells [61], achieving a surface area of 245 246 3,401 m<sup>3</sup>/g and a total pore volume of 2.50 cm<sup>3</sup>/g. It indicates upcycling food waste into engineered biochar has significant potential for practical applications such as gas 247 248 adsorption and energy storage. However, KOH activation has considerable 249 environmental impacts, including greenhouse gas emissions from activation and high-250 water consumption for the impregnation before activation and the washing after 251 activation. The environmental impacts need to be fully assessed from a life-cycle 252 perspective when designing a sustainable and feasible activation technique [62].

# 253 3.2. Modification of surface functional group

254 In addition to good textural properties, the presence of surface functional groups, such

as O-, N-, and S-rich groups, critically determines the potential application

256 performance of engineered biochar [63]. For example, typical surface O-functional

257 groups (e.g. -OH and -COOH) dramatically improve the adsorption capacity for

258 capturing CO<sub>2</sub> and removal of metal contaminants from wastewater [64-65].

259	Functional groups can be doped on the surface of engineered biochar through various
260	modifications [51, 66]. To increase the oxygenated functional groups (e.g., carboxyl
261	and phenolic hydroxyl), surface oxidation, most commonly with H <sub>2</sub> O <sub>2</sub> and HNO <sub>3</sub> , is
262	the most commonly used method [66, 67]; surface amination, such as high-
263	temperature ammonia (NH <sub>3</sub> ) treatment, can be used to increase amino groups [68];
264	surface sulfonation using concentrated sulfuric acid can be used to enrich sulfonic
265	groups on the surface of engineered biochar. In addition to these single-heteroatom
266	doping treatments, dual- or tri-doping treatments are used to enrich surface active
267	sites, enhancing application performance via synergistic effects. For example, N and S
268	dual-doping is suited for producing engineered biochar for oxygen reduction reactions
269	(ORRs), rechargeable lithium-oxygen batteries, and supercapacitors [69-72].In
270	addition, as shown in Table 1, the main advantage of food waste-derived engineered
271	biochar materials is the production of heteroatom (e.g., O, N, S) self-doped porous
272	carbons owing to the presence of protein.

<b>Biomass source</b>	Advantages	Disadvantages
Food waste	1) Relatively low ash	1) Pretreatment such as
	content;	dewatering is required
	2) Relatively low	due to high water content;
	temperature for	2) The composition is
	carbonization and	complex, normally
	activation process;	including starch, sugars,
	3) Suitable for HTC to	protein, lipids, which are
	produce hydrochar;	difficult to regulate the
	4) Production of	pore structure.
	heteroatom (e.g., N, S)	-
	self-doped porous carbons	
	if protein used;	
	5) Uniform structures	

Table 1 Comparison of food waste and lignocellulosic biomass-derived engineered
 biochar materials

	compared with lignocellulosic biomass- derived biochar.	
Lignocellulosic biomass	<ol> <li>Less energy consumption in dewatering</li> <li>Larger specific surface area compared with food waste-derived biochar</li> <li>Hierarchical pore structure</li> </ol>	<ol> <li>High ash content</li> <li>High energy consumption in the grinding process</li> <li>Non-uniform pore structures</li> </ol>

276 3.3. Machine learning-aided guidelines

Machine learning (ML), an artificial intelligence approach, is a promising and 277 278 practical alternative to laboratory experiments and computational simulations (e.g., 279 density functional theory and Grand canonical Monte Carlo) for designing and optimizing advanced materials [73]. Several major advantages of ML include 280 efficiently screening large amounts of data within a short time (at the 10<sup>-2</sup> s level), 281 282 accurately predicting material properties, and effectively identifying the inherent 283 relationship between material properties and specific applications [74]. Recently, ML 284 has been extensively used to accelerate carbon material discovery and elucidate the inherent relationships with their properties and specific applications. As of December 285 2022, Web of Science Core Database search using the search terms 'carbon materials' 286 and 'machine learning' retrieved 98 peer-reviewed publications; specifically, five 287 publications in 2019, 20 publications in 2020, 29 publications in 2021 and 38 288 publications in 2022, implying that the discovery and application of carbon materials 289 using ML models is an emerging research trend. 290

291	To the best of our knowledge, most of these publications focused on the forward
292	prediction of target features based on collected datasets of input features by
293	established models within acceptable or reasonable accuracy levels ( $R^2 > 0.8$ ) [74-77].
294	For example, Yuan et al. [77] applied ML to predict CO <sub>2</sub> adsorption performance
295	using biomass waste-derived carbon materials, and Li et al. [76] applied ML to predict
296	and optimize the properties of hydrochar derived from high-moisture municipal solid
297	waste. ML algorithms can improve solid waste-to-resource strategies and help to
298	achieve several United Nations (UN) Sustainable Development Goals (SDGs).
299	Typical forward prediction models are deduced from Pearson's correlation matrix,
300	Shapley values, and others, highlighting the importance of input features for
301	predicting target features [74, 77]. Moreover, the data generated from these models
302	must be interpreted with expert knowledge and experience to draw new conclusions.
303	In addition to forward prediction, ML algorithms are powerful for supporting inverse
304	design, greatly facilitating a paradigm shift towards a closed-loop guideline [51, 80].
305	Through synthesis and validation, ML algorithms enable guided experiments to
306	discover food waste-derived biochar functional materials with bespoke properties.
307	When integrating computational simulations with experimental validation, ML
308	algorithms can accelerate the synthesis of biochar functional materials derived from
309	food waste targeted toward a specific application.

Since ML is a data-driven approach, consistent data formatting (i.e., unit consistency)
is an essential step in the process. Missing data is a critical issue when performing

ML-aided investigations. Generally, each piece of data consists of valid values for all input variables, and those with unsolved missing data are discarded. Sometimes, the missing data issue can be solved by data imputation. Most data are manually extracted from recent publications, which are both time- and labor-consuming. Therefore, it is desirable to create open databases with standard formats to accelerate the ML-based discovery of food waste-derived advanced materials and their potential applications.

318

#### 319 4. Practical applications of food waste-derived engineered biochar

320	Engineered biochar produced from food waste has broad applications, including solid
321	fuels, adsorbents for environmental remediation (e.g., CO2 capture), and
322	electrochemical applications (e.g., energy storage and conversion) [11, 50, 65, 81].
323	Food waste-derived hydrochar produced via HTC has been widely developed for
324	direct use as solid fuels. Moreover, hydrochar produced from the HTC and pyrolysis
325	of food waste can be activated and/or modified to obtain high-performance
326	engineered biochar for energy and environmental applications.
327	4.1. Solid fuels
328	Because food waste is mainly composed of organic components (i.e., carbohydrates,
329	protein, and lipids), it has good potential for producing solid fuel with a relatively

330 high heating value. Currently, significant efforts have been taken to produce solid

fuels from food waste via HTC [82, 83]. Theppitak et al. [83] compared the pyrolysis

332	and HTC of food waste for solid fuel production. Compared with the biochar
333	produced via HTC, that produced via pyrolysis at temperatures higher than 300 $^{\circ}$ C
334	showed superior fuel properties, with a high heating value and high fixed carbon
335	content. When the moisture content of the food waste is high (i.e., $>75\%$ ), the HTC
336	process is more suitable and economically efficient. Moreover, the HTC process
337	removes protein from food waste and simultaneously increases the yield of biochar
338	with a higher heating value due to the carbohydrate-rich fraction. The HTC process
339	also resolves issues related to the heterogeneity of mixed food waste [84]. However,
340	the impacts of factors such as biochar palletization, gas composition, NO <sub>x</sub> emission
341	and by-product utilization in the following thermochemical process (i.e., combustion)
342	should be considered [85, 86]. Su et al. [24] recently studied the conversion of food
343	waste (after extracting oil) into solid fuel by the HTC process in a large-scale plant
344	(200 t/d). HTC conditions like residence time, temperature, can significantly influence
345	the physicochemical characteristics, combustion behavior, and alkali metal removal
346	capacity of the produced hydrochar. Most food wastes are composed of
347	carbohydrates, protein, and lipids, making them suitable for the production of solid
348	fuels with a high heating value. Compared with other biomass wastes such as
349	agroforestry residues, food waste also has a lower ash content, especially of alkali and
350	alkaline earth minerals, which may reduce sintering and corrosivity. However, food
351	waste has a high moisture content, so developing a cost-effective dewatering pre-
352	treatment remains a key challenge for advancing practical applications. Moreover, the
353	co-production of oil fuels and biogas can greatly improve the technical- and economic

354 feasibility of converting food waste into energy products.

# 355 4.2. CO<sub>2</sub> adsorption

356	In the context of carbon neutrality, carbon capture is essential to mitigate climate
357	change [87] owing to the current high atmospheric CO <sub>2</sub> concentration (up to 420
358	ppm) and the increasing annual growth rate of $CO_2$ (up to 2.4 ppm per year) [88].
359	Carbon capture, alternatively known as greenhouse gas removal or CO2 removal
360	mainly addresses CO <sub>2</sub> removal from large point-emission sources (e.g., power plants
361	and cement industries) and even from the atmosphere (i.e., direct air capture) [89].
362	Among the available technical approaches, CO <sub>2</sub> adsorption is widely considered a
363	promising and feasible route owing to its mild operating conditions, cost-effectiveness
364	(especially when using solid waste as a precursor for adsorbent synthesis), high
365	feasibility for commercial-scale application, and low energy consumption [11, 51].
000	The simular economic inclusion days and for a days at interview and a show motorials
300	The circular economy-inspired upcycling of food waste into novel carbon materials
367	for CO <sub>2</sub> adsorption followed by soil remediation has been proposed with the potential
368	of achieving negative carbon emissions [62], representing a sustainable waste-to-
369	resource strategy that simultaneously mitigates climate change. Moreover, surplus
370	electricity, engineered biochar material and captured CO2 gas can be generated in a
371	sustainable and multifunctional food waste-based system that integrates temperature
372	swing CO <sub>2</sub> adsorption to reach zero carbon emissions. No waste is generated in this
373	circular life-cycle system, which is beneficial for achieving UN Sustainable
374	Development Goals 11 (sustainable cities and communities), 12 (responsible 19

375	consumption and production), and 13 (climate action). Engineered biochar for $CO_2$
376	adsorption is expected to have a high CO2 adsorption capacity, low energy
377	requirement for regeneration, excellent cyclic stability, high selectivity for CO2 over
378	other gases, and fast kinetic characteristics for adsorption-desorption cycles [51, 90].
379	The CO <sub>2</sub> uptake versus the surface area of food waste (e.g., fruit waste, vegetable
380	waste, shrimp shells, coconut shells, and spent coffee grounds)-derived engineered
381	biochar for CO <sub>2</sub> adsorption is investigated [62, 91, 92], illustrating two major
382	findings. First, through effective synthesis routes, most food waste-derived engineered
383	biochar materials have a viable CO <sub>2</sub> capture performance of more than 3 mol/kg at
384	25 °C (1 bar), in accordance with the US National Energy Technology Laboratory
385	criterion for applicable and practical CO <sub>2</sub> capture. Second, the relationship between
386	$CO_2$ uptake and Brunauer-Emmett-Teller surface area ( $S_{BET}$ ) is not linear, suggesting
387	that CO <sub>2</sub> uptake does not exhibit a directly proportional relationship with $S_{BET}$ .
388	However, when plotting CO <sub>2</sub> uptake versus $S_{BET}$ for materials with pores smaller than
389	$\beta$ nm ( <i>S</i> <sub><math>\beta</math></sub> ), an excellent linear relationship was observed between CO <sub>2</sub> uptake and <i>S</i> <sub>0.8</sub> ,
390	implying that the performance of food waste-derived engineered biochar materials for
391	CO <sub>2</sub> adsorption is significantly mediated by the surface area, limited by narrow pores
392	smaller than 0.8 nm at 25 °C (1 bar) [62, 93]. As shown in a typical case study
393	presented, the spent coffee grounds generated from Starbucks® is upcycled into
394	engineered biochar with a high-performance CO <sub>2</sub> adsorption capacity of 4.54 mol/kg
395	at 25 °C (1 bar) via simple and sustainable synthesis routes [92], demonstrating the
396	great potential for commercial-scale food waste conversion for CO <sub>2</sub> capture

398	Before globally deploying CO <sub>2</sub> adsorption techniques using food waste-derived
399	engineer biochar materials as adsorbents, several existing challenges or barriers must
400	be addressed. First, CO <sub>2</sub> selectivity over other gases (e.g., N <sub>2</sub> ) is a critical factor in the
401	potential performance of commercial-scale CO <sub>2</sub> adsorption [94, 95]. To the best of our
402	knowledge, food waste-derived engineered biochar materials have relatively low CO <sub>2</sub>
403	selectivity, which could result in low CO <sub>2</sub> purity when evaluating cyclic performance
404	using pressure temperature swing or temperature vacuum swing CO <sub>2</sub> adsorption [94].
405	Moreover, low CO <sub>2</sub> purity products would require further gas sorption and separation
406	treatments to obtain high-purity CO <sub>2</sub> gas for sustainable and practical CO <sub>2</sub> utilization,
407	dramatically increasing the investment for CO <sub>2</sub> capture. Therefore, how to efficiently
408	design engineered biochar materials derived from food waste with high CO <sub>2</sub>
409	selectivity remains a significant challenge to be investigated.
410	Second, integrating ML to guide high-performance CO <sub>2</sub> adsorbent preparation is
411	considered a promising route to accelerate the deployment of practical CO <sub>2</sub> adsorption
412	applications [96]. However, thus far, only CO <sub>2</sub> adsorption capacity has been predicted
413	by applying ML to synthesize and optimize engineered biochar materials [74, 97].
414	Other critical indicators, especially CO <sub>2</sub> selectivity, also need to be incorporated into
415	ML models. In addition, following the inverse design perspective, the low $CO_2$
416	selectivity issue might be solved via several iterations of ML model development and
417	experimental validation [98].

21

Third, environmental impact and economic feasibility are the key factors to assess the 418 emerging CO<sub>2</sub> adsorption technologies from both life cycle and techno-economic 419 420 perspectives, verifying if environmental benefits and economic feasibility could be achieved [56, 62]. A paradigm shift to interdisciplinary collaboration in CO<sub>2</sub> 421 422 adsorption research is urgently required to achieve the global adoption of CO<sub>2</sub> adsorption technologies that can be deployed globally. Collaboration between material 423 424 science, data science, thermochemical engineering, and environmental engineering 425 researchers is required for developing adequate advanced CO<sub>2</sub> adsorption systems 426 using food waste-derived engineered biochar materials.

427 4.3. Energy storage and conversion

Biochar derived from food waste can be activated or modified as engineered biochar 428 materials; N-doped engineered biochar materials show particularly high potential for 429 electrochemical applications [81, 99]. To ensure stable performance, many work have 430 431 focused on the synthesis of engineered biochar materials with stable properties using 432 specific food waste instead of mixed waste. Table 2 summarizes the preparation, 433 properties, and performance of engineered biochar materials derived from various food wastes, including carbohydrates (e.g., fruit and starch), protein (e.g., fish), and 434 chitin-enriched waste (e.g., shrimp shell), for advanced electrochemical applications, 435 such as supercapacitors, batteries, ORRs, and hydrogen evolution reactions (HERs). 436 In general, the electrochemical performance of these engineered biochar materials is 437 influenced by the textural structure (e.g., hierarchical porous structure), specific 438

439	surface area, heteroatom (e.g., N, S, P, and O) doping, and active metal (e.g.,
440	transition metal) loading. Accordingly, these parameters depend on the feedstock type
441	and activation/modification methods used. Carbon materials possessing high specific
442	surface areas, hierarchical porous structure, multiple heteroatom-doping, and metal
443	catalyst loading have been shown to exhibit high electrochemical performance. In
444	particular, hierarchical engineered biochar materials synthesized from food waste
445	have been developed for supercapacitor applications (Table 2). Even in the absence of
446	metal catalysts, the maximum capacitance can reach 476,000 F/kg, with high cycling
447	stability (90.9% of the original capacitance retained after 10,000 cycles at
448	1,000 A/kg current density) [100]. This high performance is attributed to the
449	properties of fishbone, which is mainly composed of protein (N, S, and C source) and
450	CaCO <sub>3</sub> (self-template), thereby producing N- and S-doped engineered biochar. The
451	selection of a suitable type of food waste is important for the synthesis of functional
452	carbon materials. For example, shrimp shell composed of chitin (N and C source) and
453	inorganic CaCO <sub>3</sub> (self-template) is considered a promising candidate for producing N-
454	doped engineered biochar [101-103]. Gao et al. [101] prepared N-doped hierarchical
455	engineered biochar from shrimp shells containing abundant mineral scaffold (CaCO <sub>3</sub> )
456	as a self-template combined with KOH activation. Shrimp shells were first activated
457	by KOH in an Ar atmosphere to produce micropores at 700 $^\circ$ C, and then the CaCO <sub>3</sub>
458	was removed using acetic acid to form hierarchical engineered biochar, which
459	exhibited a high specific capacitance of 348,000 F/kg in a 6 M KOH electrolyte.
460	Wang et al. [103] synthesized a N and P co-doped engineered biochar material

401	embedded with bimetallic phosphide (FeCoP <sub>2</sub> $@$ NPPC), via a sol-gel and
462	carbonization (N <sub>2</sub> , 900 $^{\circ}$ C) process using sustainable N- and P-enriched chitosan and
463	transition metal salts as precursors. FeCoP2@NPPC had good catalytic activities in
464	HERs, oxygen evolution reactions and overall water splitting. It requires low
465	overpotential between 114 to 150 mV at a current density of 10 mA/cm <sup>2</sup> for HERs in
466	$0.5 M H_2 SO_4$ and $1.0 M KOH$ , while the overpotential of 236 mV for OER in 1.0 M
467	KOH solution. The FeCoP <sub>2</sub> @NPPC material was capable of transferring electrons and
468	electrolytes and increasing the contact area of the active sites and electrolytes. More
469	importantly, the wettability and conductivity of engineered biochar can be improved
470	by doping N- and P-containing functionalities.
471	Currently, viable methods in developing carbon materials from food waste for
471 472	Currently, viable methods in developing carbon materials from food waste for electrochemical applications are still in an early phase of development. The diversity
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471 472 473 474 475 476 477	Currently, viable methods in developing carbon materials from food waste for electrochemical applications are still in an early phase of development. The diversity of food waste and the high cost of synthesis processes significantly limit the large- scale application of waste conversion technologies. Therefore, achieving full valorization of food waste into biofuels, chemicals, and high value-added carbon materials remains an important challenge requiring further investigation. In addition, techno-economic and life-cycle assessments of the integrated processes based on the
471 472 473 474 475 476 477 478	Currently, viable methods in developing carbon materials from food waste for electrochemical applications are still in an early phase of development. The diversity of food waste and the high cost of synthesis processes significantly limit the large- scale application of waste conversion technologies. Therefore, achieving full valorization of food waste into biofuels, chemicals, and high value-added carbon materials remains an important challenge requiring further investigation. In addition, techno-economic and life-cycle assessments of the integrated processes based on the characteristics of various food wastes are necessary to meet the industrial

481 '	Table 2. Examples of food	waste-derived engineered	biochar materials for	electrochemical applications
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Feedstock	Preparation method	Engineered biochar properties	Electrochemical application	Electrochemical performance	Ref.
Rice	KOH activation with Bacillus cereus (N precursor)N-doped micro- and meso- porous structure with high 		Li-ion batteries (LIBs), Na-ion batteries (SIBs)	Capacity: 1,006 mA h/g in LIBs at 100 mA/g (100 cycles) and 169 mA h/g in SIBs at 100 mA/g (1,000 cycles).	[104 ]
Starch	Pyrolysis (Ar, 500 °C) combined with calcination (Ar, 800- 1,000 °C)	Hierarchical engineered biochar with high specific surface area $(2,200 \text{ m}^2/\text{g})$ and controlled porosity	Supercapacitor	Specific capacitance (229 F/g at 1 A/g in a 6 <i>M</i> KOH electrolyte), rate capability (211 F/g at eve 10 A/g) and cycling stability (94% capacitance retention after 10,000 cycles at 2 A/g).	
Potato peel	el Carbonization with sodium hypophosphite and thioacetamide combined with KOH activation $(N_2, 750 \ ^{\circ}C)$ S- and P-doped hierarchical engineered biochar with high specific surface area $(1,911.5 \ m^2/g)$ and controlled porosity		Supercapacitor	Specific capacitance of 323 F/g at 1 A/g, acceptable rate capability and cycling stability after 5,000 cycles	[106 ]
Apple waste	Hydrothermal carbonization (HTC) (200 °C) combined with KOH activation (CO <sub>2</sub> , 800 °C)	Hierarchical engineered biochar with high specific surface area (2,000 m <sup>2</sup> /g)	Supercapacitor	Specific capacitances: 260-290 F/g	[107 ]
Orange peel	HTC (150 °C) combined with KOH activation (800 °C)	N-doped engineered biochar and intercrossed, interlinked urchin-like NiCo <sub>2</sub> O <sub>4</sub> @3DNF framework	Supercapacitor	High capacitance of 1,300 F/g at a current density of 1 A/g, excellent rate and cyclic performance of 3,500 cycles.	[108 ]
Banana peel	One-step carbonization (Ar, 800 °C) and subsequent mixing with MoS <sub>2</sub>	$MoS_2$ decorated engineered biochar composite with specific surface area of 269 $m^2/g$ and suitable micropores and mesopores.	Hydrogen evolution reaction (HER)	High HER activity and excellent stability in the acid solution, including an overpotential of 150 mV at a current density of -10 mA/cm <sup>2</sup> and a Tafel slope of 51 mV/dec.	[109 ]

Sugarcane bagasse	HTC (240 °C) combined with KOH activation (800 °C)	Oxygen-rich hierarchical engineered biochar with the specific surface area of 1,436.7 m <sup>2/</sup> g and highly microporous.	Supercapacitor	The specific capacitance of 185.5 F/g and 150.7 F/g at a current density of 0.5and 20 A/g, respectively, and superior cycling performance with 96% capacitance retention after 10,000 cycles at a current density of 10 A/g	[110]
Watermelon peel	Carbonization combined with KOH activation	N-doped hierarchical engineered biochar with the specific surface area of 782.69 m <sup>2</sup> /g and pore volume of 0.44 cm <sup>3</sup> /g	Oxygen reduction reaction (ORR) for Zn-air battery (ZAB)	High specific capacity of 748.9 mA/h g and energy density of 891.2 W/h kg, close to those of the ZAB based on Pt/C	[111]
Coffee grounds	One-step carbonization/ activation with ZnCl <sub>2</sub> (600-900 °C)	N-doped hierarchical engineered biochar with the specific surface area of $1,200 \text{ m}^2/\text{g}$	ORR for ZAB	The half-wave potential was ~870 mV versus Reversible Hydrogen Electrode comparable to the best results reported for metal-free carbon-based materials in alkaline media	[112]
Coffee grounds	One-step carbonization/ activation with KOH (1,200 °C)	Hierarchical engineered biochar nanosheet with the specific surface area of 1,945.7 m <sup>2</sup> /g	Supercapacitor	Specific energy of 35.4 Wh/kg at 11250 W/kg and 23 Wh/kg for a 3 s charge-discharge current rate, corresponding to a specific power of 30,000 W/kg and good cyclic performance over 5,000 cycles	[113]
Coffee grounds	FeCl <sub>3</sub> catalytic carbonization combined with KOH activation	Hierarchical engineered biochar with high specific surface area $(3,549 \text{ m}^2/\text{ g})$ and large meso-/macropore volume $(1.64 \text{ cm}^3/\text{g})$	Supercapacitor	The high capacitance of 440 F/g at 0.5 A/g	[114]
Shrimp shell	One-step pyrolysis (Ar, 700 °C) with KOH	N-doped hierarchical engineered biochar with the specific surface area of $2,032 \text{ m}^2/\text{g}$	Supercapacitor	The specific capacitance of 348 F/g in a 6 <i>M</i> KOH electrolyte	[101 ]
Shrimp shell	One-step pyrolysis (Ar, 800 °C) with KOH	Hierarchical engineered biochar with a high specific surface area (1,271 m <sup>2</sup> /g), O-doping (9.08%) and N-	LIBs, supercapacitor	As LIB anode material: specific capacity as high as 1,507 mA h/g and 1,014 mA h/g at current densities of 0.1 A/g and 0.5 A/g, respectively, good rate performance and superior cycling	[102 ]

		doping (2.86%)		stability; as engineered biochar-based supercapacitor: specific capacitance of 239 F/g at a current density of 0.5 A/g in 6 $M$ KOH electrolyte.	
Shrimp shell	A sol-gel and carbonization (N <sub>2</sub> , 900 °C) process	FeCoP <sub>2</sub> nanoparticles embedded in N- and P- doped hierarchical engineered biochar	ORR, HER for water splitting	A low voltage of 1.60 V to deliver a current density of 10 mA $/cm^2$	[103 ]
Crab shell	One-step pyrolysis (900 °C)	N- and S-doped hierarchical engineered biochar with the specific surface area of 827.5 m <sup>2</sup> /g	ORR	ORR activity with an onset potential of 0.072 V and a half-potential of 0.110 V (vs. Ag/AgCl) in alkaline electrolytes	[115]
Fish	Pyrolysis with ZnCl <sub>2</sub> (900 °C)	N-coated hierarchical engineered biochar with the specific surface area of 850 $m^{2}/g$	ORR	Half-potential of 0.060 V lower than that on 20 wt.% Pt/C catalyst	[116]
Fishbone	One-step carbonization (850 °C) without chemical activator	N- and S-doped hierarchical engineered biochar with the specific surface area of 1,337 m <sup>2</sup> /g	Supercapacitor	Specific capacitance of 476 F/g and high cycling stability with 90.9% of the original capacitance retained after 10,000 cycles at a 1 A/g current density	[100 ]

#### 483 5. Conclusions and outlook

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Thermochemical conversions are a promising route for upcycling food waste into high 484 485 value-added fuels and advanced carbon materials due to the short residence times 486 required, high yields of a wide range of products, high conversion efficiencies and feasible deployment for practical applications. Numerous research focused on 487 optimizing thermochemical reaction conditions with controllable product distributions 488 489 and elucidation of the reaction mechanisms and evolution pathways. The production of food waste-derived gaseous and liquid products via thermochemical conversions 490 has been widely explored elsewhere. This review summarizes the processes of 491 producing biochar and hydrochar via the slow pyrolysis and HTC of food waste and 492 493 their by-products. Engineered biochar synthesis through activation (i.e., physical and chemical) and surface modifications (i.e., O-, N-, and S-doping) can be further 494 495 optimized using ML approaches. Moreover, engineered biochar-based practical applications, such as solid fuels, adsorbents (i.e., CO<sub>2</sub> adsorption), and advanced 496 catalytic conversions (i.e., electrochemical catalysis), were thoroughly reviewed. 497

Food waste is a promising and sustainable alternative to solid fuels. It is often characterized by high-water content. HTC enables the direct conversion of food waste 499 500 into hydrochar for solid fuel applications. HTC operating conditions (i.e., temperature, pressure, and residence time) significantly affect the physicochemical properties, 501 combustion behavior, and alkali metals contents of the resulting hydrochar. Compared 502 with other biomass waste, such as agroforestry residues, food waste has a lower ash 503

content, especially of alkali and alkaline earth minerals, which is beneficial for
reducing both sintering and corrosive risks during thermochemical conversions.
However, because of the high moisture content, developing a low-cost and efficient
dewatering pre-treatment remains a key challenge. The co-production of bio-oil fuels
(from waste cooking oil) and biogas (from AD pre-treatment) dramatically increases
both the technical and economic feasibility of thermochemical upcycling.

510 Biochar and hydrochar can be activated and/or modified into advanced engineered biochar materials for energy and environmental applications. Owing to its stable 511 physicochemical properties, tunable pore structures, effective surface functional 512 groups and cost-effectiveness, engineered biochar has been widely synthesized into 513 advanced carbon materials for adsorption and electrochemical applications. In the 514 context of carbon neutrality, paradigm shifts to interdisciplinary collaboration are 515 516 urgently needed to achieve the circular economy of food waste. For example, material science, data science, thermochemical engineering, and environmental engineering are 517 518 the basic requirements for developing an advanced CO<sub>2</sub> adsorption system using food waste-derived engineered biochar. Moreover, the wide range of feedstocks and lack of 519 520 synthesis guidelines for engineered biochar greatly limit its commercial applications. 521 A powerful ML approach has been considered a promising route for synthesizing 522 high-performance engineered biochar. ML can be used not only to accurately predict 523 practical performance using engineered biochar but also to efficiently provide inverse 524 design guidelines to synthesis engineered biochar for a specific application. In addition, both environmental benefits and economic feasibility need to be verified 525

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527 engineered biochar can be synthesized and applied on an industrial scale.

528	To accelerate the practical applications of food waste-derived engineered biochar
529	materials, several efforts need to be made in the future as follows:
530	(1) AD is still the dominant pre-treatment for sustainable food waste management into
531	methane and solid residue, owing to that it has a better techno-economic feasibility
532	compared with the direct thermochemical processes (e.g., pyrolysis, gasification).
533	However, it is still necessary to perform thermochemical routes for further upcycling
534	solid residue into high value-added engineered biochar materials. In the future, more
535	efforts should be made in the integrated AD and follow-up thermochemical routes for
536	full valorization of food wastes.
537	(2) For the application of food waste-derived engineered biochar as an adsorbent,
538	most of the current research has focused on the conventional pollutants removal (e.g.,
539	VOCs adsorption, CO2 capture). More efforts need to be made in the development of
540	food waste-derived engineered biochar with functionalized properties, in order to
541	adsorb and convert emerging contaminants into value-added products, simultaneously.
542	(3) Many investigations have been performed in the conversion of diverse food waste
543	to biochar materials for solid biofuel productions or carbon material applications, but
544	these studies mainly focused on the optimization of processing and utilizing
545	conditions. To date, less work has been done in the techno-economic and life-cycle
546	assessment of the whole process including biofuels and carbon materials production
547	and applications, which is beneficial to promoting the industrial applications of food 30

548 waste-derived value-added products.

549

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