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Hydrothermal carbonization of plastic waste: A review of its potential in alternative energy applications

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ABSTRACT

The significant rise in plastic consumption and waste generation, coupled with the urgent need for sustainable energy solutions, has led to innovative research seeking to convert plastic waste into valuable resources. This review focuses on the application of hydrothermal carbonization as a promising technique for transforming plastic waste into valuable products. It highlights the suitability of hydrothermal carbonization for plastic waste conversion, and presents recent reports showing promising results, prospects, and a range of potential hydrochar applications, including solid recovered fuels, catalysts, direct carbon fuel cells and supercapacitors. This review further presents the challenges in utilizing plastic hydrochar across different applications, which include feedstock variability, contamination, scalability, material properties, and environmental considerations. The need for optimized synthesis methods, stable performance, and long-term sustainability is also emphasized. The critical evaluation of the applications of hydrothermal carbonization can contribute to advancing sustainable waste management and renewable energy production.

1. Introduction

Since the onset of large-scale plastic production in the 1940s, there has been a significant increase in the production of plastic, reaching a cumulative global production of 9.5 billion tonnes in 2019 [1]. By 2021, annual global plastic production had reached over 390 mT, with China emerging as the foremost producer, contributing approximately 32 percent of global plastic production [2]. Even during the COVID pandemic, China continuously produced around 7 mT of plastic products monthly, whereas in 2020, Europe witness a decline in plastic production, followed by an increase to 58.7 mT in 2022 [3]. North America, which is the second largest producer, contributed 18 percent in global plastic production in 2021 and approximately 56.9 mT were produced in 2022 accounting to 20 % of global production. According to data published by Statista, 2023 [2] the global plastic market reached a value of 712 billion U.S. dollars in 2023 and has anticipated substantial growth in the coming decade.

This increase in plastic production has resulted in a corresponding increase in plastic waste. Between 2000 and 2019, annual global plastic waste generation doubled to 353 mT [1,4]. Plastics with lifetimes of less than five years make up over two-thirds of all plastic waste, with packaging accounting for 40 %, consumer products 12 %, apparel and textiles 11 % (Fig. 1). Only 9 % of plastic waste is recycled (while 15 % of plastic waste is collected for recycling, 6 % is ultimately discarded as recycling residue), 19 % of plastic waste is burnt, 50 % is disposed in landfills, and 22 % ends up in uncontrolled dumpsites, open pits, or in terrestrial or aquatic habitats, often in impoverished nations. In 2019, 6.1 mT (Mt) of plastic waste escaped into aquatic habitats, with 1.7 Mt

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Acronyms and abbreviations: ABS, Acrylonitrile butadiene styrene; C, Flammability index; CHF, Critical heat flux; CI, Combustion characteristic index; DCFC, Direct carbon fuel cell; D_i, Ignition index; D_f, Burnout index; EG, Ethylene glycol; GHG, Greenhouse gas; HEVs, Hybrid Electric Vehicles; H_f, Index of intensity; HGI, Hardgrove grindability index; HIPS, High impact polystyrene; HTC, Hydrothermal carbonization; HTL, Hydrothermal liquefaction; HTG, Hydrothermal gasification; IPA, Isopropyl alcohol; NiO-SDC, Nickel Oxide and Samarium-Doped Ceria; NRF, National Research Foundation; OECD, The Organization for Economic Co-operation and Development; PC, Polycarbonate; P_c, Critical pressure; PE, Polyethylene; PEMFC, Proton exchange membrane fuel cells; PET, Polyethylene Terephthalate; PP, Polypropylene; PS, Polystyrene; PVC, Polyvinyl Chloride; S, Combustion performance index; SRF, Solid recovered fuel; T_c, Critical temperature; T_f, Burnout temperature; T_i, Ignition temperature; T_p, Peak temperature; TG-DTG, Thermogravimetry-derivative thermogravimetry; TPA, Terephthalic acid.

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flowing into seas. There is an estimated 30 Mt of plastic debris in the seas and oceans, with an added 109 Mt accumulating in rivers.

Among these plastics, thermoplastics are extensively produced, used and discarded, such as polyethylene terephthalate (PET), high-density polyethylene (HDPE), polyvinyl chloride (PVC), low-density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), and others. Fig. 2 shows the global primary plastic production and its corresponding waste, generated by polymer type for 2019, where 109.9 mT of polyethylene (HDPE and LDPE) were produced, and 93.9 mT of plastic were generated (Fig. 2).

Plastic production is an energy intensive process [1,4]. In 2015, plastic production consumed about 6 % of global electricity generated from coal [5,6]. The energy used in plastic production (including the energy of raw materials) is 62–108 MJ kg^{-1}, much higher than that for paper, wood, glass, and metals [7]. By 2050, plastic production and processing could account for up to 20 % of global oil consumption and 15 % of annual carbon emission budgets [7,8]. Upgrading plastic into solid fuel or other energy-related purposes [9] could contribute to offsetting the energy input from coal with corresponding emission reductions [10,11]. About 73 % of anthropogenic greenhouse gas (GHG) emissions are from energy and about 3 % comes from waste, therefore upgrading waste to energy could also offset GHG emissions [12].

Plastic waste accumulation has emerged as a major environmental concern worldwide [13], with limited effective management strategies. Simultaneously, the increasing demand for circularity and alternative energy sources has led to the exploration of unconventional feedstocks that can contribute to a more sustainable future [11,14].

The concept of hydrothermal carbonized plastics epitomizes the synergy between scientific advancement and environmental stewardship [15–17]. Hydrothermal carbonized plastics represent a groundbreaking solution at the intersection of sustainability [18], renewable energy, and net-zero goals [19]. This innovative process of transforming discarded plastics into a valuable resource has potential far-reaching implications for sustainable transport, renewable energy generation, and achieving net-zero emissions [19] and can be a powerful tool to drive the transition towards cleaner energy alternatives. Plastic-based hydrochar could be utilized as a renewable fuel [20], powering vehicles and machinery, thus reducing our reliance on finite fossil fuels and curbing greenhouse gas emissions [19].

In the realm of sustainable transport, plastic hydrochar has the potential to revolutionize the way vehicles are powered [21–25]. Integrating plastic hydrochar into fuel systems would be a significant step towards achieving emission reduction targets and fostering cleaner air quality in urban areas [26]. This aligns with the global push for greener transportation options, helping to create a more environmentally friendly and efficient mobility landscape. As nations strive to eliminate or offset their carbon emissions, this innovation can serve as a critical bridge towards a carbon-neutral future [26]. Through the diversion of plastic waste from landfills and incineration [13,19,27], contributions to carbon capture and reduction endeavors are made, consequently accelerating the trajectory towards a sustainable environment.

This review aims to evaluate the potential of hydrothermal carbonization of plastics as a promising technique in transforming plastic waste into solid recovered fuels, catalysts, direct carbon fuel cell and supercapacitors etc., thereby addressing both the environmental issues associated with plastic waste and the need for alternative energy applications.

The paper will first delve into the principles of hydrothermal carbonization and its suitability for transforming plastic waste, and then examine the potential of hydrothermally carbonized plastic waste (hydrochar) as solid recovered fuels, supercapacitors, catalysts, and direct carbon fuel cell, highlighting promising results in recent studies. Finally, the challenges and prospects of this technology will be discussed, shedding light on potential advancements and opportunities for further research. By exploring the potential applications of hydrothermal carbonization of plastic waste, this review seeks to contribute to the growing body of knowledge in sustainable waste management, renewable energy production, net zero goals, fuel, and sustainable transport.

2. Hydrothermal technology

2.1. The hydrothermal processes

Hydrothermal processes take place at high temperatures under high saturated pressures that trigger a set of reactions which change the physicochemical properties of water (density, permittivity, ionic product), producing energy-dense fuels and valuable products or chemicals [28,29]. Compared with other waste recovery technologies, hydrothermal processes can handle a variety of feedstock, from low to high-water content and operate at lower temperatures. Hydrothermal techniques can target specific molecules or functional groups in feedstock, and can handle a wide variety of feedstocks, making them appropriate for a variety of applications. In some cases, catalysts are not required, due to the peculiar reaction environment provided by the high temperature and pressure in the presence of water [30]. This eliminates the requirement for catalyst selection, handling, and regeneration, thus simplifying the process and lowering the cost. Hydrothermal procedures can assist in reducing the discharge of hazardous gases and volatile

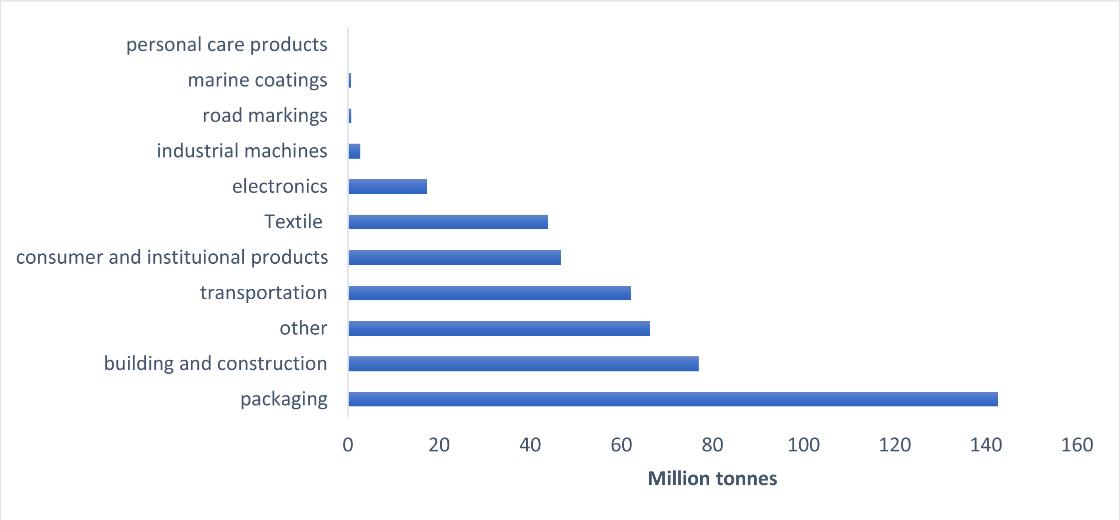

Fig. 1. Global primary plastic waste production by sector as of 2019 [1].

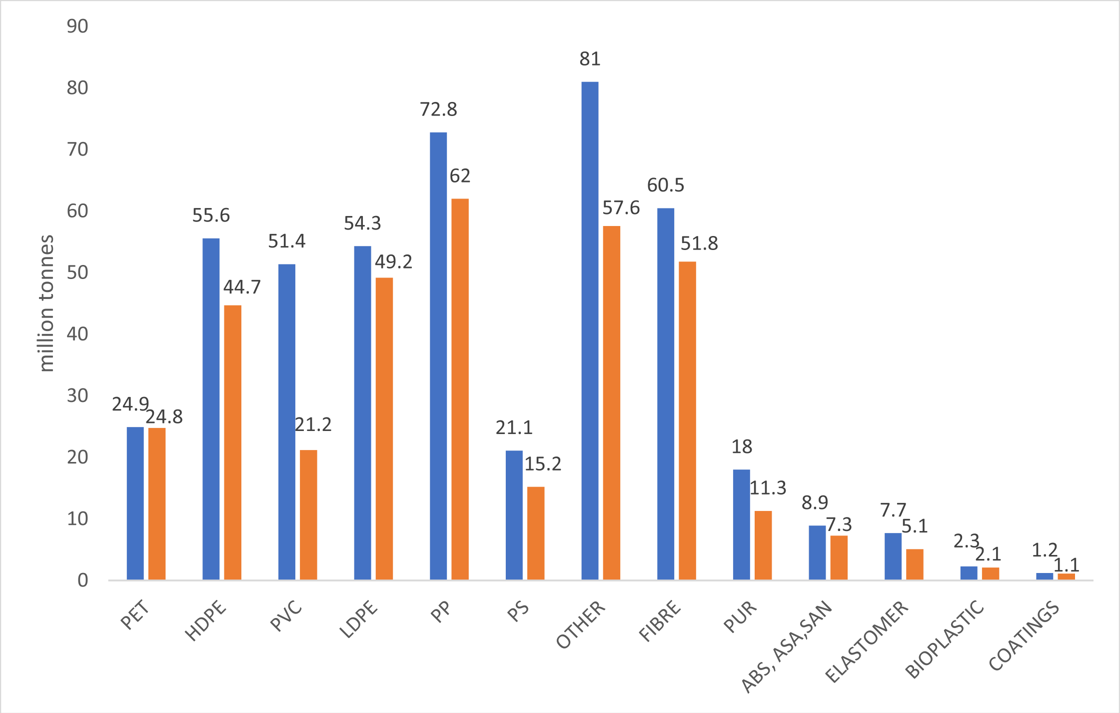

Fig. 2. Global primary plastic production and plastic waste generation by polymer type for 2019 (adapted from [1]). (🟦) Plastic produced, (🟧) plastic waste generated. Acrylonitrile Butadiene styrene (AB), Acrylonitrile Styrene Acrylate (ASA), Styrene Acrylonitrile (SAN), Polyurethane (PUR), Polyvinylchloride (PVC), Polystyrene (PS), Linear Low-Density Polyethylene (LLDPE), Polypropylene (PP), Polyethylene Terephthalate (PET), High-Density Polyethylene (HDPE).

organic compounds (VOCs) [19,31–33].

The environmental impact of hydrothermal processes depends on the energy efficiency, time and complexity, subcritical and supercritical conditions [34,35]. One promising technique receiving attention is hydrothermal decomposition using subcritical fluids [36,37]. Subcritical water is water at temperatures between 100 °C and 374 °C, with sufficient pressure (221 bar) to keep the water in a liquid state [29]. When water exceeds its critical point ($T_c = 373$ °C, $P_c = 221$ bar), it becomes supercritical water [38] (Fig. 3). Subcritical and supercritical water have been used extensively in the degradation of plastics and biomass [39]. Various reaction mechanisms can occur in parallel during feedstock conversion in subcritical or supercritical water, such as hydrolysis, depolymerization/polymerization, isomerization, dehydration, decarboxylation, aromatization, condensation, methanation, hydrogenation and dehydrogenation, amongst others [40,41]. Hydrothermal technology has been applied on polyethylene terephthalate (PET) water [42], polycarbonate (PC), polystyrene (PS), polyethylene (PE), polyvinyl chloride (PVC) and even thermosets. According to Liu et al. [43], thermosets containing breakable connections such as ester bonds, acetal linkages, and Schiff bases, are susceptible to hydrolysis within a solution, leading to the retrieval of monomers or oligomers which can then be employed in creating new thermosets or related materials.

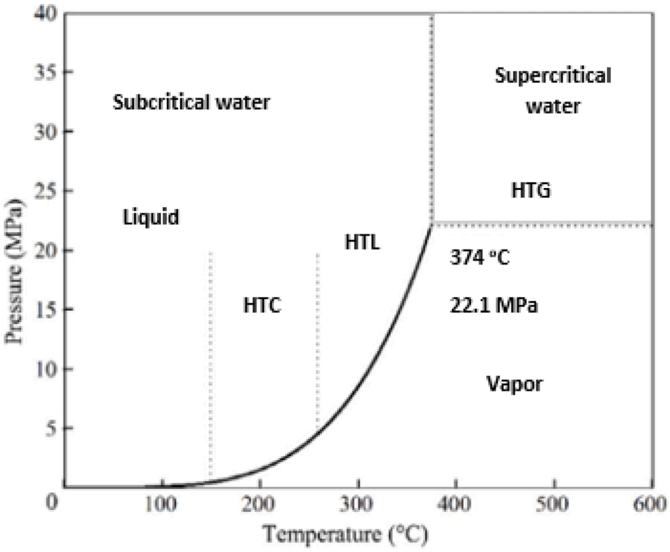

Fig. 3. Phase diagram of water, including the subcritical and supercritical pressure and temperature region [16].

Hydrothermal technology can be divided into three processes according to the temperature range: hydrothermal carbonization (HTC), hydrothermal liquefaction (HTL) and hydrothermal gasification (HTG) [28,44], see Fig. 4. Hydrothermal Carbonization (HTC) is a process typically conducted at moderate temperatures, ranging from 180 °C to 250 °C, and at relatively low to moderate pressures, typically between 10 and 40 bar. HTC reactions are relatively longer, often taking several hours to complete, and they yield hydrochar as the main product [19,31–33]. In contrast, Hydrothermal Liquefaction (HTL) involves higher temperatures, usually between 250 °C and 500 °C, and higher pressures, often exceeding 50 bar and reaching up to 200 bar or more. HTL processes are relatively shorter, taking minutes to a few hours, and produce a liquid biocrude or bio-oil as the primary product. This bio-oil can be further refined into transportation fuels, chemicals, and other valuable products [28,44]. On the other hand, hydrothermal gasification (HTG) utilizes even higher temperatures, typically ranging from 500 °C to 800 °C, and similar high pressures, usually between 50 and 200 bar [38]. HTG reactions are rapid and take only a few minutes to complete, generating synthetic gas (syngas) as the main product which can be used to produce fuels, chemicals, and electricity [34,35] (Fig. 4).

2.1.1. *Hydrothermal carbonization*

Hydrothermal carbonization (HTC) involves the treatment of biomass, including various organic wastes such as plant and plastic waste, in a high temperature (100–250 °C), high-pressure water

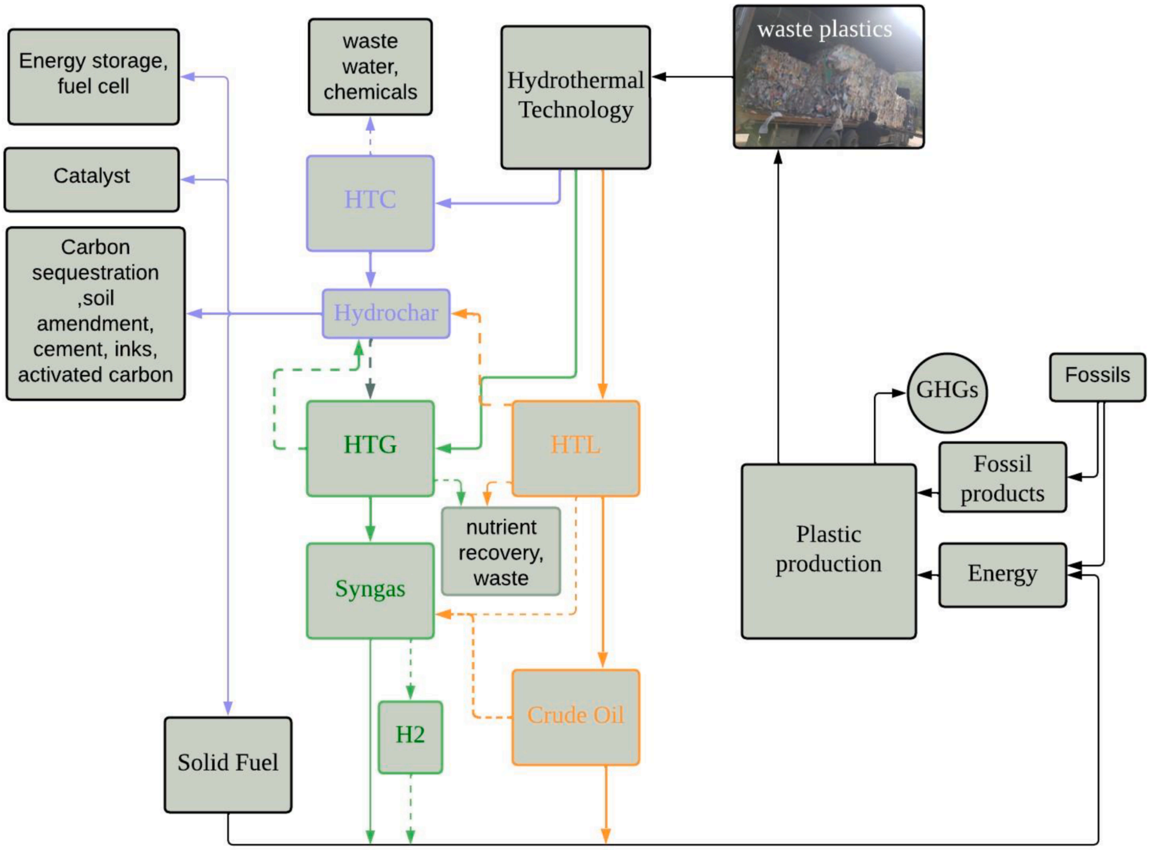

Fig. 4. Schematic overview of the hydrothermal processes. (●) hydrothermal carbonization (HTC), (●) hydrothermal liquefaction (HTL) and (●) hydrothermal gasification (HTG) pathways. Full lines indicate the production of primary products while broken lines indicate the production of secondary products.

environment where water is kept in a liquid state yet exhibits unique properties. These properties make water a highly polar solvent with increased ionization capacity and reduced dielectric constant, increasing its ability to dissolve and react with organic compounds such as polymers. For plastic waste, subcritical water hydrolysis promotes polymer bond scission and the attack on polymer bonds, resulting in the separation of individual monomer units and decomposition of the polymer into smaller fragments [16,45]. During the hydrothermal carbonization of plastic waste, besides bond cleavage, the resulting hydrochar typically retains a carbonaceous structure which is composed of the carbon backbone of the original plastic polymer [41]. The hydrochar may contain residual aromatic and aliphatic carbon compounds [41], depending on the type of plastic waste and the specific conditions of the hydrothermal process. While some chemical bonds are broken during decomposition, such as ester linkages in the case of polyester plastics, the carbon-carbon backbone of the polymer is not destroyed [46]. The hydrothermal conditions promote the rearrangement and stabilization of carbon atoms, leading to the formation of a solid carbon-rich hydrochar [20]. The HTC process may introduce new oxygen-containing functional groups [47], such as the carbonyl (>C = O) and the hydroxyl (-OH)) groups on the carbonaceous structure [48]. These groups can influence the reactivity, properties, and potential applications of hydrochar, making it suitable for various uses as a renewable fuel or precursor for carbon materials. The specific composition and structure of the hydrochar will depend on various factors, including the type of plastic waste, the temperature, pressure, and residence time during the hydrothermal process [49].

Several hydrothermal carbonization studies have been documented in the existing literature [50,51,46,52–54]. Studies by Zhao et al. [55] and Lu et al. [56] focused on the removal of harmful elements and the dechlorination of PVC during the HTC process under mild conditions. Becker et al. [57] explored the composition of condensed steam water generated during the HTC process. The condensed steam contains essential nutrients, including nitrogen, phosphorus, potassium, and others, which can be extracted and utilized as fertilizers or soil conditioners. The extraction of valuable chemicals such as levulinic acid, furfural, 5-hydroxymethylfurfural, and phenolic compounds from the condensed steam is also possible. The condensed steam can further undergo anaerobic digestion, yielding biogas as a renewable energy source [58]. Chen et al. [59] investigated the effects of temperature and pressure on hydrochar properties, for optimization purposes.

2.1.2. *Hydrothermal liquefaction*

During hydrothermal liquefaction (HTL), the process can result in the release of alcohol and carboxylic acid functionalities, and the polymer chains can be broken down into smaller hydrocarbons, including alkanes and olefins containing a mixture of saturated and unsaturated hydrocarbons exhibiting different structures [60,61]. The resulting products may encompass styrene, benzene, and other aromatic hydrocarbons, with aromatics potentially being retained in the oil fraction. Also, the formation of chlorinated hydrocarbons and other byproducts is possible.

A study conducted by Čolnik et al. [62] on hydrothermal decomposition of PE waste used supercritical water of 380–450 °C with the addition of acetic acid as a catalyst and 15–250 min reaction time. The plastic was degraded majorly into oil and wax with minor fractions of gas, solid and aqueous solution [63]. The oil phase was made of alkanes, alkenes, cycloalkanes, aromatics, and alcohols, while the gas phase contained light hydrocarbons (C1–C6) [40]. When temperatures and reaction times increased, the concentrations of the long-chain hydrocarbons (>C9, diesel fractions) decreased [64]. The plastic feedstock of PP and PS produced cyclic compounds, while HDPE predominantly generated alkanes. A review on the conversion of municipal solid waste and mixed feedstocks for commercial HTL in bio-refineries was carried out focusing on the yield and quality enhancement of bio-crude through hydrothermal liquefaction of municipal solid waste and the challenges regarding scale up. The review reported 65–70 % energy recovery ability of HTL [65]. In another study, it was reported that the biocrude yields from hydrothermal liquefaction of mixed plastic wastes and polypropylene increased, reaching up to 45 % of the initial feedstock, with the high heating values surpassing 50 % under temperatures of 350 to 450 °C, with pressures reaching up to 20 bar and a retention time of 30 min [66]. In Rahman et al. [61] PET, HDPE, LDPE, PP and PS were blended 42 %, 20 %, 20 %, 4 %, and 14 % by weight respectively mimicking household plastic waste with their corresponding single plastic counterparts as control. The HTL reaction of the plastic mixture produced 23 % liquid crude and 23 % solid product. For the control experiments, HDPE yielded the highest crude oil at 76 %, while PET predominantly produced 80 % solids. The crude oil derived from plastics contained 36−92 wt.% gasoline-range compounds. HDPE decomposition resulted in straight-chain alkanes, while products from PP and PS consisted of cyclic compounds [61].

2.1.3. Hydrothermal gasification

Hydrothermal gasification (HTG) represents a thermochemical process designed to convert feedstocks (organic materials, plastics etc.) into valuable gaseous products within a hydrothermal gasifier at elevated temperatures and pressures, breaking down complex molecular structures to simpler ones. It has received attention due to its ability to convert a wide range of raw materials, with high energy, conversion efficiency, and short reaction time [67–69]. This method is suitable for feedstocks with high moisture content because it does not require additional drying and dehydration processes, which would result in high operational expenses [70,71].

The gasification process transforms plastic waste into a mixture of gases (syngas), predominantly carbon dioxide (CO_2), methane (CH_4), hydrogen (H_2), and traces of other hydrocarbons [72,73], which can further be processed into liquid fuels through various gas-to-liquid technologies. It can also be converted to hydrogen, methanol, and ammonia [69,74]. Like biomass, plastic waste hydrothermal gasification may lead to the formation of tar compounds, which are addressed through secondary reactions like tar reforming. Inorganic components undergo mineralization, resulting in the formation of solid residues or ash [69,74].

A review explored the thermophysical chemistry of hydrothermal decomposition of waste polymers (municipal solid waste (MSW), sewage sludge, mixed plastics, petroleum waste and used tires) and biomaterials under sub and supercritical temperatures [30]. One advantage is the ability to utilize the syngas directly without compression, since it is produced at elevated temperatures and pressures, which can reduce the cost. In Bai et al. [72], polystyrene waste was converted into syngas in a batch reactor at 500–800 °C, 1–60 min reaction time and 22–25 MPa. The study focused on the optimization performance of gasification parameters and found that reducing the reaction time and increasing the feedstock concentration improved conversion efficiency to 94 % (800 °C, 60 min, 23 MPa). In Okolie et al. [30], several other studies were reported on the decomposition of plastics using hydrothermal gasification. In one of the studies, plastic compounds were broken down through gasification using subcritical and supercritical water with the conversion of diphenyl ether to phenol. Another study revealed the transformation of monomer-cast nylon through the interaction of subcritical water and a commercial heterogenous zeolite (H-Beta) catalyst, which led to a remarkable increase in the hydrolysis rate (31–60 %) at a lower temperature of 300 °C for 50 min. The zeolite catalyst provided an alternative reaction pathway with lower activation energy, allowing the reaction to occur at milder temperatures. The catalyst facilitated the hydrolysis of the monomer-cast nylon by providing active sites for the reaction, thereby reducing the energy barrier for the process.

2.2. Advantages of hydrothermal technology over conventional gasification and pyrolysis

Hydrothermal technology operates in the presence of water, making it well-suited for processing a variety of feedstock, including mixed waste plastics, providing greater flexibility in dealing with diverse waste streams that may contain residual water [37]. This helps to eliminate the dewatering stage [75]. Traditional gasification and pyrolysis processes may have limitations on the types of plastics they can effectively process. Hydrothermal technology produces more solid carbon, more surface functional groups and has proven to be a practical method for enveloping pre-existing nanostructures with carbonaceous coatings [76]. Hydrothermal liquefaction surpasses pyrolysis with advantages such as higher biocrude yields, and higher energy density products within a shorter reaction period [77]. In addition to energy recovery, hydrothermal processes may facilitate the recovery of valuable nutrients from plastic waste providing additional environmental benefits. It also facilitates the removal of heavy metals [78] and halogens such as chlorine [79].

3. Hydrothermal carbonization technology

3.1. Industrial hydrothermal carbonization processes

The hydrothermal carbonization (HTC) process can convert a variety of waste (ranging from municipal solid sewage, medical waste, industrial, biowaste, fishery, livestock to plastic waste etc.) [45,80] to different products which can be used for different purposes [81,82,107,108]. Fig. 5 shows a general description of a commercial hydrothermal carbonization process adapted from the Hokuto Kogyo hydrothermal plant, Japan [83], and the Hypersolution pilot plant in South Korea (Fig. 6).

The process starts with the supply of saturated steam from a boiler. A maximum operating temperature and pressure of 280 °C and 30 bar is set respectively [84]. Inside the HTC system, shovels or shafts continuously agitate the waste for heat transfer and homogeneity. When the waste is completely decomposed, the gas valves are opened, and the steam is discharged to the condenser as wastewater. The wastewater is then sent to a treatment system where all harmful chemicals are removed before it is discharged into the sewer system.

After decomposition, the hydrochar is discharged onto a conveyor system and transported to a drying system. The final product is passed through a magnetic separator and a sieve to remove metals and larger particles. The final hydrochar product can then be used as carbon material. These hydrochar have important characteristics that make them appropriate for a variety of uses: supercapacitors, catalyst, inks, tire additives, pelletized as solid fuel for coal furnaces, cement rotary kiln, or as additives in cement [85–87].

Hydrochar is normally acidic, lower in ash concentration, and contains more alkyl groups because of the strong demineralization from the inorganic metal washing and the presence of oxygenated functional groups. For energy related uses, the particle sizes of hydrochar are important. Hydrochar possesses uniform, spherical, low-porosity, and high specific surface area particle sizes [87,88]. The plastic hydrochar can be used for electricity generation.

For electricity generation and grid integration, the solid-engineered pulverized fuel (plastic hydrochar) is used in high-temperature combustion burners for heat production which is transferred to the steam boiler [89,90] which powers the steam turbine and generator. The electricity generated from the steam turbine generator is passed to the electric grid system which in turn is used in the HTC treatment plant as energy.

The emissions from steam are managed with the installation of a multi-pollutant air quality control system. These include twin cyclone

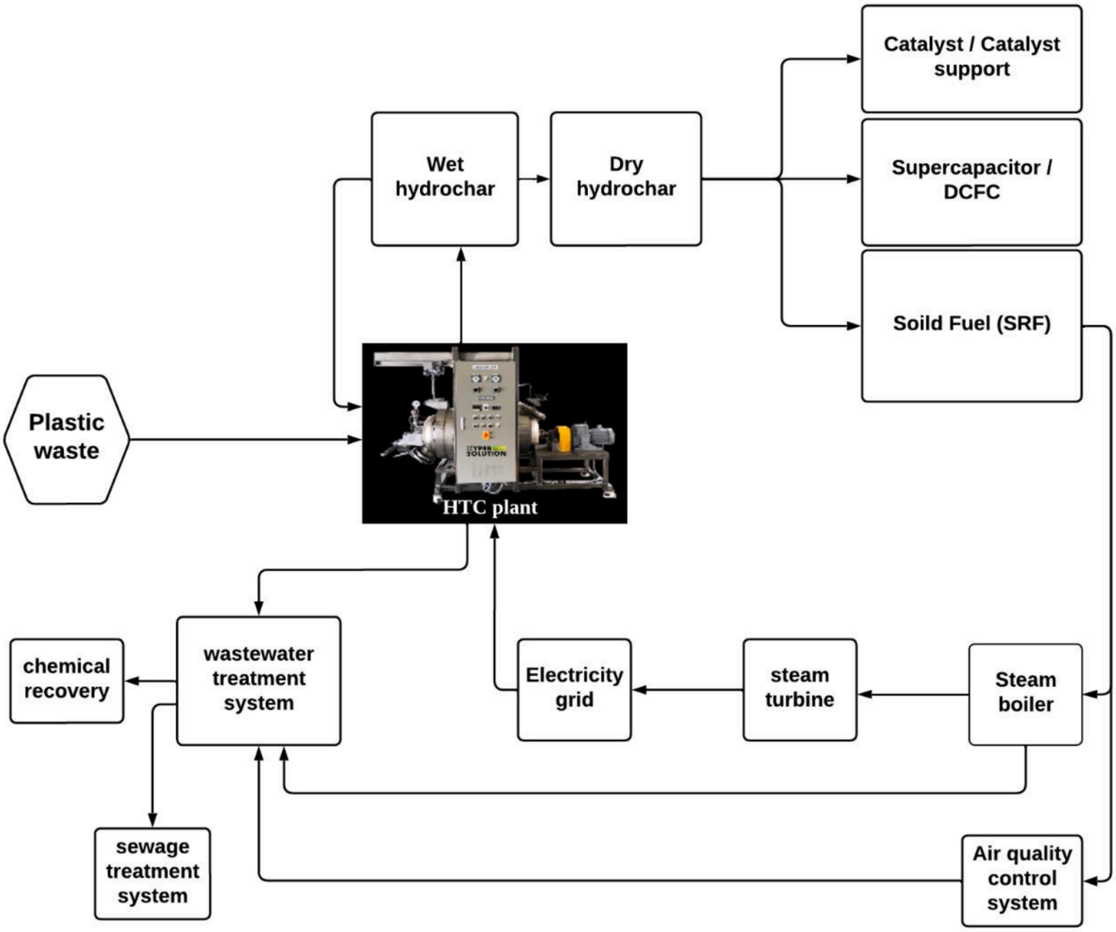

Fig. 5. Plastic hydrochar production and utilization pathway illustrating a circular economy.

for initial particulate removal [91,92], selective catalytic reduction with urea injection to reduce NO_x and VOC (volatile organic compounds) [93], fabric filter baghouse to control particulate matter (PM) and metals [94], wet scrubber with sodium hydroxide to control SO_2, HCl, HF [95] and a carbon adsorption system to control mercury (Hg) and VOC [96–98].

3.2. Advantages of plastic hydrochar over biomass hydrochar

Plastic hydrochar may exhibit a higher calorific value due to the energy-dense nature of some plastic polymers, providing a more potent fuel source compared to biomass [78,99]. Plastic hydrochar could have lower ash content compared to biomass hydrochar, contributing to cleaner combustion and potentially reducing issues associated with ash-related complications. Plastics typically have lower moisture content than biomass, which can be advantageous in terms of energy content and combustion efficiency [88]. The unique chemical composition of plastic hydrochar, influenced by the polymerization and degradation of plastic polymers, may result in enhanced reactivity during combustion, contributing to efficient fuel burnout [100]. Plastic hydrochar production offers a potential means of recycling and utilizing plastic waste, addressing environmental concerns associated with plastic disposal.

3.3. The mechanism of hydrothermal decomposition of waste plastic

During the hydrothermal carbonization of PET, it is subjected to high temperatures and pressure in the presence of water, which causes the ester bonds in the polymer chain to break down, releasing terephthalic acid (TPA) and ethylene glycol (EG), while monomers like isopropyl alcohol (IPA), and 1,4-dioxane are formed [101] and later decarboxylated (benzoic acid), dehydrated (acetaldehyde) and dimerized (diethylene and triethylene glycol) [102]. The final products from PET decomposition are TPA as solid [42], carbon dioxide, which is released as gas, and isopropyl alcohol, 1,4-dioxane, acetaldehyde, triethylene glycol, benzoic acid which are carried into the steam and later condensed as liquid. However, controlled conditions, additives, and other impurities may have a significant impact on the final products. When a blended mixture of PET and nylon-6 were decomposed in a hydrothermal system, 55 % of solid TPA was recovered [41]. However, when PET was decomposed alone, 75 % of solid TPA was recovered. Different polymers behave differently during hydrothermal carbonization of plastic waste. Čolnik et al. [101] carried out hydrothermal carbonization of PET waste under subcritical conditions of 250 °C and short reaction time, which resulted in incomplete reaction, although the bulk on the PET was hydrolyzed.

1. Ester bond hydrolysis: (PET) $C_{10}H_8O_4 + H_2O \rightarrow nC_8H_6O_4$ (TPA)+ $nC_2H_6O_2$ (EG)
2. Ester bond hydrolysis: $C_{10}H_8O_4 \rightarrow C_3H_8O$ (IPA) + $C_4H_8O_2$ (1,4-dioxane)
3. Decarboxylation of TPA: $C_8H_6O_4$ (TPA) → $C_7H_6O_2$ (benzoic acid) + CO_2
4. Dehydration of EG: $C_2H_6O_2$ (EG) → C_2H_4O (acetaldehyde) + H_2O
5. Dimerization of EG: 2 $C_2H_6O_2$ (EG) → $C_4H_{10}O_3$ (diethylene glycol)
6. $C_2H_6O_2$ (EG) + $C_4H_{10}O_3$ (Diethylene glycol) → $C_6H_{14}O_4$ (triethylene glycol)

Fig. 6. Examples of hydrothermal carbonization plant. (a) Hydrothermal carbonization plant in Hokuto Kogyo Japan [83] (b) A 200-L pilot plant, South Korea with (c) the HTC reactor cross section, (d) the rotary shaft and (e) the plastic hydrochar product. Image (b-d) are original images taken by the authors in 2022 at the Hypersolution pilot plant, Sejong, South Korea.

In the hydrothermal carbonization of polyethylene (PE) under subcritical water conditions, a series of reactions results in the breakdown of the polymer chain. Initially, polyethylene reacts with water, leading to the formation of aliphatic alcohols and other intermediate products. Subsequent transformations involve the dehydrogenation and fragmentation of the polymer backbone [103]. The generated intermediate species may include alkenes, aldehydes, and ketones. Further reactions may lead to the formation of smaller hydrocarbons, such as methane, ethylene, and propylene, as well as alcohols and carboxylic acids. The final products of polyethylene decomposition are diverse and can encompass gases like carbon dioxide and methane, as well as liquid byproducts such as alcohols and acids. The overall process is sensitive to reaction conditions, and the presence of impurities or catalysts may influence the nature and yield of the final products. These reactions are similar in the case of polypropylene.

1. Hydrolysis: $(C_2H_4)_n + H_2O \rightarrow C_2H_5OH$
2. Dehydrogenation: $C_2H_5OH \rightarrow C_2H_4 + H_2O$
3. Formation of aromatic compounds: $C_2H_4 \rightarrow C_6H_6 + 3H_2O$

In the case of polystyrene (PS), it is first depolymerized into styrene monomers where they undergo elimination reaction to form benzyl alcohol. This benzyl alcohol undergoes subsequent transformations, leading to the generation of benzaldehyde and benzoic acid as intermediate products. Following this, some distinct reactions take place: benzaldehyde undergoes decarboxylation, shedding a carbon dioxide molecule, while benzyl alcohol experiences dehydration, losing a water molecule. Concurrently, two benzoyl radicals undergo dimerization, forming a dimer product. The hydrothermal energy breaks the C—C bond, producing phenyl radicals, phenylmethyl radicals, and alkyl radicals. Various organic products, including diphenyl compounds with an extended carbon chain between phenyl radicals, result from the combined reactions of different free radicals benzene [46]. A phenylmethyl radical bonds to two alkyl radicals, forming (1-methylethyl)-benzene [46,104], which, through an elimination reaction, becomes α-methyl styrene. Linear alkyl radicals with three carbon atoms bridge the gap between two individual phenyl radicals, resulting in the formation of 1,3-diphenyl-propane. Similarly, pentyl radicals link aromatic radicals, leading to chain termination, and the likelihood of cyclization reactions increases with the carbon chain, resulting in the formation of 1,2-diphenylcyclopropane. Furthermore, intermolecular rearrangement between dissociative phenyl radicals leads to the emergence of 2-phenyl-naphthalene and multi-benzene compounds [46, 104–106]. These chemical changes contribute to the breakdown of the polystyrene polymer, yielding various final products. Potential degradation products include styrene monomer, styrene dimer, styrene trimer, toluene, ethyl benzene, isopropyl benzene, and diphenyl compounds including 1,2-diphenylcyclopropane,1,3-diphenylpropane, (1-methylpropane-1,3-diyl) dibenzene, 2-phenyl-naphthalene, anthracene, (1-methylethyl)-benzene, α-methylstyrene, (1-methylpropane-1, 3-diyl) dibenzene, 2-phenyl-naphthalene, 4′-phenyl, terphenyl, and quaterphenyl [104]. The specific products generated depend on various factors, including reaction conditions, temperature, and the presence of catalysts. Note that the exact pathways and products may vary based on specific reaction conditions and the presence of impurities in the hydrothermal feedstock:

1. $(C_8H_8)_n \rightarrow C_6H_5CH{=}CH_2$ (styrene monomers)
2. $C_6H_5CH{=}CH_2 \rightarrow C_6H_5CH_2OH$ (benzyl alcohol)
3. Breakdown of C—C Bonds: $C_6H_5CH_2OH \rightarrow C_6H_5$ (phenyl radicals), $C_6H_5CH_2$ (phenyl methyl radicals), CH_3(alkyl radicals)
4. $C_6H_5CH_2 + 2CH_3 \rightarrow C_6H_5CH_2CH_3$ (1-methylethyl)-benzene

5. $C_6H_5CH_2CH_3 \rightarrow C_6H_5CH(CH_3){=}CH_2$ (α-methyl-styrene)
6. Linear alkyl radicals+2-phenyl radicals→1,3-diphenyl-propane
7. Pentyl radicals + Aromatic radicals→1,2-diphenylcyclopropane
8. Dissociative phenyl radicals→2-phenyl naphthalene

The hydrothermal carbonization of PVC goes through hydrolysis, oxidation, aromatization, polymerization, and esterification reactions [48,107]. The HTC of polyvinyl chloride (PVC) forms aliphatic alcohols and releases hydrochloric acid. Subsequently, dehydrochlorination occurs, leading to the generation of alkene, water, and additional hydrochloric acid. Concurrently, hydroxyl substitution takes place, where hydroxyl groups (-OH) substitute for chlorine atoms, releasing hydrochloric acid [79,108]. Aromatization then follows, transforming hydroxyl-substituted compounds into aromatic structures such as polyaromatic hydrocarbons (PAHs). Esterification involves the reaction of hydroxyl groups with carbon dioxide (CO_2), resulting in the formation of esters. Collectively, these reactions contribute to the chemical changes that PVC undergoes during hydrothermal carbonization. The aromatic hydrocarbon (C—H, C═C), hydroxyl (–OH), amino group (–NH), methylidyne (≡C—H), cycloalkane or aliphatic hydrocarbon (C—H, CH_2, CH_3), Carbonyl groups (C═O), hydrogen bonded carbonyl (-O-), phenol, alcohol, ether, esters, are some of the functional groups identified throughout the hydrothermal carbonization of PVC [47].

3.4. Dechlorination of PVC using HTC

Chlorine is a hindering factor to chemical recycling (degradation) of PVC, unlike the other counterparts (PP, PE, and even PS) which do not have chlorine. However, due to HTC conversion technology, PVC can be dechlorinated while bromine (which acts as a flame retardant), can be successfully removed. A 100 % dechlorination efficiency is attainable, however, under hydrothermal carbonization temperatures, 93 % of PVC can be dechlorinated. With the addition of a catalyst or a longer reaction time, 100 % dechlorination is possible [108]. Adolfsson et al. [109] reported that citric acid acting as a catalyst under 240 °C can facilitate the removal of chlorine completely after 15 h. However, other findings showed a 98 % chlorine removal at 240 °C, 3 MPa for 2 h [56]. Sodium hydroxide also acts as a catalyst in dechlorination, however, at higher concentrations it may lead to surface poisoning. Li et al. [79] reported a 94 % dechlorination at 240 °C with 1 % NaOH. Hydrothermal technology is also suitable for dechlorination of plastic waste [108], with the co-hydrothermal carbonization of PVC and bagasse proven to be highly effective. Liu et al. [110] reported that bagasse supplied a large amount of -OH in the hydrothermal system, causing the substitution of -Cl with -OH, leading to the conversion of organic-Cl to inorganic-Cl which is migrated to the liquid. Substitution reaction is the dominant dechlorination pathway, although elimination reactions partly contribute. Bagasse has been blended with acrylonitrile butadiene styrene (ABS) and high impact polystyrene (HIPS) for hydrochar production [110].

4. Hydrochar

4.1. General properties of hydrochar as an energy alternative

4.1.1. Proximate analysis

The proximate analysis of a compound is a measure of its fixed carbon, ash content, volatile matter, and moisture content [20]. Analysis of these parameters usually follows the ASTM 2009 standards. Fixed carbon is inversely proportional to volatile matter, and is derived by subtracting the ash, moisture, and volatile matter from 100 [111]. The fuel ratio is dependent on fixed carbon and volatile matter. During the HTC process, the volatility reduces while the fixed carbon increases and the amount of ash left after combustion is the ash content [112]. A good hydrochar should have a lower ash content since this is an insoluble material that inhibits the combustion process [112]. Bardhan et al. [111] compared the proximate analysis of some selected feedstocks before carbonization (PVC, food waste, raw coal, sub-bituminous coal, lignite coal and low-rank coal). They found out that polyvinyl chlorides had the highest volatile matter range of 93 to 95 %, with the smallest amount of fixed carbon (4–6 %) and ash content (0.44 %). The volatile matter of coal was the smallest (34–59 %) with high ash content (2–9.6 %) which was like that of food waste (1.7–9.2 %). All the coal types demonstrated extremely high fixed carbon content.

4.1.2. Ultimate analysis

Ultimate analysis is the measure of a fuel carbon, hydrogen, oxygen, nitrogen, and sulfur contents. For hydrochar, determining the hydrogen/carbon (H/C) and oxygen/carbon (O/C) molar ratios is a key factor in determining the degree of deoxygenation and aromatization. A high O/C ratio implies low hydrophobicity and high polarity, whereas a high H/C ratio indicates low aromatic content [113] . The plotted H/C and O/C ratios of lignite and bituminous coal are used to assess the hydrochar molar ratios, where coalification degrees of the products are compared [114]. The hydrochars produced by hydrothermal carbonization of PVC plastics at 220–260 °C in 60 min revealed that hydrothermal temperature had a substantial effect on hydrochar chemical structure, with the van Krevelen diagram showing a similar maturation rate to lignite coal. [48]. This suggested that higher temperatures during HTC positively influenced the fuel characteristics of the PVC hydrochar. This was evident as the ratios of O/C and H/C shifted towards those of bituminous coal. Elevated temperatures also caused a reduction in the H/C and O/C ratios, resulting in higher aromaticity, demethylation, and dehydration of the hydrochar [48]. Hydrothermal carbonization was carried out on marine mixed plastic waste (PP, PE, PET, Nylon) at different temperatures (200, 250 and 300 °C) where the van Krevelen diagram showed that the mixed plastic waste hydrochar fell in the fuel oil region, indicating a high-quality fuel oil suitable for coal power plants etc. [20]. Another study investigating the co-hydrothermal carbonization (co-HTC) process at 230 °C using coal waste and food waste found that this increased the elemental carbon content to a maximum of 49.5 %. The co-HTC treatment in an acidic environment created by the HTC of food waste also had a synergistic effect, reducing the sulfur content to a minimum value 1.4 % compared to the raw coal waste values 8.5 % [115]. It was also observed that during the hydrothermal carbonization of paper mill waste, higher reaction temperatures facilitated dehydration and decarboxylation, resulting in hydrochars with lower H/C and O/C ratios, which improved the fuel quality. The HTC treatment effectively produced cleaner solid fuel with low nitrogen and sulfur content, enabling the recovery of hydrochar with higher carbon content, improved energy densification, and enhanced combustion properties [116]. Fig. 7 presents the van Krevelen diagram of different plastic hydrochar as reported by some authors [20,117,118].

4.1.3. Heating value

The heating value of a substance (energy or calorific value, expressed in MJ/kg) is the amount of heat released during its combustion. Energy values vary per fuel type or material but are important parameters in determining fuel quality. Most plastics have an energy content of 18–44 MJ/kg, which is higher than that of most coals (Fig. 8). The higher the heating value of a waste polymer, the better its recycling or energy recovery potential. Some studies have reported that blending plastics with other materials such as biomass or coal produces heating values higher than those of their corresponding feedstocks [119]. For example, Bardhan et al. [111] found that the heating value of blended corncob and PVC hydrochar was 32.8 MJ/kg, higher than the values of the individual feedstock (corncob = 19 MJ/kg; PVC = 20 MJ/kg). The higher energy content of hydrochar could be attributed to a higher carbon content in the blended hydrochar (76 %) compared to the individual feedstocks (44 %) [111]. The chemical reactions occurring during hydrothermal carbonization of the blend can lead to the formation of new compounds or structures with higher energy content and the combination of corncob and PVC could lead to a more efficient conversion

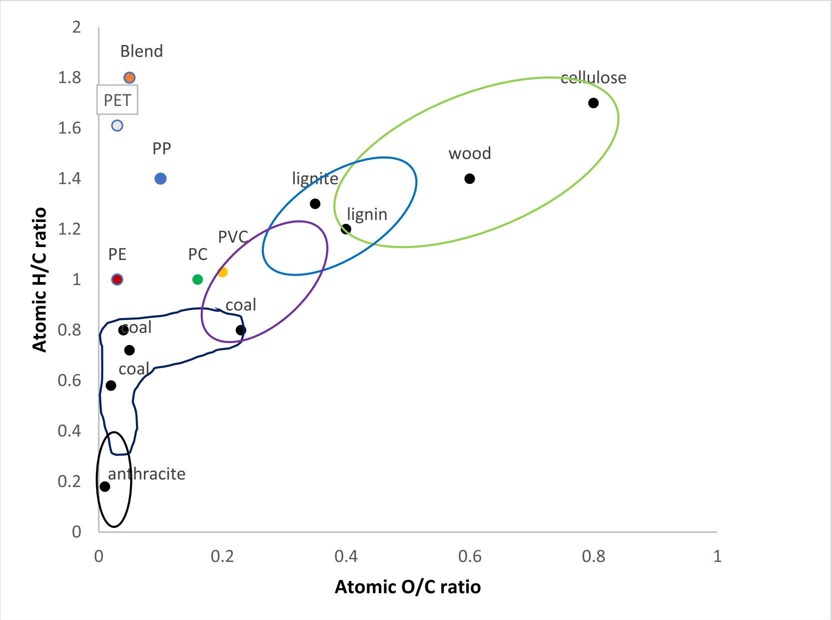

Fig. 7. Van krevelen diagram of plastic hydrochar from different polymer types. (PET & PVC [20], PE, PP [117], Blend (PP, PET, PP, Nylon [118]). The light green ring/circle indicates the biomass region; the second ring (blue indicates the peat region, the third ring (purple) indicates the lignite (low coal) region, the fourth ring (dark blue) indicates the sub-coal region, and the fifth ring indicates the anthracite coal region.

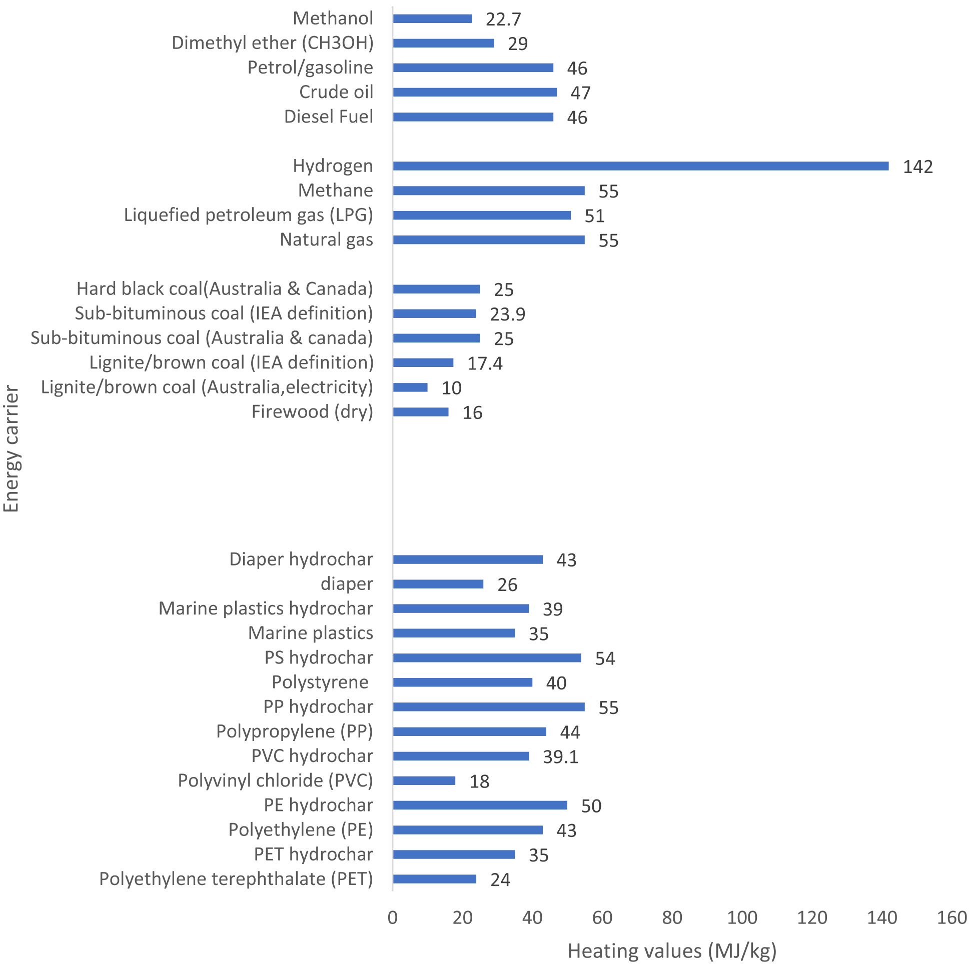

Fig. 8. Energy content of different energy carriers including plastics [47,122].

process, resulting in the formation of energetically favorable compounds in the hydrochar.

4.2. Hydrochar as solid recovered fuel (SRF)

4.2.1. Potential of hydrochar as SRF

Most petrochemically derived materials have a high energy content compared to coal and biomass. Hydrothermal carbonization conversion improves the energy quality of a material. In Iñiguez et al. [20], marine plastics were decomposed using HTC and the heating values increased from 35 to 39 MJ/kg. Also, PVC decomposed through HTC saw heating values increase from 18 MJ/kg to 39.1 MJ/kg [47]. Polypropylene and polyethylene have higher energy values than their counterparts, hence can serve as better feedstocks for hydrochar production through hydrothermal carbonization. The feasibility of plastic waste hydrochar as an energy source is dependent on the amount of energy required to offset the energy used in plastic production (plastic production processes are energy intensive and vary between plastic types and processes). The energy content (heating value) of a feedstock alone is not a measure of fuel quality, but the chemical compositions of the material, emissions and energy required for production also play a part. For example, in Fig. 9, although polypropylene has the highest heating value (44 MJ/kg), this does not mean it is a better solid-fuel feedstock than the others. Polyethylene has the highest energy quotient which translates to having the highest energy recovery potential. However, when these polymers are further processed into day-to-day materials such as bottles, plastic bags or cups, the total energy consumption will increase according to the plastic polymer types and processing methods (e.g., mixing, extrusion, molding, and thermoforming). Fig. 10, shows the energy used in the different PET production processes. This processing may alter the energy quotients. In addition, another important energy consideration is the amount of energy required during the plastic waste recovery or conversion process (HTC process).

In Marczak [120], PET plastic production uses over 71.2 MJ/kg in the production of a virgin polymer where 34.1 MJ/kg of energy was used in the extraction of raw materials (like crude oil) and 37.1 MJ/kg was used by fuels in the production of the polymer. A further 44 MJ/kg of energy was used in processing this virgin polymer into different shapes, sizes, and types etc., giving about 115 MJ/kg in energy input in total. In other words, producing 1 kg of usable PET product (having a chemical energy of 24 MJ/kg) from crude oil (having a chemical energy of 47 MJ/kg), requires 115 MJ. The energy content (heating value) of the final PET product (24 MJ/kg) is almost 5 times smaller than the overall energy required to produce it (from crude oil extraction to finished product).

If PET waste is decomposed using HTC, the energy content of the product (hydrochar) might be improved up to 35 MJ/kg (unpublished data). Shen et al. [121] reported that the heating values of waste PVC materials increase from 26 MJ/kg to 30.4 MJ/kg when blended with cellulose, 30.8 MJ/kg when blended with lignin and 29.1 MJ/kg when blended with xylan. Additionally, carbonizing plastic diapers under different temperatures increased the hydrochar heating values from 26 MJ/kg to 43 MJ/kg [122]. An integrated approach should therefore be developed that can recover the lost energy during plastic production process [119,123].

Since plastics have higher heating values than coal, plastic hydrochar could be used in coal kilns if they meet the required combustion properties. If the energy produced by plastic pulverized hydrochar as a solid recovered fuel (SRF) is injected back into the production process, 20 % of the greenhouse gases emitted in its production process will be reduced. Given that plastics generated over 1.8 billion tonnes of GHGs in 2019, accounting for over 3.4 % of global GHGs emissions, then considering the energy quotient of PET, PET energy recovery could reduce about 360 mT of GHG/year, while Fig. 11 illustrates the possible circular pathway where plastic waste can contribute in the production of plastic.

4.2.2. Combustion properties of SRF in coal furnaces

For coal furnace injection, material grindability and combustion characteristic index are some of the properties of interest. Material grindability is a measure of how easily a material can be ground or crushed [124]. This is mostly used in coal and coke quality estimates [125], so if hydrochar must be used in coal or coke furnaces, then its grindability must be similarly estimated. The Hardgrove grindability index (HGI) is a function of proximate analysis which can be used to measure the grindability of most carbonized materials. While most biomass hydrochars are obtained in a powdered form, plastics hydrochar is hard (just like raw coal) which can then be ground or used whole. The HG indices are inversely proportional to amount of energy needed to produce a grind hydrochar [126]. The HGI of the hydrochar is higher than that of anthracite coal (HGI 47.9). An example is the PET hydrochar reported in Ye et al. [117], where its HGI was 98.0, while that of the PVC hydrochar was 67.1. These findings suggest that the hydrochar aligns with the requirements for blast furnace injection. The combustion characteristic index measures the reactiveness of the fuel during combustion considering the burning velocity, ignition ease, and burnout

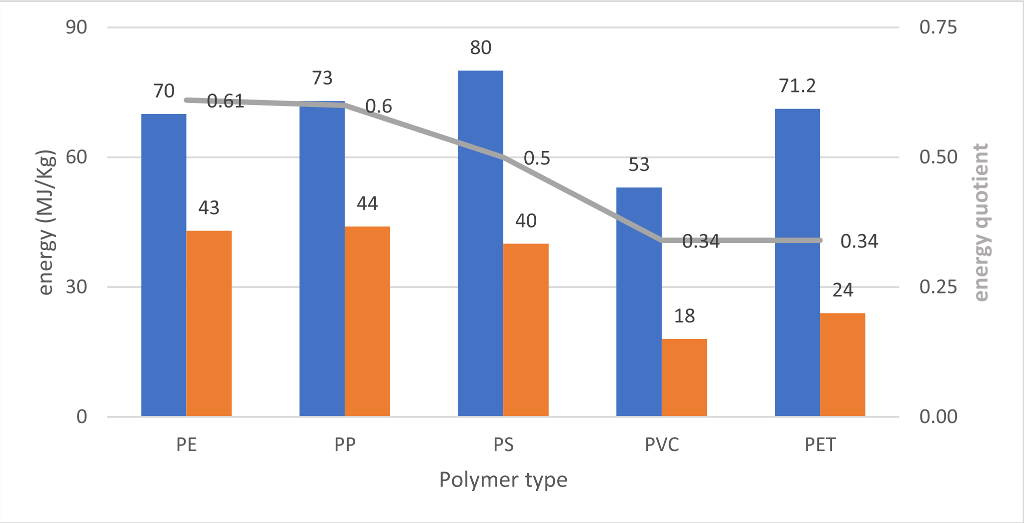

Fig. 9. Energy consumption to make virgin polymers with their calorific values and the energy quotient of the different plastic polymers. (🟦) Energy consumption (MJ/kg), (🟧) calorific value (MJ/kg) and the (—) energy quotient (right vertical axis).

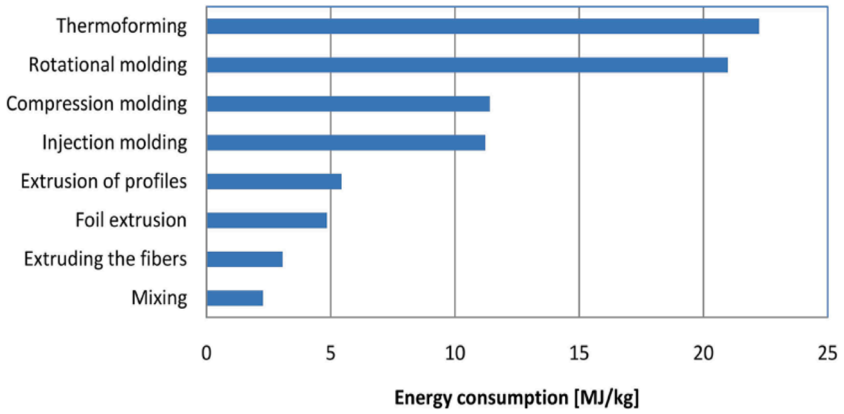

Fig. 10. Energy consumption of PET polymer into PET bottle [120].

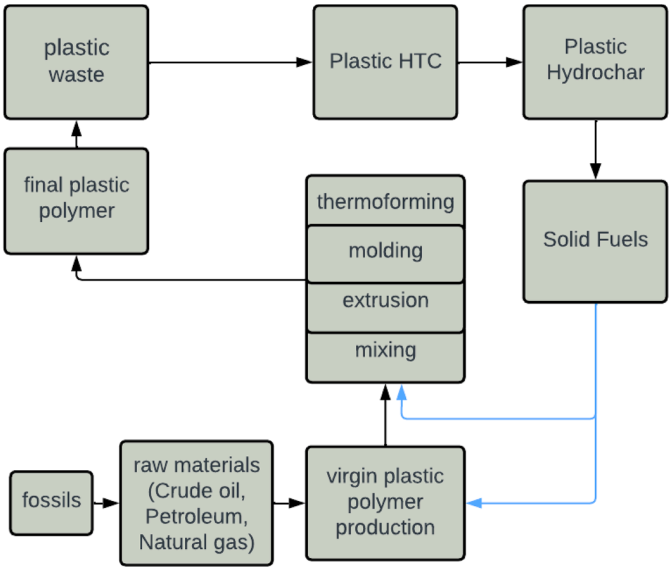

Fig. 11. Feedback loop showing the flow of energy produced by plastic hydrochar (in HTC) in the production of pure plastics. (🟦) Energy pathway from plastic hydrochar.

temperature [127]. Other combustion parameters necessary for blast furnace injection can be obtained from TGA/ DTG curves. These are ignition temperature T_i (°C), which is the temperature responsible for the weight loss of 1 % hydrochar/min, the peak temperature T_p (°C), and the burnout temperature T_f (°C), which is the temperature where the material stopped burning due to the absence of fuel [128]. There are other indices like the ignition index [129], burnout index, combustion performance index (S) [130], index of intensity (H_f), flammability index (C) [131], and critical heat flux (CHF) [132–135]. Materials with better fuel combustion behavior and performance are characterized by lower ignition and burnout temperatures, lower ignition and burnout index values, higher combustion performance index, higher index of intensity and flammability index [87]. Compared to coal, hydrochar tends to have lower ignition and burnout temperatures, lower ignition and burnout index, higher flammability index and index of intensity, higher combustion performance index. These properties suggest that hydrochar can serve as a good fuel, making it an attractive option for energy generation [136]. Hydrochar exhibits superior combustion performance compared to bituminous and anthracite coal, demonstrating exceptional reactivity in the raceway zone, enhancing fuel burnout. There is a significant difference in the combustion behaviors of waste plastic hydrochar and biomass hydrochar [87]. Plastic hydrochar is more reactive than biomass hydrochar possibly due to its possession of more oxygen-containing functional groups (such as hydroxyl (-OH), carbonyl ($C{=}O$), and carboxyl (-COOH) groups) shorter carbon chains, and lower aromaticity, increase porosity and high surface area [100]. These functional groups can promote easier breaking of chemical bonds and contribute to the release of volatile compounds.

4.3. Potential of hydrochar as supercapacitor

Given that plastic hydrochar shares similar properties with biomass hydrochar, there is potential for its use in supercapacitors as well. Various feedstocks, including wormwood, bamboo, hemp straw, *enteromorpha sp*, jatropha oilcake, carrageenan, rice husk, corn starch, fresh milk, salacca peel, shells of bamboo shoot, and rice straw have been utilized in supercapacitor experiments as hydrochar [99,137–142]. This suggests that both plastic and biomass-derived hydrochar may serve as promising materials for supercapacitor applications, showcasing the versatility of hydrochar across different feedstocks.

Supercapacitors have emerged as a promising energy storage technology due to their high-power density, fast charge/discharge rate and long cycle life [143]. However, the limited energy density is a major obstacle to widespread adoption. Various approaches have been explored to overcome this, including the use of advanced materials such as hydrochar [103,143]. Hydrochar shows immense potential for improving the performance of supercapacitors. Synthesis conditions such as temperature, reaction time, and feedstock composition can be optimized to tailor hydrochar properties to specific supercapacitor requirements [24]. As an example, adding $NaHCO_3$ to hydrochar enhanced the porosity leading to larger specific surface areas (>2300 $m^2\ g^{-1}$ and >3000 when $KHCO_3$ is used) with pore volume of approximately 1.4 $cm^3 g^{-1}$, which are indicative of high energy storage capacity in supercapacitors [24,144]. Hydrochar porosity can be modified by tuning synthesis parameters, resulting in improved ionic accessibility and electrolyte penetration by promoting charge transfer reactions and promoting adsorption of electrolyte ions [145]. Incorporating hydrochars into the supercapacitor electrodes significantly improved the electrochemical performance. The high specific surface area of the hydrochar increases electrode-electrolyte contact, resulting in improved ion diffusion and faster charge transfer [146,147]. Additionally, the unique porous structure of hydrochar provides ample space for storing ions, resulting in higher specific capacitance of 174.5 $F\ g^{-1}$ [146,148]. The high mesopore ratio, specific surface, and oxygen functional groups on the hydrochar surface also contributes to the increase in capacitance through pseudocapacitive reactions, further enhancing the overall performance of supercapacitors both as adsorbent [149] and an energy storage material [150]. Vijayakumar et al. [151] demonstrated the application of activated carbon fiber obtained from the carbonization of cotton waste. The cotton waste biomass contained few impurities hence was easy to purify. Plastic materials can provide a better carbon fiber for activation with KOH at 950 °C, although the presence of impurities (such as heavy metals) may require more effort in washing. However, more research is required on the conversion of plastic hydrochar into supercapacitors.

Various composite and hybrid systems have been developed to further optimize the performance of carbon-based supercapacitors [23]. By combining hydrochar with other carbonaceous materials such as graphene, carbon nanotubes, and activated carbon, the strengths of each component can be combined to enhance their electrochemical properties [22,152]. In Mu et al. [153] PET plastic waste was selectively transformed into three-dimensional porous carbon nanosheets (PCS) through carbonization, and then combined with MnO_2 nanoflakes to generate PCS-MnO_2 composite structures which generated a resilience of 5000 cycles of charge and discharge at a current density of 10 A g^{-1} with specific capacitance of 210.5 F g^{-1} and an areal capacitance of 0.33 Fm^{-2}. Additionally, hybrid systems combining hydrochar with transition metal oxides or conducting polymers have been investigated to achieve higher specific capacities and improved cycling stability. Ding et al. [146] added nickel to hydrochar as a graphitization catalyst, which effectively changed the graphitization degree of hydrochar and improved its specific capacity by 149 % with a specific capacitance of 174.5 F g^{-1}, higher than activated carbon/nickel hybrid. Liu et al. [154] added potassium catalyst to corn straw hydrochar, transforming it from amorphous structures to graphene-like sheets due to the increase in pore sizes. Supercapacitors from this graphene-sheet hydrochar had a very high electrochemical performance with a high specific surface area of 1781 m^2/g. Liu et al. [22], reported an outstanding electrochemical performance achieved during chemical activation process of hydrochar-derived carbon. The microstructure of the hydrochar aids the production of numerous graphene-like sheets with well-developed micropores, even with the application of minimal quantity of sodium-based catalysts. Another study [155] achieved the complete conversion of PET to solid fractions of carbon nanomaterials through hydrothermal synthesis. The supercapacitance performance showed a specific capacitance of 250.8 F/g, energy density of 34.83 Wh/kg, and power density of 999.9 W/kg with a current density of 0.5 A/g and exhibited a high cycle stability with high capacitance retention of 96.8 % and a current density of 1.5 A/g after 10,000 cycles. Dehydration and polymerization from hydrothermal carbonization can form hollow, delicate, functional carbon nanomaterials at temperatures lower than 200 °C under the presence of magnetic mediums like iron ions [156]. Hydrothermal carbonization can create controlled magnetic carbon nanocomposites (iron carbide nanoparticles, magnetite/carbon nano rods, carbon-coated magnetite nanoparticles, silver-coated iron oxide nanoparticles and graphene oxide) with improved structure, including high surface area, organized nano sizes, material stability, and magnetic properties [156].

Sustainable energy mobility is an important aspect of the transition to a greener and more sustainable future. By exploiting the unique properties of hydrochar, supercapacitors can improve the performance and efficiency of energy storage systems in a variety of sustainable transportation applications [157]. Carbon-based supercapacitors can contribute to sustainable energy mobility in several ways. For example, the incorporation of carbon-based supercapacitors into electric vehicles will increase energy storage capacity, enabling longer range and faster charging times [158]. Hydrochar-based supercapacitors can be used as energy buffers in HEVs, capturing and releasing energy during regenerative braking and acceleration. Hydrochar-based supercapacitors may offer high power density and fast charging capabilities, making them ideal for efficiently managing power fluctuations in hybrid vehicles, resulting in improved fuel efficiency and reduced emissions [157,159]. Hydrochar-based supercapacitors facilitate the integration of renewable energy sources such as solar and wind into the transportation sector. Using carbon-based supercapacitors in charging stations and infrastructure can optimize their performance and reduce the load on the power grid. By storing energy during times of low demand and releasing energy during times of peak demand, supercapacitors ease the grid, enable faster charging, longer driving range, and efficient energy management, supporting a more stable and efficient EV charging infrastructure. This contributes to a more sustainable green transportation sector and a sustainable transition to energy mobility.

4.4. Potential of hydrochar as catalyst for fuel production

Biodiesel generation is aided by a variety of catalysts, the most common of which are metal catalysts. However, biocatalysts and carbon-based catalysts, such as biochar or hydrochar, are preferred due to their lower cost, reusable nature, large surface area, stable structure, good thermal and mechanical stability, chemical inertness, improved acid density, and environmental friendliness [160–162]. Hydrochar has the potential to serve as a heterogeneous catalyst in the synthesis of biodiesel, achieved through surface modification with either acidic or basic groups [163,164]. For instance, sulfonated hydrochar, generated by exposing hydrochar to sulfur trioxide gas or sulfuric acid, demonstrates effective acidic catalytic properties in biodiesel production [165]. Alternatively, hydrochar can be infused with metal oxides like those of calcium, magnesium, or potassium, creating basic catalysts for biodiesel synthesis [163,166]. These catalysts offer numerous advantages over traditional counterparts, including notable catalytic activity, stability, reusability, and cost-effectiveness [88]. Furthermore, hydrochar can function as a catalyst support in various fuel production processes such as pyrolysis, gasification, and reforming [76,167].

Recyclable biochar-based catalysts show improved activity during simultaneous esterification and transesterification processes of non-edible oils and the inclusion of K and Fe boosts activity even further [168,163]. In Dharmalingam et al. [169], a biocatalyst with KOH blend produced maximum biodiesel. By sulfonating char with strong sulfuric acids, a carbon-based solid acid catalyst was created. Catalyzing the transesterification of vegetable oils and the esterification of free fatty acids were achieved and when the catalyst was treated with 10 M potassium hydroxide, the catalyst displayed increased transesterification activity due to porosity development, high acid density and increased surface area [170]. The researchers also explored the impact of alcohol to oil molar ratio, reaction time, and catalyst loading on the esterification reaction facilitated by the sulfonated biochar. They observed that increasing alcohol to molar ratio, reaction time, and catalyst loading led to higher conversion of free fatty acids [170]. Biodiesel yield of 94 % was achieved with 5 wt.% carbon-based catalyst loading in a microalgae biodiesel production [171]. In another study [172], peat-derived biochar was chemically activated with KOH to create cost-effective solid-base catalysts for transesterification. The modified catalyst showed significantly increased surface area and pore volume. The catalyst achieved a biodiesel yield of 93.4 % with 5 wt.% catalyst, 8:1 methanol/oil molar ratio, and 150 min reaction time. The stability of this catalyst was maintained after 10 cycles of use, with over 81.6 % yield. Considering that hydrochar has even higher surface area, pore volume and many other attributes than biochar, hydrochar has the potential to act as a better catalyst. Acidified hydrochar was reported in Ghalandari et al. [173] as a catalyst to convert waste PVC to biocrude through hydrothermal liquefaction. The addition of hydrochar to waste PVC leads to an increase in carbon and hydrogen contents and a decrease in oxygen content, thereby generating plastic crude oils with higher heating values [173].

Hydrochar catalysts could also be useful in hydrogen production. Global production of hydrogen is about 75 Mt per year as pure hydrogen and 45 Mt per year as part of mixed gases. This represents 3 % of the world's final energy demand [174]. Biochar as a catalyst has the potential to improve hydrogen output by up to 220.3 % and it is used in methane catalytic cracking and dry methane reforming, with hydrogen conversion rates ranging from 13.4 % to 95.7 % [175]. Steam reforming and water gas shift are significant steps widely used for producing hydrogen from hydrocarbons such as natural gas and biological feedstocks [176]. The presence of hydrochar or biochar catalysts can promote the gasification reactions, increase the yield of desired products, and reduce the formation of undesirable byproducts, such as tar or char [177]. Hydrochar catalysts can enhance the conversion efficiency, selectivity, kinetics, and stability of these processes, contributing to waste valorization and the production of renewable fuels [178]. Ma et al. [179] used biochar as a catalyst in the catalytic steam reforming of bio-oil compounds. The best results were achieved at 900 °C, leading to a high hydrogen yield (89.1 %) and concentration (76.0 %). The study also found that acid treatment reduced the catalytic activity of biochar by removing inherent acidic and alkaline earth metal species during the process. Also, the incorporation of biochar as a Ni support material presents an opportunity to reduce the overall reforming process expenses while promoting sustainability and thereby facilitating the augmentation of catalyst sites and functionalities [180]. This is done by harnessing the inherent surface oxygen-containing groups and alkali and alkaline earth metals. Carbon-derived catalysts have a strong degree of resistance to deactivation. In Harun et al. [181], biochar-derived catalyst was used and showed extended life (no deactivation for over 60 h) compared to other catalysts, due to carbon nanotube formation, high surface area of 3250 m^2 g^{-1} and its microporosity. Cotton-derived biochar catalyst has also been shown to increase production of hydrogen (64.0 vol%, 92.08 mg g^{-1} biomass) [177].

Hydrothermal carbonization has also been used to develop a platinum (Pt) catalyst for proton exchange membrane fuel cells (PEMFCs) using coconut shell-derived carbon [25]. The surface area of the catalyst is enhanced through thermal activation, making it suitable for catalysis with well-distributed Pt particles. The electrochemical surface area of the catalyst is like a commercial vulcan-based catalyst, although its oxygen reduction activity is slightly lower due to differences in carbon properties, such as graphitization degree, surface oxygen groups, and electrical conductivity. Increasing the peak temperature and retention time during the hydrothermal carbonization process reduces the polarity while enhancing the stability, thermal resistance, and aromatic nature of hydrochar materials [24]. However, hydrochar produced under lower peak temperatures and shorter retention times yields porous carbon with higher porosity.

4.5. Potential of hydrochar as a direct carbon fuel cell

Hydrochar holds significant promise as a fuel supply for direct carbon fuel cells (DCFCs), which offer distinct advantages compared to traditional fuel cells, including enhanced efficiency, reduced emissions, and lower costs [182]. DCFCs convert the chemical energy stored in a carbon-based fuel directly into electrical energy without an intermediate conversion step like hydrogen gas generation [183]. In this process, carbon-based fuels such as coal, coke, biomass, or even carbon-containing wastes are electrochemically oxidized at the fuel cell anode. The basic structure of a DCFC is like that of other fuel cells, consisting of an anode, a cathode, and an electrolyte [184] (Fig. 12). However, there are some significant differences in DCFC design and operation. Anodes are typically made of porous carbon materials that allow direct interaction between the carbon fuel and the electrolyte [185]. Electrolytes used in DCFCs are typically molten mixtures of carbonates or solid oxide materials that can operate at high temperatures [186]. When a carbon-based fuel is fed to the anode, an electrochemical oxidation process takes place. Carbon in the fuel reacts with oxygen ions in the electrolyte to produce carbon dioxide and release electrons [187]. The electrons then flow through an external circuit to produce an electrical current that can be used for various purposes. At the cathode, air or oxygen supplied by the system reacts with electrons and carbonate ions (for molten carbonate electrolytes) or oxygen ions (for solid oxide electrolytes) to produce oxygen gas [188] and regenerate the ions used in the anode. DCFCs have several advantages over other types of fuel cells. DCFCs are electrically efficient and can operate at high temperatures, enabling efficient cogeneration. They can use a wide range of carbon-based fuels, including low-grade and waste carbon sources, which could help reduce greenhouse gas emissions and improve the efficiency of carbon-intensive industries.

To date, there is little information on the utilization of hydrochar as a direct carbon fuel cell. However, hydrochar possesses similar properties to biochar, which has been shown in many studies as being suitable for direct carbon fuel cells. Biochar may be used as anode, cathode, and catalyst in fuel cells with a maximum power density of 0.4346 cm^{-2} [175]. DCFC verification experiments on rubber wood biochar and rice husk biochar produced a power density of 2.21 mW cm^{-2} and 0.07 mW cm^{-2} respectively. High fixed carbon, low ash content and the presence of oxygen functional groups favors the power density [189]. Another

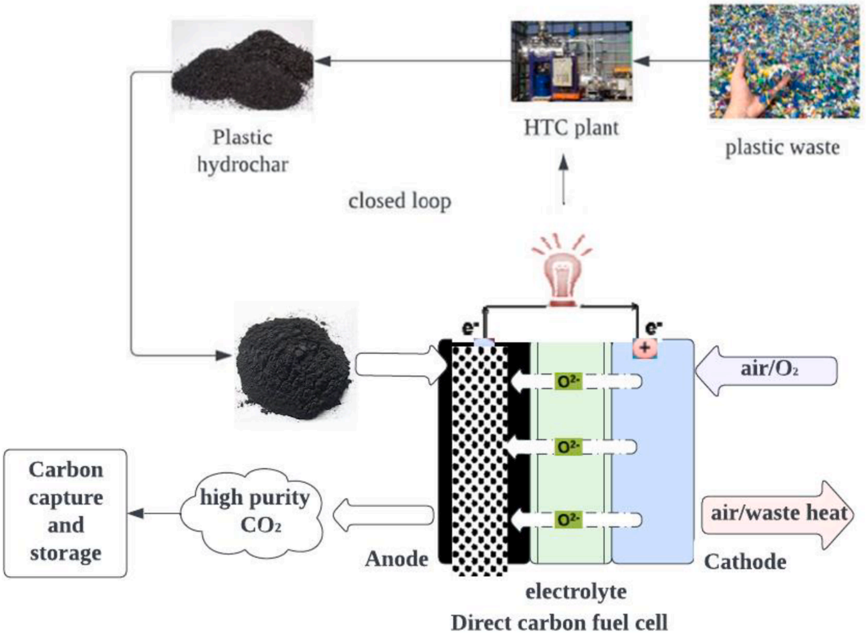

Fig. 12. Integration of hydrothermal carbonization technology (hydrochar) for direct carbon fuel cell (DCFC).

study determined the power density of three different biochar made of bare biochar and biochar-carbonate blend used as feedstock for direct carbon fuel. The findings revealed power outputs of up to 15.5 mW/cm^2 at 800 °C, owing to characteristics like porosity, acidity, volatile matter, carbon, hydrogen content and oxygenated surface functionalities [190]. In [191], researchers tested a bi-layer pellet for a dual-layer solid oxide fuel cell (DCFC) that utilized almond shell biochar as a power source. An enhancement in both the stability and performance of the cell was achieved by incorporating an NiO-SDC anode layer into the bi-layer pellet, which originally consisted of only a cathode and electrolyte. The introduction of the three-layer pellet cell at a temperature of 700 °C significantly improved the peak power density, from the initial value of 127 mW cm^{-2} to 150 mW cm^{-2}. Through the convergence of two cutting-edge technologies, plastic hydrochar and direct carbon fuel cells (DCFCs) hold immense potential for transforming the landscape of sustainable energy (Fig. 12).

5. Challenges and research prospects of plastic-based hydrochar

The practicality of using plastics for hydrochar remains understudied and lacks comprehensive data [192]. Very limited studies exist on the energy balances, emissions, carbon offset, optimization, and overall cradle-to-grave analysis of hydrothermal carbonization of plastic waste [192,193], with most research focused on the HTC of biomass and PVC dechlorination. Even many of the studies on HTC have been done at lab scale levels (ranging around 0.2 to 2-L reactor volume) which are often limiting in terms of reactor design, size, pressure, reaction time etc.

The quality and consistency of plastic hydrochar as an SRF can vary based on the type of plastic feedstock and the hydrothermal carbonization process. Inconsistent hydrochar quality can lead to variations in energy content and combustion performance. The energy content or calorific value of plastic hydrochar should be optimized to ensure efficient combustion, but achieving a consistent and high calorific value across different batches of hydrochar can be challenging in practice. The combustion properties of plastic hydrochar, such as ignition temperature, flame characteristics, and emissions profile might differ from traditional solid fuels, and so understanding and managing these properties will be crucial for ensuring safe and efficient combustion. An additional complication is that plastic waste can often be contaminated with various substances, including non-combustible materials, heavy metals, and other pollutants. During the HTC conversion process, these additives may undergo transformations, potentially leading to the generation of by-products that can be challenging to manage. Some additives may resist complete decomposition, affecting the quality of the hydrochar produced. Contaminants in plastic waste may interfere with the intended energy recovery processes. For example, the presence of certain substances may alter the combustion characteristics, leading to incomplete combustion and reduced energy output. Additives or impurities in plastic waste can contribute to the corrosion or erosion of equipment, affecting the longevity of processing units and increasing maintenance requirements.

At a practical level, incorporating plastic hydrochar into existing solid fuel combustion systems might require slight modifications to equipment and processes to accommodate its unique characteristics. Finally, combustion of plastic hydrochar can result in unwanted emissions of greenhouse gases, particulate matter, and other pollutants. Assessing the economic feasibility of using plastic hydrochar as a SRF involves full consideration of the costs of production, processing, transportation, and compliance with regulations, as well as potential revenue from energy generation. Understanding the stability of plastic hydrochar during storage, transportation, and handling is also crucial to prevent degradation and ensure consistent combustion performance.

Hydrochar faces challenges as a catalyst, including variability in catalytic activity and selectivity, potential instability and durability issues, concerns about pore structure and accessibility, the risk of catalyst deactivation over time, and the need for effective regeneration methods. Reusability and scalability pose additional challenges, along with the complexity of understanding reaction mechanisms. Despite these obstacles, ongoing research aims to address these issues and optimize performance of hydrochar as a catalyst in diverse applications.

Hydrochar, like biochar, holds promise as a carbon source for supercapacitors, but challenges still exist. Ongoing research is focused on tailoring hydrochar synthesis methods for specific supercapacitor requirements. Other priorities are scaling up hydrochar production for large-scale use and investigating long-term stability. Supercapacitor performance relies on the quality, structure, and properties of plastic hydrochar-derived carbon, which must exhibit high surface area and conductivity. Contaminants from plastic feedstock and carbonization can impede conductivity and reduce capacitance. Achieving optimal specific capacitance and energy density with hydrochar-derived carbon presents difficulties due to non-optimized carbon properties. Designing efficient electrodes requires careful optimization, ensuring adhesion, uniformity, and ion accessibility. Long-term supercapacitor stability over charge-discharge cycles depends on plastic hydrochar resistance to degradation and capacity loss. Electrolyte compatibility is crucial; certain plastic hydrochar-derived carbons may hinder ion diffusion or cause electrolyte breakdown. Cost competitiveness with other supercapacitor materials also must be considered. Comparing plastic hydrochar-derived supercapacitors with established materials is vital to demonstrate performance viability.

The emergence of DCFC technology marks a significant advancement in energy conversion, yet it remains in its nascent stages. Incorporating hydrochar into DCFCs presents constraints, including the necessity for elevated operating temperatures, the occurrence of carbon deposits, and low power density. To overcome these obstacles, research is needed to investigate the diverse approach to enhance the performance of hydrochar-based DCFCs including methods like pre-treatment, activation, functionalization, and hydrochar doping. Some of the challenges to be addressed include the demand for durable and efficient electrode materials, alongside the need to manage carbon dioxide emissions. Plastic waste carries various contaminants, including additives and dyes, which can undermine performance by clogging electrodes and impairing efficiency due to the formation of residues. During operation, electrodes crafted from plastic hydrochar may be vulnerable to fouling and degradation due to impurities and contaminants, which can lead to diminishing electrode performance, in turn requiring frequent cleaning or replacement as mitigation. Furthermore, the carbonization process for plastic hydrochar can yield gas and tar byproducts, potentially obstructing electrode pores and hampering fuel cell efficiency. The variability in carbon quality can lead to uneven power generation and reduced overall efficiency. Striking the balance between optimizing the carbonization process for plastic hydrochar and ensuring the generation of high-quality carbon suitable for DCFC application presents a significant research avenue. Designing and fabricating electrodes compatible with hydrochar-derived carbon material is another challenge, demanding both conductivity and stability. Transitioning from laboratory-scale research to large-scale production and implementation raises challenges of maintaining consistent quality, scalability, and cost-effectiveness. Ensuring the stability of plastic hydrochar carbon over the long term is essential for the sustained performance and durability of DCFC systems. Factors such as material degradation, corrosion, and structural changes must be meticulously assessed. Finally, even if these technical hurdles are overcome, the regulatory and safety landscape also comes into view, where the utilization of plastic waste and the hydrochar production process could trigger concerns related to emissions, waste management, and potential hazardous releases.

Due to concerns of microplastics, research should focus on complete decomposition of plastic hydrochars, to minimize the potential leaching of microplastics and any impacts on soil or water quality, to ensure safe and sustainable use.

6. Conclusions

Hydrothermal carbonization offers several advantages over alternative processes such as hydrothermal liquefaction (HTL) or gasification (HTG). The simplicity of hydrothermal carbonization, and its lower operating temperatures, in contrast to the more complex hydrothermal liquefaction or gasification processes, contributes effectively to minimizing energy consumption and operating costs. It also boasts higher carbon conversion efficiency, leading to increased yields of hydrochar. Hydrothermal carbonization produces hydrochar with lower emissions, thereby mitigating the environmental impact associated with waste management and energy production. Hydrothermal carbonization has emerged as a promising technique for transforming plastic waste into valuable products, with recent studies reporting promising results, suggesting the cost effectiveness and efficiency of hydrothermal carbonization. The suitability of hydrothermal carbonization of plastic waste conversion has been highlighted, and a range of potential hydrochar applications, including solid recovered fuels, catalysts, direct carbon fuel cells, and supercapacitors, have been presented. However, the practicality of using plastics for hydrochar remains understudied and lacks comprehensive data. The quality and consistency of plastic hydrochar as an energy source, the feedstock variability, contamination, scalability, material properties, and environmental considerations are some of the challenges of utilizing plastic hydrochar across different applications. Additionally, understanding the stability of plastic hydrochar during storage, transportation, and handling is crucial to prevent degradation and ensure consistent combustion performance. Despite these challenges, hydrothermal carbonization of plastic waste holds great promise for accelerating the trajectory towards a sustainable environment. Addressing the challenges associated with plastic hydrochar will be crucial in realizing the full potential of hydrothermal carbonization as a sustainable solution for addressing both the environmental issues associated with plastic waste and the need for alternative energy applications. The critical evaluation of the applications of hydrothermal carbonization can contribute to advancing sustainable waste management and renewable energy production.

CRediT authorship contribution statement

Clovis Awah Che: Conceptualization, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Philippe M. Heynderickx:** Funding acquisition, Project administration, Resources, Supervision, Validation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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