

New Trends in Pyrolysis Methods: Opportunities, Limitations, and Advantages



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Abstract The expanding demand for environmental treatment increasingly requires different types of engineered char with high performance. In this context, new trends in pyrolysis methods have emerged and contributed to the sustainable development of pyrolysis technologies, namely microwave-assisted pyrolysis, co-pyrolysis, pyrolysis under non-inert ambiances, hydrothermal carbonization (wet pyrolysis), and integrated pyrolysis techniques. The outstanding advantages of these technologies over conventional pyrolysis include: increase in biomass conversion efficiency of the process, use of nonconventional raw material, increase in adsorption capacity of biochar by enhanced activated oxygen species, porosity, and functional groups, and removal or immobility of contaminants. The biochar products can be widely applied in various environmental fields, such as carbon capture and sequestration, soil amendment, adsorption of contaminants in soil, water, and air, and energy production. Nevertheless, challenges remain for these new trends, such as the increase in cost for the installation and operation, the lack of knowledge of the mechanism involved during pyrolysis, the difficulty in scaling up, etc. Further studies are recommended to facilitate the application of these new trends, such as pilot tests or field experiments to evaluate the real effects of biochar products prior to large-scale applications or their long-term risk during use, or prediction of properties of biochar and their impacts on environmental applications using modeling or machine learning approaches.

Keywords Advantages · Biochar · Limitation · New trends · Opportunities · Pyrolysis

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1 Introduction

Although widely applied in biochar production, products from conventional pyrolysis are increasingly unable to meet demands in energy production and environmental applications. The expansion of the raw materials for biochar production, as well as the need to create biochar with new properties with high adsorption capacity and environmental safety, are increasingly focused (Mohammadi et al. 2020). In addition, optimizing performance with maximal energy saving represents a crucial factor for the sustainable development of pyrolysis technologies (Veiga et al. 2020). In this context, new trends in pyrolysis methods, namely co-pyrolysis (Veiga et al. 2020; Rodriguez et al. 2021), microwave-assisted pyrolysis (Motasemi and Afzal 2013; Fang et al. 2021), (Yu-Fong et al. 2015; Veiga et al. 2020), pyrolysis under non-inert ambiances (SASAKI et al. 2009; Shen et al. 2017; Mian et al. 2018), hydrothermal carbonization (wet pyrolysis) (Zhou et al. 2019; Olszewski et al. 2020), and integrated pyrolysis (Yek et al. 2019, 2020; Liu et al. 2020) have emerged with the potential of synthesizing engineered biochar with enhanced properties and better quality (Fig. 1). These new pyrolysis conditions affect both physical properties (e.g.,

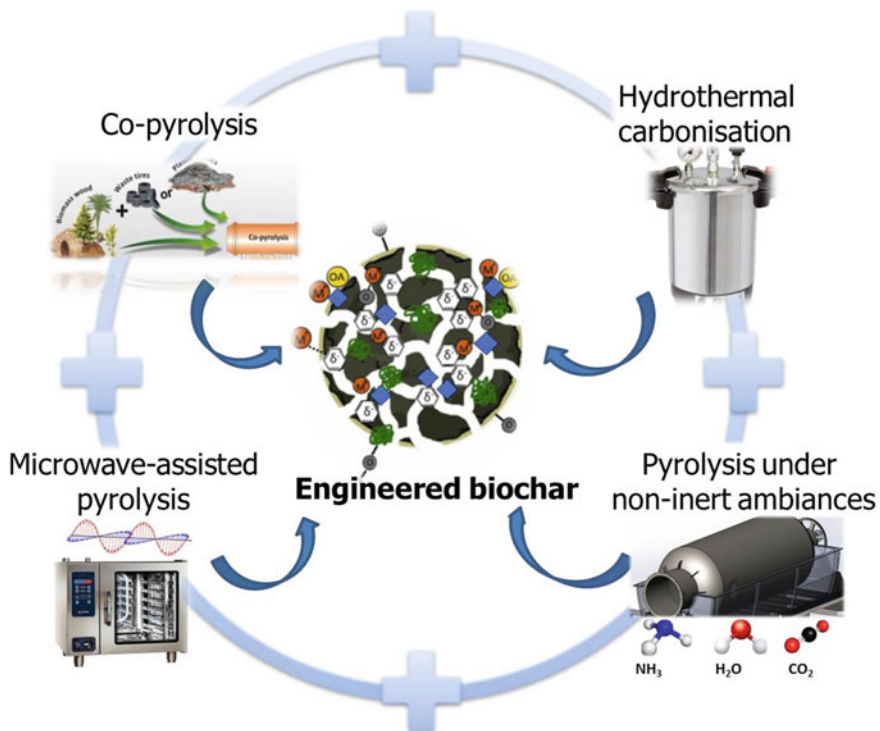


Fig. 1 New trends in pyrolysis of biomass

increase of specific surface area and porosity) and chemical properties (e.g., the introduction of functional groups and induce activated oxygen species on biochar surface) of the biochar, promoting the adsorption or degradation of different contaminants depending on the specific approach employed.

This chapter aims to summarize new trends in pyrolysis methods. Recent studies, advantages, and limitations/challenges related to each method are deeply discussed. Several research directions to facilitate the application of these new trends are also proposed.

2 Principle of New Pyrolysis Methods

2.1 *Co-pyrolysis*

Co-pyrolysis is a process that involves two or more feedstock as raw materials. For biochar applications, the biomass can be mixed with various types of other materials, from other biomass to municipal and industrial wastes (Johannes et al. 2013; Sewu et al. 2019; Rodriguez et al. 2021). The principle of co-pyrolysis is very similar to conventional pyrolysis. Prior to co-pyrolysis, the samples should be dried and well mixed to ensure an adequate homogeneity of the raw material. The ratio of raw material is an important parameter in co-pyrolysis that defines the quantity/quality of the biochar produced (Yao et al. 2021). The important key to the success of this technique is the contact of the different raw materials during co-pyrolysis (Fei et al. 2012; Johannes et al. 2013; Diao et al. 2021). Synergistic effects can be achieved through a series of radical interactions during the co-pyrolysis, namely initiation, formation of secondary radicals (depolymerization, formation of monomers, favorable and unfavorable hydrogen transfer reactions, intermolecular hydrogen transfer, isomerization via vinyl groups), and termination by disproportionation or recombination of radicals (Önal et al. 2014). Synergistic effects during co-pyrolysis can be complicatedly varied, depending on the type and contact of components, pyrolysis conditions (residence time, temperature, and heating rate), removal or equilibrium of volatiles formed, addition of catalysts, etc. (Fei et al. 2012; Johannes et al. 2013).

2.2 *Microwave-Assisted Pyrolysis*

Intrinsic drawbacks of conventional pyrolysis systems can be listed as heat loss to the environment, heat transfer resistance of materials, nonselective heating, a short lifetime of the system due to continuous heating, etc. (Salema and Ani 2011; Nan et al. 2015). Due to these limits, microwave-assisted pyrolysis (Fig. 2) is proposed as a novel method to effectively improve pyrolysis performance by enhancing chemical reactions with the help of heat transfer profiles through microwave irradiation

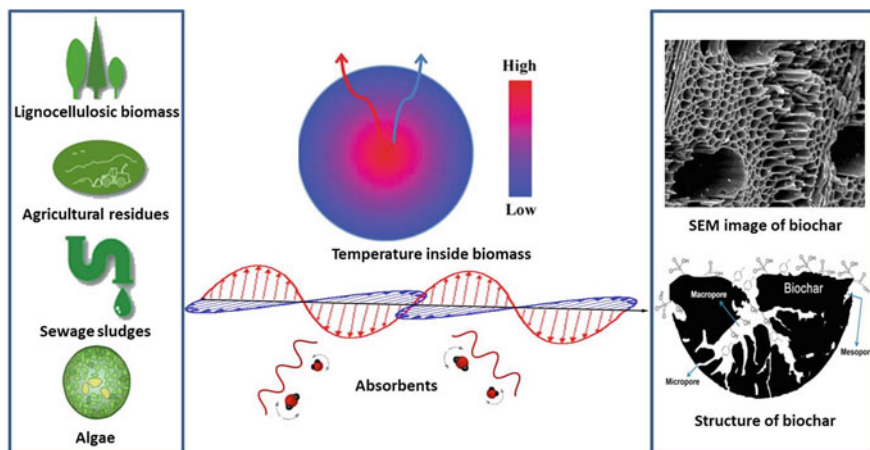


Fig. 2 Illustration of microwave-assisted pyrolysis of biomass

(Lidström et al. 2001; Fang et al. 2021). Microwaves fall in between the infrared and radio wave regions of the electromagnetic spectrum, i.e., wavelengths ranging from 0.01 to 1 m, corresponding to the range of frequency between 0.3 and 300 GHz (Motasemi and Afzal 2013; Calles-Arriaga et al. 2016). Most industrial microwave-assisted pyrolysis processes are conducted at the range of frequency between 0.915 and 2.45 GHz, which are assigned for heating applications (Mushtaq et al. 2014). Several reviews exist in the literature, highlighting the importance and the advantages of microwave-assisted pyrolysis (Lidström et al. 2001; Luque et al. 2012; Motasemi and Afzal 2013; Huang et al. 2016; Zaker et al. 2019).

The heating mechanism of microwave heating is mainly attributed to the interaction of dipoles or ions in the electric field, known as the dielectric response (Guerra et al. 2006; Taqi et al. 2020). The heating medium or reactants absorb electromagnetic energy from microwaves volumetrically to achieve self-heating in a uniform and rapid way. This contributes to a rapid moisture release from the raw material, improving its specific surface area and pore structure (Zbair et al. 2018; Taqi et al. 2020). A vital property of microwave heating is the formation of hot spots, as a result of the heterogeneity of the microwave field or dielectric characteristics of the material, generating a much higher inner than outer temperature (Vadivambal and Jayas 2010; Wang et al. 2016; Zhang et al. 2018). The temperature of the hotspots can significantly increase the pore creation inside the biomass.

Commonly, quartz reactors are employed inside a microwave chamber where biomass is placed. The microwave-assisted pyrolysis system is usually purged with N_2 , He, or CO_2 to ensure a limited-oxygen environment (Zubairu et al. 2012; Lam et al. 2017). As biomass is often a poor absorbing material, microwave absorbers (also called dielectrics) are often homogeneously blended with the raw material before pyrolysis, to ensure a stable heat transfer and a high enough pyrolysis temperature inside the reactor (Ethaib et al. 2020). The selection of a suitable microwave

absorber is the key to the improvement of biochar characteristics. Similar to conventional pyrolysis, operational parameters can greatly affect the biochar yield and characteristics during microwave-assisted pyrolysis, such as microwave power—which has a direct link with pyrolysis temperature, or residence time (Mushtaq et al. 2014; Huang et al. 2016). Other than that, the types of absorbers and/or catalysts mixed with the raw material also determine the properties of the char produced (Fang et al. 2021).

2.3 *Pyrolysis Under Non-inert Ambiances*

Unlike conventional pyrolysis which often uses inert gases (especially N_2) to form an oxygen-limited environment, the use of non-inert atmospheres such as steam, CO_2 , or NH_3 during pyrolysis has emerged as a promising trend for pyrolysis (Lee et al. 2017a; Chen et al. 2018; Grottola et al. 2019). As opposed to post-pyrolysis activation using reacting agents at high temperatures (usually superior to $700\text{ }^\circ\text{C}$), steam, CO_2 , or NH_3 are introduced from the beginning of the pyrolysis process at low temperatures to create a non-inert atmosphere. Flow rates and concentrations of these gases are well controlled to obtain desired pyrolysis conditions suitable for each type of raw material. An example of a Macro-TGA system for investigating pyrolysis under different atmospheres is presented in Fig. 3. The reactor, containing a ceramic tube (1), is placed in an electrical furnace with three independent heating zones (T_1) to maintain a uniform temperature. The pyrolysis atmosphere is generated by a mixture of N_2 and a pyrolysis agent (CO_2 , NH_3 , or H_2O) in selected proportions. Gas flows can be precisely controlled by mass flow meters (M_1). The gas mixture at atmospheric pressure moves across a 2-m-long coiled tube (3) located in the upper part of the reactor before reaching the sample. Then the sample holding (4) containing the sample is lifted from the bottom to the desired position.

Compared to conventional pyrolysis, pyrolysis under non-inert ambiances offers some special characteristics. Steam can efficiently penetrate the biomass during pyrolysis, enhancing desorption, distillation, and efficient removal of the pores (Pütün et al. 2006; SASAKI et al. 2009). Meanwhile, the CO_2 atmosphere hinders polymerization reaction and secondary char formation by reacting or cracking tar compounds that may lead to its formation (Choi et al. 2020; Lee et al. 2020). In addition, CO_2 has an affinity to react with hydrogenated and oxygenated groups, resulting in a carbon-rich char (Shen et al. 2017). Regarding NH_3 -assisted pyrolysis, a significant activation of N-containing surface functional groups on biochar occurs as NH_3 breaks down to some radicles of NH_2 , NH , and H^+ and they chemically react with the biochar surface oxides groups (Mian et al. 2018).

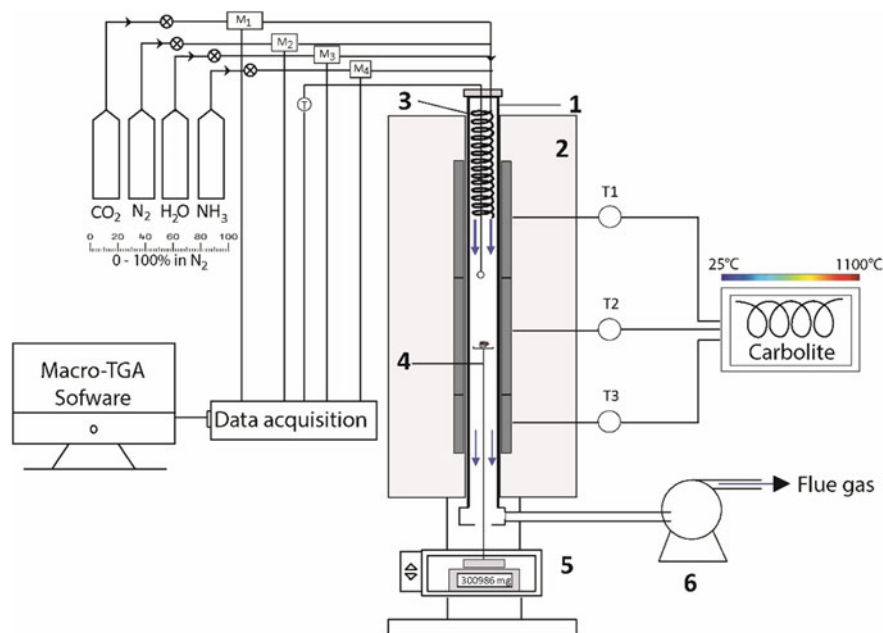


Fig. 3 A Macro-TGA system for pyrolysis under non-inert ambiances. The desired atmosphere is generated by different gases (CO_2 , H_2O , NH_3 , or a mixture of each with N_2) and regulated by flow meters (M_i). (1) Ceramic tube, (2) Electrical furnace, (3) 2-m coiled preheater, (4) Sample holder, (5) Weighing scale, (6) Extractor, (T_i) Regulation thermocouples

2.4 Hydrothermal Carbonization (Wet Pyrolysis)

During the hydrothermal carbonization (or wet pyrolysis) process, the raw material is heated in subcritical water (between 100 and 374 °C) or supercritical water (above 374 °C) at autogenic pressures in a high-pressure reactor (Ischia and Fiori 2021). Many similar reaction mechanisms occur during both dry pyrolysis and wet pyrolysis. Macro-molecules degrade to form liquid and gaseous products, while solid–solid interactions led to a rearrangement of the original structure (Libra et al. 2011). However, the difference in the pyrolysis atmosphere plays a defining role in the characteristics of the resulted biochar. The degradation of biomass during wet pyrolysis is initiated by hydrolysis, which exhibits lower activation energy than most of the pyrolytic decomposition reactions. Therefore, the principle biomass components are less stable under wet pyrolysis, leading to lower decomposition temperatures. Hemicelluloses decompose between 180 and 200 °C, most of the lignin between 180 and 220 °C, and cellulose above approximately 220 °C (Hydrothermal degradation of polymers derived from plants 1994). For comparison, in dry pyrolysis of biomass, hemicellulose generally decomposes at temperatures between 220 and 315 °C, cellulose between 315 and 400 °C, and lignin between 160 and 900 °C (Yang et al. 2007). This method actively introduces hydroxyl and carboxyl functional groups onto the

surface of biochar, improving the biochar contaminant adsorption. Moreover, with such low process temperature and high-pressure conditions, most of the organics in the input raw material remain, and/or are transformed into solids, and only a little gas is produced (<5%) (Libra et al. 2011). Therefore, hydrothermal carbonization is highly suitable for the production of biochar.

2.5 Integrated Pyrolysis Techniques

Several recent attempts seek to combine the above-mentioned techniques, namely microwave-assisted hydrothermal (co-)pyrolysis and microwave steam/CO₂ (co-) pyrolysis. During microwave-assisted hydrothermal carbonization, the raw material is subjected to microwave heating to obtain moderate temperatures (180–250 °C) in a tight-closed space for a short reaction time (dozens of minutes to a few hours). The decomposition of biomass then occurred under high pressures (2–10 MPa, usually autogenously). Meanwhile, in microwave steam/CO₂ (co-) pyrolysis, microwave heating is initially conducted to transform biomass into a precursor, after which pyrolysis was performed under a CO₂ or steam atmosphere (Yek et al. 2019, 2020). Lots of complex reactions take place during these processes, such as dehydration, decarboxylation, hydrolysis, polymerization, poly-condensation, and aromatization. With these techniques, water inside biomass has good absorption properties of microwave energy due to its high dielectric constant (δ') and loss tangent ($\tan \delta$), which help in increasing the heating rate and reduce the energy consumption for the process (Tsubaki et al. 2012, 2016; Antonetti et al. 2015).

3 Advantages of New Pyrolysis Methods

3.1 Co-pyrolysis

The co-pyrolysis technique could significantly improve the biochar yield and its quality without any improvements in the system process (Yao et al. 2021). Some minor modifications may be needed, but only for the feed preparation system. The interaction or synergy between agricultural and industrial materials in co-pyrolysis can improve certain properties of biochar, such as reduction of electrical conductivity, increase in water-holding capacity, neutralizing power, stability, and enabling the release or concentration of macro and micronutrients (Rodriguez et al. 2021).

Another important advantage of co-pyrolysis lies in the synthesis of novel biochar composites. Sewu et al. (2019) produced a biochar composite from the co-pyrolysis of bentonite and kelp seaweed (Sewu et al. 2019). The product had a higher carbon sequestration potential and adsorption capacity of dye than the kelp biochar. In another study, biochar/layered double hydroxides composite was synthesized using

co-pyrolysis after pre-loading MgAl-layered double hydroxides on the surface of rice husk powder through precipitation (Lee et al. 2019). Results showed a significant enhancement of phosphate adsorption due to the effective adsorption of anionic contaminants by the biochar through ion exchange with negatively charged groups located between hydroxide layers. Co-pyrolysis of cigarette industry-based waste with bentonite and calcite (5:1 w/w) at 700 °C helped in enhanced Pb removal in comparison to the control biochar with no mineral additives (Ramola et al. 2020). Similarly, rice husk and calcite (4.2:1 w/w) co-pyrolyzed together imparted synergistic effects for superior adsorption capacity for phosphate removal at lower concentration in comparison to control rice husk biochar and calcite separately (Ramola et al. 2021).

Table 1 presents some recent studies on co-pyrolysis of biomass, the effect of co-pyrolysis on the resulted char, and its applications.

3.2 *Microwave-Assisted Pyrolysis*

High heating rates obtained during the process are a major advantage of microwave-assisted pyrolysis, as this can help produce a type of biochar with a higher surface area and pore volume than those obtained with conventional pyrolysis (Luque et al. 2012; Zhang et al. 2017b; Zaker et al. 2019). Furthermore, it is indicated that biochar obtained from this technique achieves a high uniformity and cleanness (Yagmur 2012; Huang et al. 2016). This method also comfortably accepts the addition of cheap absorbers or catalysts, or the blending of materials together during pyrolysis without any modification of the system to create biochar products with better adsorption capacity (Chen et al. 2008; Fang et al. 2021). Some up-to-date microwave-assisted pyrolysis studies are presented in Table 2.

3.3 *Pyrolysis Under Non-inert Ambiances*

This technique is relatively energy-efficient and eco-friendly compared with other approaches aiming at improving the adsorption capacity of the biochar (e.g., pyrolysis at high temperatures, catalyst pyrolysis). Steam has the effect of removing tar and other trapped products of incomplete combustion during pyrolysis on the surface of biochar, creating a clean final product (SASAKI et al. 2009; Kurian et al. 2015). Meanwhile, CO₂-assisted pyrolysis helps simultaneously improve the quality of pyrolysis gas and biochar produced (Lee et al. 2017b). In a recent study, the CO₂-assisted pyrolysis of teabags increased the gas yield, particularly hydrogen, and prevented the formation of pollutants (e.g., phenolic compounds, benzene derivatives, and polycyclic aromatic hydrocarbons) (Lee et al. 2021). The biochar product also had a high calorific value (HHV = 26.8 MJ kg⁻¹) comparable to that of coal. Regarding NH₃-assisted pyrolysis, it can be considered a novel way to synthesize

Table 1 Some recent studies in co-pyrolysis of biomass

Raw material	Pyrolysis conditions	Effect on biochar properties	Application	References
Dried municipal sludge and tea waste	<ul style="list-style-type: none"> - Temperature: 300 °C in 2 h - Raw material ratio: 1:1 	<ul style="list-style-type: none"> - Enhanced active adsorption sites - Formation of some new aromatic groups 	Cadmium removal from aqueous solution	Fan et al. (2018)
Agricultural wastes (poultry litter, swine manure) and industrial wastes (construction wood, tire, PVC plastic)	<ul style="list-style-type: none"> - Temperature: 300-700 °C - Raw material ratio: 1:1 	<ul style="list-style-type: none"> - Low H:C (0.06) and O:C (0.30) molar ratios - Reduction of electrical conductivity - Increase in water-holding capacity, neutralizing power, and stability 	Carbon sequestration in soil, soil quality improvement	Rodriguez et al. (2021)
Rape straw and phosphate rock	<ul style="list-style-type: none"> - Temperature: 500 °C in 2 h - Heating rate: 10 °Cmin⁻¹ - Raw material ratio: 5:1 to 2:1 	<ul style="list-style-type: none"> - Increase in carbon retention (up to 27.5%) - Decrease in aromaticity and graphitization - Positive effect on Pb removal capacity with a low additive amount of phosphate rock (ratio 5:1) 	Improvement of carbon sequestration and enhancement of Pb removal	Gao et al. (2019)
Seaweed-rice husk-pine sawdust	<ul style="list-style-type: none"> - Temperature: 500 °C in 1 h - Raw material ratio: 7:3 	<ul style="list-style-type: none"> - Enhancement of thermal stability, aromaticity, pH balance, ash content, and yield 	Soil ameliorant, contaminant remediation	De Bhowmick et al. (2018)
Bituminous coal and wheat straw Wu et al. (2019)	<ul style="list-style-type: none"> - Temperature: 950 °C in 0.5 h - Raw material ratio: 3:1 - Heating rate: 10 °C min⁻¹ 	<ul style="list-style-type: none"> - Increase in specific surface area - Inhibitory of the ordering and uniformity of microscale structure 	Enhancement of kinetics during biomass gasification	Wu et al. (2019)

Table 2 Recent studies on microwave-assisted pyrolysis of biomass

Raw material	Pyrolysis conditions	Effect on biochar properties	Application	References
Empty fruit bunch	<ul style="list-style-type: none"> - Power: 2.6 kW - Frequency of 2.45 GHz - Temperature: 253 °C - Duration: 90 min 	<ul style="list-style-type: none"> - High HHV (26.4 MJ kg⁻¹) - Higher O/C and (N + O)/C ratios - Stable burning characteristics 	Solid fuel in power generation to substitute coal	Azmi et al. (2019)
Switchgrass	<ul style="list-style-type: none"> - Power: 700 W - Frequency: 2.45 GHz - Temperature: 300 °C - Catalyst: K₃PO₄, clinoptilolite, bentonite, and their mixtures (catalyst-to-biomass ratio 10–30%) 	<ul style="list-style-type: none"> - Increase in specific surface area and plant nutrient contents - High sorption affinity and high cation exchange capacity 	Improvement of water-holding capacity and fertility of sandy soil	Mohamed et al. (2016)
Sugarcane bagasse	<ul style="list-style-type: none"> - Power: 600 W - Duration: 30 min - Catalyst: Ferric oxide 	<ul style="list-style-type: none"> - Increase of porous structures and pore size - Increase of specific surface area - Ferromagnetic properties observed 	Adsorbent for the removal of Cd ²⁺ from aqueous solutions	Noraini et al. (2016)
Soapstock	<ul style="list-style-type: none"> - Power: 1000 W - Frequency: 2.45 GHz - Temperature: 550 °C - Bentonite: soapstock ratio: 1:1:2 	<ul style="list-style-type: none"> - Decrease in carbon content, increase in ash content and porosity - Increase in removal efficiency of Cd²⁺ compared to biochar without catalyst (77.9% vs. 50.5%) 	Adsorbent for the removal of Cd ²⁺ from aqueous solutions	Dai et al. (2017)
Horse manure	<ul style="list-style-type: none"> - Power: 1000 W - Frequency: 2.45 GHz - Catalyst: coconut shell-based activated carbon 	<ul style="list-style-type: none"> - High heating value (35.5 MJ kg⁻¹) - High surface area and pore volume 	Adsorbent or soil improvement additives	Mong et al. (2020)

N-doped biochar. Promotion of stable N-containing groups and a significant decrease of O-containing groups in the biochar could be obtained with this technique (Chen et al. 2020). The functional char with more active sites and surface functional groups obtained by NH₃ ambient pyrolysis could also significantly enhance its adsorption capacity (Shen et al. 2017; Mian et al. 2018). Table 3 summarized some recent studies on pyrolysis under non-inert ambiances and its effects on the biochar product.

3.4 Hydrothermal Carbonization

The advantage of hydrothermal carbonization lies in the capability of converting the “wet” raw material into carbonaceous solids at a relatively high yield in the absence of an energy-intensive drying process, hence lowering the requirement of excess auxiliary drying equipment (Zhou et al. 2019; Olszewski et al. 2020). This solution helps handle a wide range of unconventional sources of biomass, such as sewage sludge, municipal solid waste (bio-fraction), livestock and aquaculture residues, as well as the very new types of biomass of interest such as algae. To some extent, hydrothermal carbonization helps minimize the harmful effects to the environment from these types of biomass, because some of them are continuously generated in large quantities and require expensive management or treatment stages. Moreover, hydrothermal carbonization can be followed by an activation process to produce functional biochar. The latter is superior to ordinary biochar in some features such as adsorption abilities versus flue gases or heavy metals (Tu et al. 2021). Table 4 presents some examples of wet pyrolysis and the corresponding results on the char product.

3.5 Integrated Pyrolysis Techniques

The combination of several pyrolysis techniques in a correct way can optimize the strengths and limit the weaknesses of each method. Therefore, integrated pyrolysis techniques provide various advantages, such as high conversion efficiency, low processing temperatures, and the capability to process wet and aqueous raw materials (Liu et al. 2020; Luo et al. 2020; Zhang et al. 2021a). As an example, in our study, biochar prepared from cashew nut shell, bagasse, macadamia nut shell under conventional pyrolysis process at different temperatures and residence times in an N₂ environment were compared to microwave-hydrochar generated in a 1200-W microwave hydrothermal system (temperature: 200 °C in 15 min, with a biomass-to-water ratio of 1:10). Results highlighted a significant increase in the porosity, expressed by the higher position of the N₂ adsorption–desorption isotherms in Fig. 4.

Table 3 Recent studies on pyrolysis under non-inert ambiances

Raw material	Pyrolysis conditions	Effect on biochar properties	Application	References
Poplar, giant reed	<ul style="list-style-type: none"> - Temperature: 600° - Steam mass flow rate: 0.25 g s⁻¹ 	<ul style="list-style-type: none"> - High surface area (347.5 m² g⁻¹) - Lower metals mobility 	Immobilization of retained potentially toxic elements	Grottola et al. (2019)
Populus nigra	<ul style="list-style-type: none"> - Temperature: 480–700 °C; - Steam flow rate: 0.25 g s⁻¹ 	<ul style="list-style-type: none"> - Biochar with narrow microporosity and average pore sizes from 0.55 and 0.6 nm 	CO ₂ adsorption and separation	Gargiulo et al. (2018)
Bamboo	<ul style="list-style-type: none"> - Temperature: 500 °C in 45 min; - Steam flow rate: 5 ml.min⁻¹ 	<ul style="list-style-type: none"> - Low surface area (2.12 m² g⁻¹) with mesoporous structure - Enhancement of active sites 	Adsorption of Cu ²⁺ and tetracycline	Wang et al. (2020)
Red pepper stalk	<ul style="list-style-type: none"> - Temperature: 600 °C - CO₂ concentration: 50% (in N₂) and 100% - Total flow rate: 500 ml min⁻¹ 	<ul style="list-style-type: none"> - Increase in surface area compared to biochar produced with N₂ (109 m² g⁻¹ vs. 32.46 m² g⁻¹) - Lower pH and electrical conductivity - Higher ash content, degree of carbonization, hydrophobicity, and polarity 	Waste management, energy recovery, and biochar production	Lee et al. (2017b)
Bamboo waste	<ul style="list-style-type: none"> - Temperature: 600 °C - NH₃ concentration: 40% in Ar - Total flow rate: 200 ml min⁻¹ 	<ul style="list-style-type: none"> - Increase of N₂ content (4.85 wt%) and N-containing groups; - Increase of specific surface area (369.59 m² g⁻¹) - Excellent electrochemical property (120 F g⁻¹) 	Production of N-doped biochar	Chen et al. (2018)

(continued)

Table 3 (continued)

Raw material	Pyrolysis conditions	Effect on biochar properties	Application	References
FeCl ₃ -laden agar biomass	<ul style="list-style-type: none"> - Temperature: 600 and 800 °C in 1 h - NH₃ concentration: 28% in N₂ - Total flow rate: 500 ml min⁻¹ 	Effect on biochar properties <ul style="list-style-type: none"> - High Cr (VI) adsorption capacity (up to 142.86 mg g⁻¹) - Increase of magnetic properties and activated N-functional groups 	Removal of Cr (VI)	Mian et al. (2018)

Table 4 Recent studies on hydrothermal carbonization

Raw material	Pyrolysis conditions	Effect on biochar properties	Application	References
Bamboo	<ul style="list-style-type: none"> - Temperature: 220–260 °C in 1 h - Biomass: solution ratio: 1:10 (0.1 M HCl, H₂SO₄, and HNO₃) - Heating rate: 3 °C min⁻¹ 	<ul style="list-style-type: none"> - Low specific surface (3–16 m² g⁻¹), small pore size (1–10 nm) - HCl and HNO₃ can form a single micron carbon sphere - H₂SO₄ can form irregularly shaped hydrochar particles groups 	Production of functional hydrochar microspheres	Zhang et al. (2021b)
Olive pulp	<ul style="list-style-type: none"> - Temperature: 190–250 °C between 2 and 15 h - Biomass: deionized water ratio: 1:3.15 - Heating rate 5 °C min⁻¹ 	<ul style="list-style-type: none"> - Increase the residence time to 15 h increased the hydrochar yield by 8%; - Higher temperatures and longer residence times increased HHV from 24 to 30 MJkg⁻¹, and decreased electrical resistivity from 800 to 200 mΩm 	Production of pellets at low compression pressure without a binder for use as metallurgical reducing agents	Surup et al. (2020)
Defective coffee beans	<ul style="list-style-type: none"> - Temperature: 150–250 °C for 40 min - Biomass: deionized water ratio: 1:10 	<ul style="list-style-type: none"> - Decrease in ash content, N content, H/C, and O/C molar ratios with increasing temperature 	Production of functional and energy-dense solid biofuels	Santos Santana et al. (2020)
Tobacco stalk	<ul style="list-style-type: none"> - Temperature: 220 °C in 6 h - Biomass: catalyst: deionized water ratio: 2:1:30 	<ul style="list-style-type: none"> - High surface area (1498 m² g⁻¹) and volume (0.712 cm³ g⁻¹) - Nitrogen functions on the surface significantly increased 	Enhancement of CO ₂ capture performance	Huang et al. (2021)
Wood and parchment of <i>Coffea arabica</i> , <i>Eucalyptus</i> sp. wood, and giant bamboo	<ul style="list-style-type: none"> - Temperature: 180–240 °C in 3 h - Biomass: deionized water ratio: 1:8 	<ul style="list-style-type: none"> - HHV, fixed carbon content, and energy density increased with increasing reaction severity - High HHV (24.6–29.2 MJ kg⁻¹) at temperatures ≥ 220 °C 	Energy and value-added products generation	Mendoza Martinez et al. (2021)

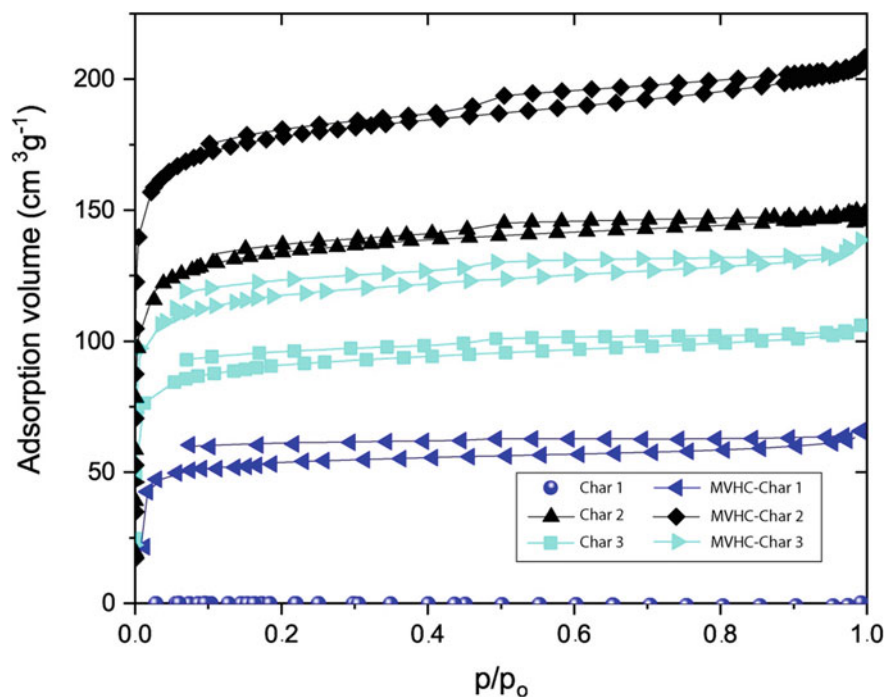


Fig. 4 The N_2 adsorption–desorption isotherms of different “normal” biochars (Char) and microwave-hydrothermal chars (MVHC-Char). Char 1: Cashew nut shell, Char 2: Macadamia nut shell, Char 3: Bagasse

4 Limitations of New Pyrolysis Methods

4.1 Co-pyrolysis

The primary disadvantage of co-pyrolysis lies in the instability of the product quality. Given that this technique deals with many types of biomass, the mechanism involved in co-pyrolysis generally goes through a series of complex reactions that is hard to control, especially on large scales (Abnisa and Wan Daud 2014). Moreover, it varies from one raw material to the other, and is highly dependent on the mixing ratio and the pyrolysis conditions. Knowledge of the synergistic effects during pyrolysis, therefore, remains poor.

4.2 *Microwave-Assisted Pyrolysis*

The energy conversion efficiency of microwave-assisted pyrolysis is relatively low. The energy conversion of the input electric energy to heat by microwave heating is typically in the range of 20–60% (Yao et al. 2014; Sun et al. 2016; Rosa et al. 2017). Moreover, the energy loss in the transformer and the magnetron during microwave-assisted pyrolysis of biomass is approximately 26%, and the power loss caused by the heat loss is approximately 29% (Xiqiang et al. 2014; Zhang et al. 2017a). The interference between electromagnetic irradiation and thermocouple sensors also limits the accurate measurement of the temperature in a microwave reactor (Yin 2012). In addition, the penetration depth in materials of microwave-assisted pyrolysis is quite limited, inhibiting the size of the reactor (Sajjadi et al. 2014; Sun et al. 2016).

4.3 *Pyrolysis Under Non-inert Ambiances*

The biggest limitation of substituting N_2 with other atmospheres for pyrolysis lies in the cost of the substitutes. The higher cost of pure NH_3 and compressed CO_2 may hinder the development of this technique. For the case of steam, it needs another piece of equipment to generate superheated steam, or supercritical steam before injecting it into the pyrolysis system. This requires cheaper gas/chemical sources with comparable efficiency. In addition, the optimum concentration of reactive agents in pyrolysis depends on many factors. The quality of the biochar produced is not necessarily proportional to the increase in the concentration of these gases or chemicals (Chen et al. 2018; Yang et al. 2018), asking for intensive research for each individual process and material.

4.4 *Hydrothermal Carbonization*

Hydrothermal carbonization (wet pyrolysis) is an energy-intensive process, from heating up the water in a high-moisture content raw material, powering the process to drying the char product after the process, so the energy balance should be well established if the generated char is used for energy purposes. A longer treatment time and further treatments that are often required for wet biomass waste also present disadvantages of this technique (Ischia and Fiori 2021). Moreover, some previous studies report lower stability of the biochar produced from wet pyrolysis, in comparison with the char produced from dry pyrolysis (Liu et al. 2021). Thus, possible toxic effects or risks of the biochar produced from wet pyrolysis in long-term applications, such as in soil amendment, have to be carefully evaluated. This knowledge is still

lacking in the literature. The high-pressure requirement for raw material decomposition is also a big limitation to the upscale of this technology. Further improvements in hydrothermal carbonization need to be explored to use simplified devices.

4.5 *Integrated Pyrolysis Techniques*

The complexity of integrated pyrolysis techniques represents the biggest limitation for their applications on larger scales. The characteristics of the product are influenced by a set of various factors with mutual effects, where the degradation mechanism of the raw material depends on the nature of each raw material in each specific condition. The sensibility of these methods versus pyrolysis factors is relatively high, causing high difficulty in process control. To date, no pilot studies have been published, and information on the economic efficiency and stability of these methods is very sketchy and requires more research in the future.

5 Conclusion and Future Prospects

Various pyrolysis strategies of biomass have emerged as new trends in biochar production to enhance sustainability. These trends include co-pyrolysis, microwave-assisted pyrolysis, pyrolysis under non-inert ambiances, hydrothermal carbonization (wet pyrolysis), and integrated pyrolysis techniques. Each process has its own advantages—such as treatment of a wide range of unconventional raw materials, synthesis of novel biochar composites, or enhanced biomass conversion efficiency, but also limitations—such as instability of the product quality, high investment costs, or difficulty in up-scaling. The effects of these novel methods on the biochar characteristics can be categorized into four groups: (a) improvement of specific surface and pore structure, (b) increase of certain functional groups, (c) promotion of the activated oxygen species, and (d) immobility of heavy metals in biochar. Opportunities for the application of these techniques are numerous, from adsorption of contaminants, soil amendment, reduction of risks of using contaminated biomass-derived biochar, and energy production.

Further investigations should be implemented to facilitate the application of engineered biochar. Information on the relationship between the raw materials, catalysts, and pyrolysis conditions on the characteristics of the resulted biochar is still very limited and incomplete, requiring more systematic studies. As new characteristics of biochar are developed, pilot tests or field experiments are also recommended to evaluate the real effects of biochar prior to large-scale applications or their long-term risk during use, especially for biochar derived from different types of sludge or other contaminated biomass. Novel statistical analysis methods such as modeling or machine learning would be a good idea to predict the properties of biochar and their impacts during environmental applications.

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