Engineered hydrochar production methodologies, key factors influencing agriculture wastewater treatment, and life cycle analysis: A critical review

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Abstract. For intensive food production, a range of chemical compounds are used to increase production, reduce the amount of weeds, and prevent pest infestation. Therefore, agricultural wastewater discharge to water bodies creates human health and environmental risks. This highlights the need for technologies to remove organic and inorganic pollutants, where adsorption using carbon-based materials has emerged as a cost-effective and promising alternative for agricultural wastewater treatment with high removal efficacy and alignment with the circular economy concept by generating value-added products, achieving energy conservation and reducing the environmental footprint. Among the different adsorbent materials, hydrochar (HC) has attracted attention because, compared to the thermal processes used for synthesizing other carbon-based materials, it requires relatively milder production conditions and possesses higher adsorption capability for water pollutants. Although HC holds advantages for the adsorption of water pollutants, HC modification using different methods has been found to improve the properties, including adsorption capacity. Accordingly, engineered hydrochar (EHC) has attracted research attention. However, past research publications show that the investigations have focused on material characterization and removal rates, with little knowledge created of the environmental impacts of HC production, application, and disposal. This study reviews current knowledge on EHC synthesis, characteristics, water treatment applications, and life cycle analysis. Initially, production methodologies were investigated to understand their influence on key surface physical and chemical characteristics. This was followed by assessing EHC efficacy for water and wastewater treatment. Finally, the environmental footprint of EHC production, application, and disposal was evaluated to identify critical knowledge gaps and to provide recommendations for future research.

Keywords
Carbon-based materials; Hydrochar; Synthesis; Pollutant adsorption; Water treatment; Life cycle analysis.

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1. INTRODUCTION

Agricultural production is relevant to the current water crisis because of significant freshwater consumption [1] and the resulting wastewater containing diverse organic and inorganic pollutants [2–6]. Developing effective agricultural wastewater treatment technologies, therefore, meets a significant need to ensure water security [5,7,8]. Different agricultural wastewater treatment technologies have been proposed, among which adsorption using engineered materials has gained interest because of its low cost and ease of use [2,9–12]. Despite high pollutant removal efficiency, some adsorbent materials (e.g., carbon nanotubes) possess a significant environmental footprint generating concerns about their sustainable full-scale production and application [13–18]. To avoid these undesirable indirect consequences, adsorbent production should be aligned with the circular economy concept, creating value-added products, extending the life cycle for achieving energy conservation and reducing the environmental footprint in line with Sustainable Development Goal 6 (SDG6) (clean water and sanitation) and other SDGs related to sustainable and clean production, and waste valorization [19–22].

Some carbon-based materials (CBMs) have gained attention because their production is based on using waste biomass as feedstock, requires relatively mild production conditions [19,23–26], and generates key properties which play major roles influencing their effectiveness in environmental applications [27–31]. Hydrochar (HC) is a CBM produced using hydrothermal carbonization (HTC) that requires mild temperature conditions (180-250°C) compared to other thermal processes used for CBM production [32–34]. HC is reported to have the ability to adsorb organic and inorganic contaminants. This suggests its high potential for use as an adsorbent [35–37]. Compared with other CBMs, HC possesses relatively low porosity and specific surface area, showing seemingly lower pollutant adsorption rates [38,39]. However, HC can be activated/modified [40] to improve its properties for different applications (e.g. water treatment, carbon capture, catalysis, and energy storage) [19,41]. The activated/modified HC, usually termed engineered HC (EHC), has been found to hold advantages for the adsorption of water pollutants. Most past studies which have focused on EHC have been devoted mainly to material characterization and pollutant adsorption rates with little information available on the environmental impacts and life cycle of production/application [42].

In this study, state-of-the-art methodologies for EHC production were investigated to assess its environmental footprint and its fit to the circular economy concept. The key physical and chemical characteristics of EHCs were evaluated to assess their effectiveness for agricultural wastewater treatment applications. Further, the life cycle analysis (LCA) of different EHCs was investigated to understand the current state of knowledge, identify critical knowledge gaps, and provide recommendations for future research.

2. HC production processes

HC is generated from biomass using HTC in the 180-300 °C temperature range and autogenous pressure (2-16 MPa) [43,44]. Cellulose- or hemicellulose-related biomass is the most commonly reported feedstock. HTC has been suggested to break down biomass through polysaccharide hydrolysis into smaller oligosaccharides and monosaccharides (e.g., glucose and xylose) [45]. Subcritical water conditions in HTC convert monosaccharides into intermediate organic compounds through isomerization, dehydration, fragmentation, and

condensation reactions (Figure 1). Glucose and xylose isomerization reactions produce fructose and lyxose [46], while its decomposition leads to organic acids (e.g., acetic, lactic, propionic, levulinic, and formic). Glucose dehydration leads to furfural-related structures which decompose in aldehydes, phenols, and carboxylic acids [47]. Aldol condensation and intermolecular dehydration produce furfural condensation and polymerization [47] and, subsequently, HC [48–50] with combined polyaromatic/polyfuran structure [51].

The influence of feedstock composition on HC chemical and physical characteristics has been explained using HTC biomass transformation pathways [52] [20,53,54]. For example, the production of homogeneous HC from solid waste, a freely available and low-cost feedstock has been identified as having significant drawbacks related to the presence of biomass with mixed long- and short-chain organic structures [19,20]. To produce homogenous HC, biological biomass reduction has been suggested [55–57]. Nevertheless, only a few studies have focused on understanding the role of feedstock chemical and physical properties on HC characteristics, which constitutes a significant knowledge gap that merits further attention. This is because an in-depth understanding of the influence of feedstock on HC characteristics will allow the selection of the most suitable biomass for a specific application and for identifying feedstock modifications to optimize HC performance.

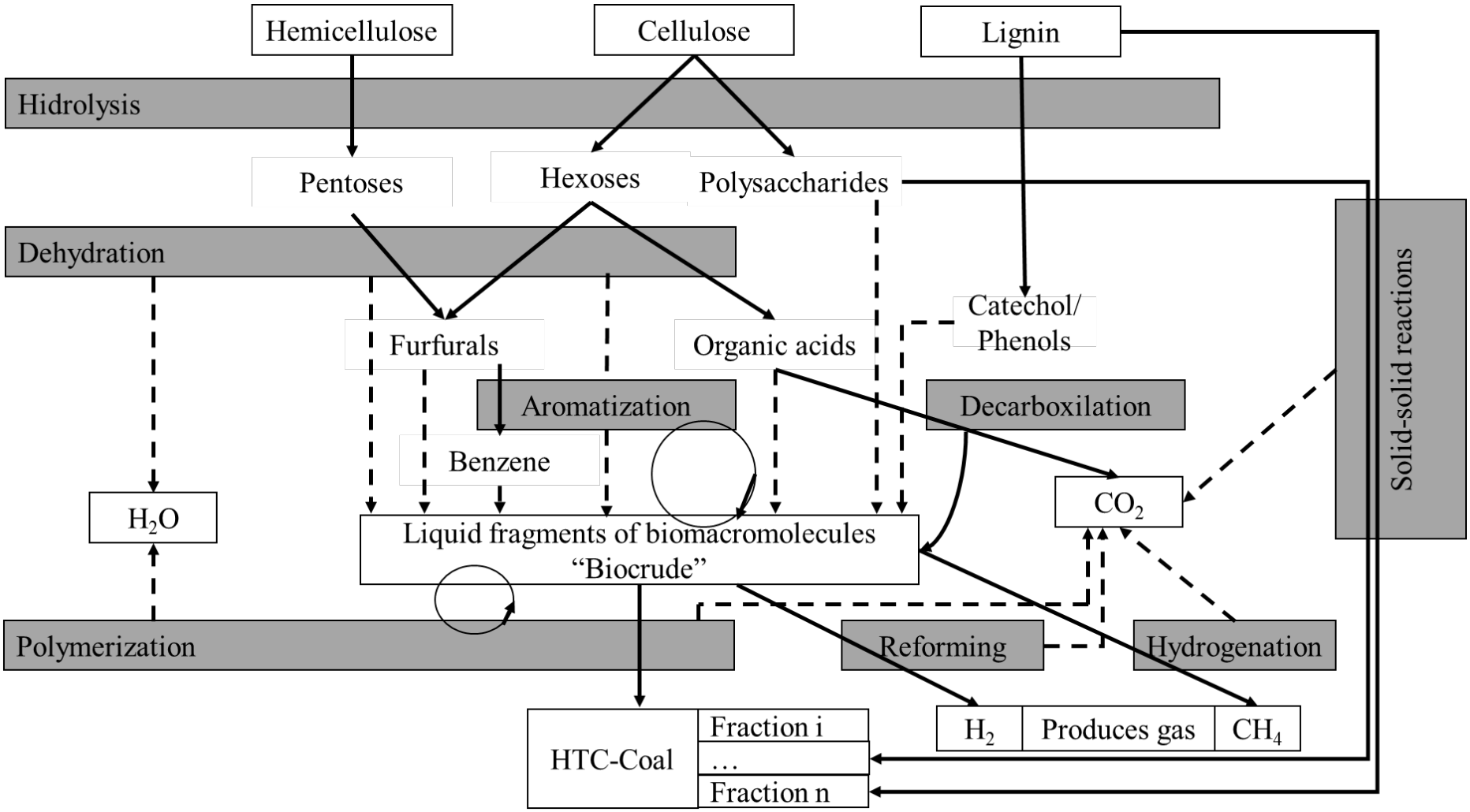

Figure 1. Schematic of HC formation mechanisms (adapted from Kruse et al., 2013) [53].

3. Modification of HC characteristics

Controlling HC characteristics by limiting feedstock type may involve significant difficulties because not all biomass would be appropriate or its cost may limit feedstock usage. Therefore, identifying abundant, low-cost, and reliable biomass is necessary for efficient HC production. In addition, suitable in-situ or post-production modifications should be considered when the resulting product lacks requisite performance characteristics.

The modification of HC properties by changing carbonization temperature and residence time has been reported to have generated materials lacking adsorption capacities to compete with other CBMs. Therefore, further modification processes (e.g., alkali or acid activation) to produce EHCs have been explored to enhance the adsorption of pollutants in water [19,20,58–60]. For example, alkali activation with KOH has been found to enhance the Pb^{2+} adsorption capability of EHC over five times compared to unmodified HC [35].

EHC is prepared using two main pathways, namely, in-situ treatment and post-treatment. For in-situ HC modification, acid or alkaline solutions are added to the biomass before HTC to promote specific reaction pathways [61]. Post-production modifications involve the addition of chemical agent(s) to change the surface characteristics after HTC. A deeper analysis of how HC production process modification impacts adsorption characteristics is discussed in the following sections.

3.1. Acid/alkaline modification

In-situ modifications. Adding acid or alkaline solutions before HTC accelerates polysaccharide degradation (glucose and furfural production), enhancing HC adsorptive properties [36,59,62]. When acidic conditions are used, protic acids catalyze hydrolysis through nucleophilic substitution accelerating cellulose transformation to glucose [63]. After hydrolysis, dehydration reduces organic molecules (Figure 1), generating EHC through different pathways. Identifying specific acidic conditions (e.g., acid type and concentration) to limit uncontrolled organic reactions during HC synthesis merits in-depth investigation because it could enable the production of HC with tailored chemical and physical characteristics appropriate for specific applications [64,65]. Very few studies, however, have reported the use of acidic or alkaline solutions as solvents for HTC (Table 1). A more in-depth understanding of the benefits of in-situ acidic or alkaline modifications before HTC is a pending research task that may help to reduce EHC production time and/or improve material homogeneity and performance reliability.

Alkaline conditions lead to saponification during hydrolysis, transforming esters into alcoholic and/or carboxylic groups and reducing EHC functional groups [66]. Alkali counter ions (e.g., Na^+, K^+) have been reported to produce carboxylate groups on the EHC surface and serve as cation exchange sites [67,68]. However, there is a paucity of information available on alkaline in-situ modification (Table 1), which is another knowledge gap worthy of attention because it would allow the adoption of alternative EHC production pathways to achieve enhanced properties for advanced applications in water treatment.

Table 1. In-situ modifications for EHC production.

Modification type	Reagent	Feedstock	Thermal treatment	Reference
Acid treatment	N-cyclohexyl sulfamic acid	Sawdust	190°C; 12 h; O_2	[36]
Acid treatment	HCl	Small world rabbit food	250°C; 20 h; O_2	[62]
Alkaline treatment	NaOH	Small world rabbit food	250°C; 20 h; O_2	[62]
Functional group modification	Ammonium sulfate	Sucrose	200°C; 4 h; O_2	[40]
Functional group modification	Triethylenetetramine	Glucose	190°C; 48 h; O_2	[69]
Metal immobilization	Fe	Pinewood sawdust	200°C; 1 h; O_2	[70]
Metal immobilization	Fe	Pinewood powders	180°C; 20 h; 10°C min^{-1}; O_2	[71]
Metal immobilization	Fe	Sewage sludge	180°C; 3 h; O_2	[56]
Metal immobilization	Fe	Rice straw	200°C; 3h; O_2	[58]
Metal immobilization	La	Rice straw	200°C; 4-14h; O_2	[72]
Metal immobilization	Ca	Wood powder	200°C; 12 h; O_2	[73]
Metal immobilization	Ca	Pinewood sawdust	220°C; 4 h; O_2	[74]
Metal immobilization	Si/Mg	Pine sawdust	180°C; 12 h; O_2	[75]
Functional-coated	Ti_3AlCl_2	Glucose Cellulose Pinewood sawdust	180°C; 48 h	[76]
Functional-coated	Montmorillonite	Microcrystalline cellulose	200°C; 2-24 h; autogenic pressure; O_2	[77]
Functional-coated	Zero valent iron	Alkali lignin	200°C; 18 h	[78]

Post-production modifications. Table 2 shows the different post-production methods reported using alkali, acidic, or oxidant solutions. Alkali post-production activation is the most frequently used method, particularly potassium hydroxide because it is well-known for creating oxygen-containing surface functional groups on EHC and substituting hydrogen in carboxylic groups with potassium [79]. In Table 3, significant surface area and porosity increases are reported for post-production treatment with enhanced HC properties depending on the synthesis method adopted. Acidic activation consistently produces a higher surface area (ca. two-fold) compared to alkaline activation or unmodified material (Table 3). However, only a limited number of studies were found which have reported using acidic HC activation [34]. There is a significant need to understand the benefits and limitations of alkali and acidic HC activation, which is considered a pending research task that merits further exploration because it can provide essential knowledge for improved EHC synthesis methods and pathways for customized materials for water treatment.

Table 2. Acidic/alkaline treatment and chemical oxidation for HC modifications.

Chemical treatment	Feedstock	Chemical	Chemical activation	Secondary heat treatment	Reference
Alkaline	Rattan	NaOH	HC/NaOH mass ratio 3:4	600°C; 1 h; 10°C min^{-1}; N_2	[80]
Alkaline	Sewage sludge	KOH	HC/KOH mass ratio 0.5	650°C; 2.5 h; N_2 atmosphere	[55]
Alkaline	Sawdust, wheat straw, cornstalk	KOH	HC: 4 gL^{-1}; [KOH]: 2 N	None	[81]
Alkaline	Grape pomace	KOH	HC:10 gL^{-1}; [KOH]:2 M	None	[35]
Alkaline	Commercial sucrose	KOH	KOH/HC mass ratio 1:1-4:1	N_2; 300-800°C; 2 h; 10°C min^{-1}	[79]
Alkaline	Corn cobs	KOH	1:10 HC/KOH; [KOH]:3 M	230, 260°C; 0.5 h; O_2	[82]
Alkaline	Rice Husk	KOH NaOH	1:3 – 1:6 KOH/HC	800°C; 1 h; N_2	[83]
Alkaline	α-D-glucose	$KHCO_3$	4,6,8 $KHCO_3$/HC ratio	N_2; 850°C; 1-5h; 5°C min^{-1}	[84]
Alkaline	Sucrose	KOH	1:0.25-1:3 HC/KOH	800°C; 2 h; 5°C min^{-1}; N_2	[85]
Alkaline	Hickory wood Peanut hull	KOH	1:1 KOH/HC; 50% KOH	600°C; 1 h; N_2	[34]
Alkaline	Garlic peel	KOH	1:2 HC/KOH ratio	600-800°C; 1 h; 2°C min^{-1}; N_2	[86]
Acidic	Orange peels D-glucose	HNO_3	33.3 gL^{-1} HC; 30, 50,70% HNO_3	None	[87]
Acidic	Rice Husk	H_3PO_4	1:4,1:6 H_3PO_4/HC ratio; 85% H_3PO_4	800°C; 1 h; N_2	[83]
Acidic	Hickory wood Peanut hull	H_3PO_4	1:1 H_3PO_4/HC ratio; 85% H_3PO_4	600°C; 1 h; N_2	[34]
Oxidant	Sawdust	H_2O_2	10 g HC in 300 mL 20%H_2O_2	None	[88]
Oxidant	Peanut hull	H_2O_2	10 g HC in 300 mL 20%H_2O_2	None	[89]

Table 3. Physical characteristics of engineered HC from post-treatment modifications.

Modification	Feedstock	Surface area, m^2 g^{-1}		Porosity, cm^3 g^{-1}		Reference
		Unmodified	Modified	Unmodified	Modified	
Alkaline/KOH	Garlic peel	306	1,262	0.17	0.70	[86]
Alkaline/KOH	Sucrose	33	2,604	0.027	1.69	[85]
Alkaline/KOH	α-D-glucose	525	3,400	0.22	2.40	[84]
Alkaline/KOH	Sawdust	4.4	0.69	0.013	0.002	[81]
	Wheat straw	9.1	0.42	0.041	0.003	
	Cornstalk	8.6	1.84	0.034	0.006	
Alkaline/KOH	Glucose	75	1,197	0.1	0.74	[90]
Alkaline/KOH	Corn cobs	2.8	5.24	0.005	0.011	[82]
Alkaline/KOH	Commercial sucrose	33	2,604	0.027	1.44	[79]
Alkaline /$KHCO_3$	α-D-glucose	525	3,050	0.22	2.10	[84]
Alkaline /K_2CO_3	Salix psammophila	6.9	1,230	0.041	0.810	[91]
Alkaline /K_2CO_3	Rice straw derived	6.22	1,334	0.06	1.07	[92]
Alkaline/KOH	Hickory wood	8	222	0.121	0.05	[34]
	Peanut hull	7	571	0.01	0.075	
Acid/H_3PO_4	Hickory wood	8	1,436	0.121	0.028	[34]
	Peanut hull	7	1,091	0.01	0.079	
Acid/HNO_3	Orange peels	34.1	20.25	0.047	0.042	[87]
	D-glucose	7.3	4.49	0.012	0.012	
Functional group/Protonate amine	Bamboo	26.2	11.76	0.089	0.026	[93]
Functional group/Melamine	Glucose	75	148	0.1	0.15	[90]
Functional group/Triethylenetetramine	Glucose	335	313	0.217	0.144	[69]
Metal immobilization/Fe	Salix psammophila	7.16	349	0.006	0.24	[94]
Metal immobilization/Fe-Ni	Salix psammophila	7.16	515-1,351	0.004	0.315-0.549	[95]
Metal immobilization/Fe	Salix psammophila	6.86	673	0.041	0.646	[91]
Metal immobilization/Fe	Coffee waste	17.2	34.7	0.59	0.13	[30]

Metal immobilization/Fe	Rice straw derived	6.22	674	0.06	0.72	[92]
Metal immobilization	Rice Husk	44.47	167.17	0.019	0.065	[96]
Functionalized particles	Sawdust	1.52	13.66	0.004	0.043	[97]
Functionalized particles	Sawdust	1.52	2.97	0.004	0.027	[97]
Functionalized particles	Sawdust	1.52	3.73	0004	1.348	[97]
Biological modification	Sawdust	3.24	10.33	-	-	[98]

3.2. Modification of functional groups

In-situ modifications. The addition of amino-functional groups has been reported to enhance HC's chemical and physical characteristics [19,59,99]. However, as shown in Table 1, only a few studies have investigated amine HC modification. Ammonium sulfate has been used before HTC, impacting long-chain, liquid by-products (e.g., furfural, 5-methyl furfural) synthesis pathways and the resulting performance was compared with experiments without ammonium sulfate addition [40]. Acetic acid was the only HTC by-product identified when ammonium sulfate was used for HC modification, suggesting changes in the production pathway. No information is available about by-product generation using other production conditions. This is considered a significant knowledge gap that merits further research to better understand the effect of nitrogen compounds on the EHC production pathway and the enhancement of HC adsorption abilities.

Post-production modifications. A two-step post-thermal modification process for EHC has been suggested using acidic or alkaline activation to increase hydroxyl functional groups, followed by amino functionalization to increase nitrogen content [100]. This lack of information is considered an interesting research area worthy of attention to better understand the influence of different chemical modifications for producing EHC with specific functional groups and allowing the addition of amine groups leading to the reduction of synthesis steps.

3.3. Metal functionalized particles

In situ modifications. Few studies are available on variations of chemical and/or physical characteristics after metals addition before HTC (Table 1). Iron-induced polymerization and re-arrangement have been found to activate functional groups on the HC surface [58]. Calcium was found to generate side-chain lignin, ester bond cleavage, and carboxyl group deprotonation when added as HTC alkaline treatment [74]. Little is known about the effects of other metallic elements (e.g., other transition metals, metalloids, rare earths) on HC production. The limited information available suggests that performance improvements could be achieved using single or hybrid metallic-modified EHCs for adsorbing organic pollutants and heavy metals [101]. Accordingly, understanding in-situ HC modification with metals is a pending research task that merits further investigation to identify alternative production pathways and to create EHC for fit-for-purpose applications.

Post-production modifications. Iron-impregnated EHC has been investigated to understand the effect of iron concentration (e.g., 33 vs. 77% v/v Fe) on adsorption performance [48]. Post-production modifications were found to generate relatively higher surface area (e.g., 337 vs. 794 m^2 g^{-1}) and pore volume (i.e., 0.1593 vs. 0.2547 cm^3 g^{-1}) compared to in-situ modification. In addition, studies have reported iron association with oxygen-containing functional groups during HTC leading to improved EHC dispersion. After post-production treatment, Fe ions were found attached to hydrophilic groups by electrostatic attraction within the outer hydration shell. However, no studies are available comparing engineered and non-engineered materials which inhibits the evaluation of different impregnation pathways or understanding chemical and physical characteristics in the resulting EHC. As shown in Table 4, only iron has been investigated and information concerning other metals and metallic ions remains unavailable. This is another significant knowledge gap that merits attention as metallic elements are reported to improve the performance of other CBMs for different applications [102–104].

Using HC to prevent agglomeration and functionalized particle recovery has been reported in past studies [103,105,106]. For example, attaching zero-valent metals (e.g., Fe/Ni) to HC is reported to help prevent oxidation [107,108] and particle agglomeration [109]. However, very few studies are available where HC has been used to support zero-valent metals (Fe/Ni) [109], clays [97], or semiconductors (Ag_3PO_4) [110]. This knowledge, if available, would help to understand the interactions between HC and functionalized particles to reduce the material's limitations (e.g., agglomeration) or generate other properties that may lead to enhanced adsorption ability.

Table 4. Immobilization of metal composites.

Feedstock	Metal	Magnetic or non-magnetic	Second thermal treatment	Reference
Rice Husk	Ni	Non-magnetic	800° C; 1 h; O_2	[104]
Pinewood sawdust	Fe	Non-magnetic	600°C; 1 h; N_2 (200 mL min^{-1})	[70]
Rice husk	Fe/Mn	Non-magnetic	-	[96]
Rice straw derived	Fe	Magnetic	1.5 h; N_2 (1 L min^{-1})	[92]
Sugarcane bagasse	Fe	Magnetic	-	[111]
Salix psammophila	Fe	Magnetic	700°C; 2 h; N_2 (1 L min^{-1})	[94]
Salix psammophila	Fe/Ni	Magnetic	500-800°C; 1.5 h; N_2 (1 L min^{-1})	[95]
Salix psammophila	Fe	Magnetic	70°C; 3 h; N_2	[91]
Orange peel	Fe	Magnetic	-	[112]
Coffee waste	Fe	Magnetic	-	[30]

3.4. Other engineered modifications

HC production involves long processing times and uneven heating patterns [113]. Alternative heat treatment (e.g., microwave radiation) has received increasing attention because it is faster and can provide uniform heat transfer [114,115]. Microwave radiation generates selective HC autogenic reactions (i.e., higher acetyl rupturing), which enables the fine-tuning of EHC characteristics [115] at lower production cost [115,116]. No detailed cost analysis has been reported, and the technology readiness level (TRL) remains low preventing fair comparison with other technologies capable of full-scale HC production. This is considered a significant knowledge gap requiring further attention. Microwave-assisted HC production has also been reported pointing towards an interesting future research avenue for EHC production.

Using microorganisms to digest organic matter on HC surface is reported to increase surface area [98] through alkane conversion via anaerobic digestion. The process has been reported to change HC surface charge and hydrophobicity, suggesting that chemical activation modifications could also be achieved using biological treatment [57]. This approach provides the potential to avoid chemical usage, significantly reducing the carbon footprint of the process. Consequently, HC production using biological modifications is a highly interesting research avenue requiring further exploration, as minimal research has been undertaken to date.

4. EHC adsorption applications

4.1 Heavy metal removal.

Heavy metal adsorption using EHC for water treatment (Table 5) is reported as highly cost-effective. For example, iron-modified EHC has been shown to have high adsorption capacity for Pb and Cd (Q_{max} = 417 and 323 mg g^{-1}, respectively) [109,111]. However, these studies have used differing experimental conditions preventing fair comparison. The persistent lack of systematic studies with comparable conditions is another significant knowledge gap which merits attention because reporting on HTC conditions and feedstock used in a study is essential to identify specific influences exerted by key variables on EHC characteristics and to assess pollutant adsorption ability and selectivity.

Table 5. Efficiency of engineered hydrochar for heavy metal removal.

Modification	Contaminant	Removal, %	Notes	Reference
Acid/N-cyclohexyl sulfamic acid	Cu	100	$[Cu]_0$:0-100 mg L^{-1}; [HC]:0.5 g L^{-1}; pH:2-9	[36]
Alkaline/KOH	Cd	20 – 100	$[Cd]_0$:5-300 mg L^{-1}; [HC]: 2 g L^{-1}; pH: 2-10 30º C	[81]
Alkaline/KOH	Pb	56 – 100	$[Pb]_0$: 100 mg L^{-1};[HC]: 0.4-4 g L^{-1}; pH: 2-7;25,35,45º C	[35]
Alkaline/KOH	NH_4^+	75 – 88	$[NH^{4+}]_0$:1,324 mg L^{-1};[HC]:10 g L^{-1}; pH:4.5-8.5;25, 35, 45º C	[82]
Functional groups/Triethylenetetramine	Pb Cu	0 – 100 0 – 100	$[Pb]_0$: 4 g L^{-1}; $[Cu]_0$: 4 g L^{-1};[HC]:5-1,200 mg L^{-1};pH: 5; 30º C	[69]
Functional group/Maleylated	Cd	35 – 100	$[Cd]_0$: 40-210 mg L^{-1};[HC]: 0.8 g L^{-1};pH:2-10; 20-30°C	[117]
Functional group/Polyaminocarboxylated	Cu	58 – 100	$[Cu]_0$:40-220 mg L^{-1};[HC]: 0.8 g L^{-1};30,40,50° C;pH:2-5	[118]
Functional groups/Polyethylene imine	Cr Ni	25 – 100 50 – 100	$[Cr]_0$: 20 mg L^{-1}; $[Ni]_0$: 50 mg L^{-1};[HC]: 2.5 g L^{-1};pH: 5.5	[100]
Immobilized material/Bentonite	NH_4^+	12 – 100	$[NH^{4+}]_0$: 200 mg L^{-1};[HC]: 1-10 g L^{-1};pH: 6; 30º C	[119]
Immobilized material/Ti_3AlC_2	Cd Cu	60 – 10 41 – 100	$[Cd]_0$: 5-100 mg L^{-1}; $[Cu]_0$:5-100 mg L^{-1};[HC]:1 g L^{-1}; 25°	[76]
Immobilized material/Lanthanum	P	77 – 100	$[P]_0$: 40,100 mg L^{-1};[HC]: 5-4 g L^{-1}; 25° C	[72]
Immobilized material/Fe	Pb Cd	35 – 100 32 – 100	$[Pb]_0$: 25, 400 mg L^{-1}; $[Cd]_0$: 25, 400 mg L^{-1}; [HC]: 0.4-2 g L^{-1}; pH_{Pb}: 1.8-7.1; pH_{Cd}: 2.2-8.5; 25,30,40°C	[111]
Immobilized material/ Zero valent Ni-Fe	Pb	100	$[Pb]_0$: 50-500 mg L^{-1}; [HC]:2 g L^{-1}; pH:3-6	[109]
Immobilized material/Calcite	Cu	37 – 100	$[Cu]_0$:30-300 mg L^{-1}; [HC]: 0.8 g L^{-1}; pH: 3-6.5	[73]

4.2 Adsorption of organic compounds

EHC has been reported to be primarily for dye adsorption (Table 6), where chemical modifications (acid or alkali), functional modifications, and metal immobilization were found to produce encouraging results. Nevertheless, the need for systematic studies with comparable variables (e.g., hydrochar type, dye structure) remains to be investigated to assess the influence of HC modifications and associated structural changes. The need for an in-depth understanding of the adsorption mechanisms involved [120–122] is considered a significant knowledge gap as this knowledge would help to correlate adsorption pathways with HC modifications. Once adsorption pathways are known, the processes can be influenced by specific modifications (e.g., electrostatic attraction, hydrogen bonding) [87]. Other mechanistic pathways (e.g., ion exchange) have been proposed for methyl orange adsorption using HC functionalized with quaternary ammonium groups [123]. Despite the high efficiency shown by HC for the adsorption of organic and inorganic compounds (Tables 5 and 6), little is known about the efficacy of EHC for the adsorption of emerging contaminants (Table 6). This is considered another significant knowledge gap that merits attention based on the successful use of other engineered CBMs for the adsorption of emerging contaminants [103,124].

Table 6. Efficacy of engineered hydrochar for the removal of organic contaminants.

Modification	Contaminant	Removal, %	Notes	Reference
Acid/HNO_3	Methylene blue	0 – 100	$[MB]_0$:0.1-1.0 g L^{-1};[HC]: 2 gL^{-1}; pH: 2-11; 10, 30,50º C	[87]
Acid/N-cyclohexyl sulfamic acid	Benzotriazole	85 – 100	$[BTA]_0$:0-0.1g L^{-1}; [HC]: 0.5 gL^{-1}; pH: 2-9	[36]
Alkaline/NaOH	Methylene blue	82 – 100	$[MB]_0$:25-350 mg L^{-1}; [HC]: 0.8 g L^{-1}; pH: 3-11;30º C	[80]
Functional groups/Triethylenetetramine	Acid Red 1 Methylene green 5	0 – 100 0 – 100	$[AR1]_0$:4gL^{-1};$[MG5]_0$:4gL^{-1}; [HC]:5-1,200mgL^{-1}; pH: 5;30ºC	[69]
Functional groups/Protonated amine	Methyl orange	61 – 100	$[MO]_0$: 0.3-1.2 gL^{-1}; [HC]:0.8 g L^{-1}; pH:4-12	[93]
Functional group/Maleylated	Methylene blue	70 – 100	$[MB]_0$:0.5-1.3 g L^{-1};[HC]:0.8 g L^{-1};pH:2-10;20-30°C	[117]
Functional group/Quaternary ammonium	Methyl orange	34 – 100	$[MO]_0$: 0.2-1 g L^{-1};[HC]: 0.4,1 g L^{-1}; pH:6.2-6.7; 30, 40, 50° C	[123]
Functional group/Polyaminocarboxylated	Methylene blue	83 – 100	$[MB]_0$:0.4-1.2 mg L^{-1};[HC]: 0.8 g L^{-1}; 30, 40, 50° C; pH:2-10	[118]
Immobilized material/Fe	Malachite green	50 – 100	$[MG]_0$:125,187,250 mg L^{-1}; [HC]:0.5 g L^{-1};25,35,45° C;pH: 2-12	[91]
Immobilized material/Fe-Mn	17 β-estradiol	31 – 100	$[E2]_0$:0.2-8 mg L^{-1};[HC]: 0.05 g L^{-1};pH:3-12; 28º C	[96]
Immobilized material/Fe	Triclosan	38 – 100	$[TCS]_0$:10-40 mg L^{-1}; [HC]: 50 mg L^{-1}; pH:3-10	[92]
Immobilized material/Fe	Tetracycline	31 – 100	$[TC]_0$:5-80 mg L^{-1}; [HC]: 1 g L^{1};25-43° C	[94]
Immobilized material/Fe	Triclosan	100	$[TCS]_0$:10-50 mg L^{-1};[HC]: 500mg L^{-1};pH:2-4; 30° C	[95]

5. Life cycle analysis (LCA)

LCA evaluates the environmental, social, or economic impacts associated with all stages of the life cycle of a commercial product, process or service. The process collects information that is analyzed depending on the major stages of the product/process/service life. Environmental LCA (LCA-E) follows a sequence of steps that address one or more life stages at a time, starting by setting a goal and scope and performing a life cycle inventory followed by a life cycle impact assessment (Figure 2) [125,126]. Once the inventory analysis is completed, the impact assessment focuses on classifying the environmental impacts of all the processes involved and modeling and translating these into potential impacts such as human health, ecosystem quality, climate change impacts, and resource usage (e.g., use of non-renewable energy) [127]. Figure 3 illustrates the LCA process and interactions between different steps along with different impact categories and associated impacts [128].

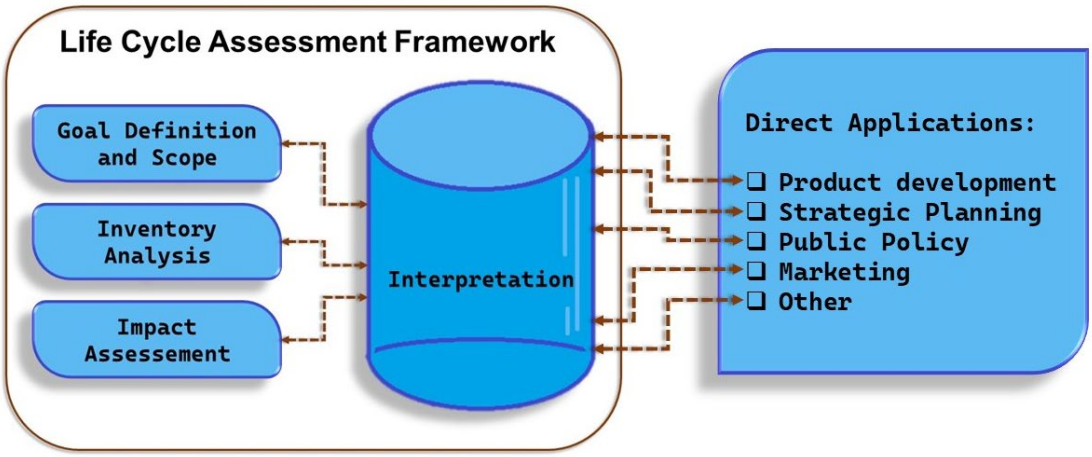

Figure 2. Steps of an LCA.

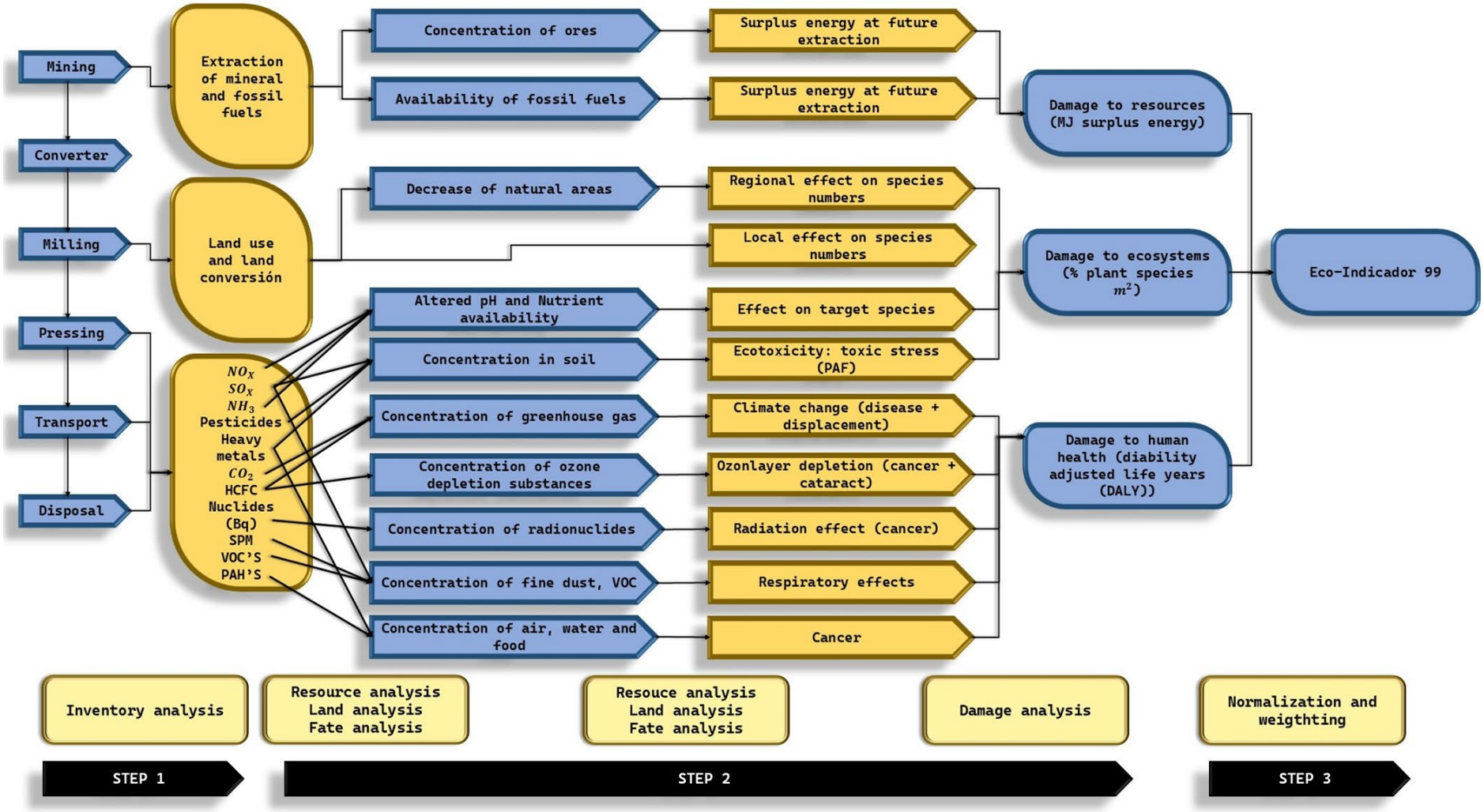

Figure 3. LCA steps and interactions and different impact categories and associated impacts.

5.1. LCA-E for EHC

To date, very few studies have been devoted to LCA-E for HC production and most of these have focused on broad goals and scope (initial LCA steps, Figure 3), assessing environmental impacts, identifying critical production parameters or comparing performance with similar materials [128–130] (Table 7). As shown in Table 7, only one study was found on LCA-E for EHC where acetic acid was used as a catalyst for EHC production [131]. Understanding the environmental impacts generated by EHC and unmodified HC production for different boundary conditions (e.g., cradle-to-grave, production only) and applying LCA-E is an important research avenue worthy of further investigation.

Energy consumption contributes to HC environmental impacts accounting for at least 50% of the overall greenhouse emissions (GHE) [132–134]. As a result, energy conservation is usually a major task when investigating HTC and materials with short detention times are preferred (Table 7). From Table 1, however, in situ produced materials are reported with detention times 3 times longer than other materials and, therefore, improving synthesis methods with reduced detention time has become an urgent pending research task. Further, energy consumption analysis of modified materials prepared using post-treatment (see Tables 2 and 4) suggests that a high amount of energy is required for the two processes used to produce CBMs. This has significant negative impacts related to greenhouse gas emissions and seriously threatens the environmental viability. Hence, LCA-E studies comparing energy consumption for the same material using in situ and post-treatment are needed to fill this knowledge gap.

From LCA-E standpoint, feedstock influences the impacts associated with the collection, processing and disposal (i.e., LCA-E inventory) by contributing to emissions depending on where feedstock is collected, the resources used to move the feedstock from the collection point to the processing point [135], feedstock characteristics, and desired EHC properties [40,127,134].

LCA-E boundaries are defined depending on the process selected for analysis (Figure 4) [136]. When the LCA-E boundary focuses on EHC production, feedstock particle size has been found to dictate the required pretreatment [137,138] involving higher energy consumption for small-size feedstock. Some biological pretreatments (e.g., enzymatic, anaerobic digestion) can generate small-size particles without increasing the environmental impacts [125,133,139]. However, very little is known about these approaches and further research is required to better understand the trade-offs involved which is also identified as a knowledge gap requiring further investigation [98].

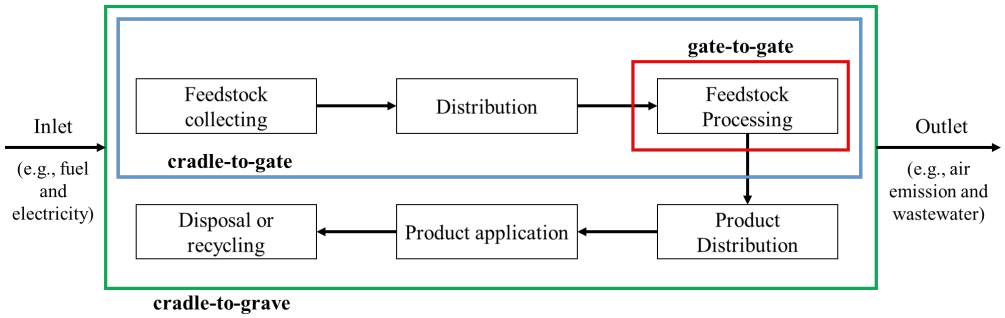

Figure 4. LCA-E boundary delimitation.

HTC generates gas, liquid and solid by-products from which only the solid (i.e., HC) product has been studied with some detail. Only a limited number of studies are available where the other two phases have been investigated [40], which makes the information available on EHC production inadequate for a comprehensive LCA-E. The three main by-products must be included to realistically identify the overall environmental impacts [132,140]. For example, in some cases, the generated biogas mix could be recycled for heating thereby reducing the environmental impacts [131], while in some others, the feedstock (e.g., poultry litter), can produce toxic gases resulting in negative environmental impacts (e.g., H_2S, NOx, SO_2) and may not be suitable for HC production [141]. Liquid phase by-products, depending on their chemical composition, could be distilled to recover valuable components with the potential for reuse [127,129,141].

Few studies have analyzed LCA-E for EHC with a focus on agricultural applications with most of them reporting significantly low environmental impacts [126,140]. No studies on LCA-E for EHC applications in water treatment are available, generating a significant knowledge gap worthy of attention as the potential for reuse of exhausted materials has been suggested for enhancing agricultural production.

Table 7. LCA studies on EHC.

Feedstock	HTC treatment	EHC application	Reference
Olive mill waste	200, 225. and 250° C, 2 h	Energy production	[125]
Poultry litter	180-250° C, 1:3 s/w ratio, 1h	Energy production	[141]
Food waste	Enzymatic pretreatment, 150, 250, 350° C, 1:4 s/w ratio, 20 min	Energy production	[133]
Sewage sludge	220° C, 4 h	Agriculture, Energy	[132]
Sewage sludge	170, 210° C, 2,10 h	Agriculture, Energy	[126]
Olive Pomace	170, 210° C, 2, 10 h	Energy	[134]
Sugarcane bagasse	240° C, 30 min	Energy	[130]
Sewage sludge	208° C, 1 h	Energy	[139]
Peat moss Miscanthus	240° C, 15 min	Energy, Agriculture	[140]
Chlorella vulgaris	180-220° C, 45 min, 10 wt.% acetic acid	Energy	[131]
Green waste	NR	Energy	[127]
Food and packing	225, 250, 275° C, 4, 16, 96 h	Energy	[129]

7. CONCLUSIONS

EHC production and application for water treatment is a continuously expanding research field. Significant knowledge gaps remain constraining wider EHC applications. The following are the main findings of this study:

- EHC is used for organic and inorganic contaminants adsorption. It is considered an environmentally friendly biosorbent with relatively low environmental impacts compared to other CBMs. Nevertheless, environmental impact analysis is needed to ensure a fair comparison with other similar materials and for use in different treatment applications.
- In-situ HTC produces EHC through selective production pathways. However, the way different key factors influence production pathways, and constraining synthesis process optimization remains poorly understood.
- Post-treatment modifications are the most frequently reported for EHC production. However, better in-depth knowledge of the synthesis process and standardized methods are needed.
- LCA-E has been used to identify the environmental impacts of CBM production. Only a limited number of LCA-E studies on HC and EHC production are available, resulting in a dearth of knowledge and significantly constraining the assessment of the factual contribution of EHC to the circular economy.
- EHC materials' hazards analysis is a factor to be considered in LCA-E. However, little is known on the topic, highlighting a significant knowledge gap because LCA-E cannot be accurately undertaken without toxicity information.
- Narrowing down LCA-E to the production process for EHC could create an acceptable approach. Nevertheless, process boundaries should encompass feedstock collection and process application to better understand the environmental benefits of a specific material.

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