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Recent advances in bio-based carbon materials for anaerobic digestion: A review

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ABSTRACT

In recent years, the great interest in improving anaerobic digestion (AD) process resulted in the use of different types of additives or accelerants to exploit several profitable synergies, including improved CH_4 yield and digestate quality. However, the use of carbon-based materials also reduces operational instability and substrate-induced inhibition that hinder microbial breakdown activity of the organic matter. In this review, recent advances made by different research groups in order to enhance the performance of AD system are reviewed, emphasizing the utilization of low-cost carbon-based materials in the AD process, with a particular focus on the use of bio-based carbons such as biochar, activated carbon, and carbon cloth. The bio-based carbons in AD system support bacterial syntrophy and accelerate to direct interspecies electron transfer (DIET), and enhance methane yield. Moreover, this review gives emphasis on the fabrication of bio-based carbon materials and their applications as additives and their working mechanism in the AD system. Finally, this manuscript debates optimized AD technology to enhance biogas yield via bio-based carbons for energy production and sustainable environment for betterment of human health.

1. Introduction

The burning of fossil fuel and solid organic wastes cause the accumulation of greenhouse gasses (CO_2 and CH_4, etc.) in atmosphere, directly linked with environment and public' health [1]. Reduction in greenhouse gasses and production of clean energy recommends the clean and sustained environment for next generations. For this purpose, China has committed to reduce CO_2 emission of 40–50% by 2020 [2]. Due to current pandemic lockdown over the world, globally 5% reduction was occurred in CO_2 emissions in 2020 as compared to 2019, mainly caused by a 8%, 4.5% and 2.3% reduction in emissions from coal, oil and natural gas respectively [3]. In this regard, AD can give guarantee of clean environment and safe disposing of organic solid wastes along with production of bioenergy [4–6]. A household-based AD digester can produce as much biogas that can replace approximately 1.5 tons of fuelwood, alike yearly biomass collecting of 2333 m^2 of wood land, and can reduce about 2 tons of CO_2 emissions [7].

The AD is a complex biochemical process and consists of four stages (hydrolysis, acidogenesis, acetogenesis, and methanogenesis). These stages carried out by various type of microorganisms (*hydrolytic bacteria, acidogens, acetogens, and methanogens*), which are responsible for converting the complex organic matters into biogas mainly composed of methane CH_4 (50–70%), CO_2 (25–50%), and N_2 (0–10%) [8]. Acetogenesis stage results into the release of H_2 in large quantity, which imposes toxic effects on the anaerobic microbes which are necessary for this process. Moreover, the large H_2-partial pressure made the process thermodynamic infeasible. However, syntrophic metabolism and a good cooperation are required between *acetogens* and autotrophic *methanogens* to use this huge amount of produced H_2 [9,10]. Some efficient electron transmissions between *acetogens* and *methanogens* can utilize H_2 as electron carrier by improving organic biodegradation and CH_4 yield. During methanogenesis, direct interspecies electron transfer (DIET) has revealed a greater electron transfer efficiency than interspecies hydrogen transfer (IHT) and interspecies formate transfer (IFT) [10–12]. In addition, the substrate-induced inhibition is considered as one of most important technical challenges that occurs due to the metabolic intermediate products of substrates, and causes process instability, foaming and drastic drop in pH [13,14].

DIET can accelerate AD performance by improving its rate and efficiency. Naturally occurring DIET can only enhance the performance of that AD system, containing relatively diluted and simple organic wastes.

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Abbreviations

AD	Anaerobic digestion
AC	Activated carbon
GAC	Granular activated carbon
CNTs	Carbon nanotubes
CH_4	Methane
VFAs	Volatile Fatty Acids
COD	Chemical oxygen demand
OLR	Organic loading rate
DIET	Direct interspecies electron transfer
HTC	Hydrothermal carbonization
BET	Brunauer-Emmett-Teller
MP	Microwave pyrolysis
OmcZ	Outer-surface cytochrome

Moreover, DIET can be accelerated with simple and low cost modifications, to improve AD of more complex and concentrate solid organic wastes such as agricultural wastes and sludges. The conventional AD have several challenges like volatile fatty acids accumulation and H_2 that might be more toxic to methanogenic archaea and acetogenic bacteria, respectively, and slow electron transfer through H_2 or formate. These challenges limit the performance of the conventional AD [15,16]. AD integrated with DIET can increase CH_4 production and also controls the digester souring. Moreover, it can bypass the required shuttle system that microorganism use for electron transfer by cell-to-cell syntrophy. This syntropic association naturally present in methanogenic groups. Furthermore, the biogas yield can be enhanced by accelerating the DIET phenomenon, using conductive materials like carbon-based materials (graphene, carbon nanotube, activated carbon, and etc.) [17].

The addition of additives can easily resolve the problems related to AD process efficiency such as slow degradation of organic waste, poor stability, low biogas yield, weak syntrophic metabolism, and low concentrations of electron carriers (IHT/IFT) [15,18–22]. The bio-based carbons, can be prepared from biomass wastes at low cost through different fabrication methods including pyrolysis, microwave pyrolysis, hydrothermal carbonization and two-step activation, have great potential to enhance DIET mechanism. The bio-based carbons can also reduce many other serious problems related to AD process such as low CH_4 production rate, lengthy lag time, and problem to unsuitable conditions [8,11,23].

The principle motivation behind this manuscript is to give comprehensive information on the utilization of bio-based carbon materials as a mediator to accelerate the DIET for energy retrieval from organic waste. It also surveys the previous logical attempts to assess the performance of the AD process with various low-cost bio-based carbon materials and to identify their real impacts on biogas yield. Moreover, this review summaries the results of previous studies about bio-based carbons. Subsequently, these studies were deeply analyzed and research gaps were found in related problems that need to be solved, and also recommended guidelines for future direction.This work comprehensively compares and evaluates the effect of using bio-based carbons as additives on DIET for the first time, and provides a guide for enhancing AD performance.

1.1. Current status of anaerobic digestion

Globally, the bioenergy consumption was what could be compared to 460.1 million tons of oil (Mote) in 2017 with representing 16.5% of the expansion in 2018–2023 [24]. All over the world, about 50 million domestic AD plants (small-size), 132,000 commercial biogas plants (engineering biogas projects), and 700 upgrading AD plants are functioning, and about 15,000 commercial AD plants are working only in Europe [25]. In 2015, the biogas production was about 31.24 billion cubic meter (bcm) in European Union (EU), which will be increased upto 70 bcm in 2030 based on available resources [7]. In China, according to rural biogas development, total biogas production would be 20.7 bcm in 2020 including digester volume of 22.77 million m^3. Globally, the total production of energy from biogas was about 2.8×10^5 TJ in 2000, which reached 1.3×10^6 TJ in 2014, showing an annual increase of 13.2% [26]. Recently, EU have installed about 17,783 biogas plants with the electricity generation capacity of 10.5 GW. Germany is leading with 10, 971 biogas plants followed Italy, France, Switzerland and the UK with 1655, 742, 632 and 613 biogas plants, respectively [27].

Most of the studies on AD for improving biogas yield published in the two periods with time span from 2008 to 2012 and 2013–2020. During 2008–2012 about 700 papers were published, whereas a number of publications were increased about 2.5 times (1050 papers) during 2013–2020. Based on these publications, the proposed strategies for enhancing biogas yield can be categorized into six broader areas: parameters optimization (15–16%), pretreatments (20–22%), additives (4–5%), co-digestion (26–33%), bioreactor optimization (23-14%), and genetic technology (9–13%). These publications dedicated that the percentage of parametric and bioreactor optimization for AD during 2013–2020 years has reduced. Latest novel research emphases on changing genetic technology (single-stage to two-stage and multi-stage), magnetic field application in AD, co-digestion, and potential utilization of additives for optimizing AD system [21,28–30].

Although the great potential for AD to expressively contribute to renewable energy portfolio, the biogas still remains an under utilized renewable resource for energy production with only $4.7 \times 10^{11}-9.5 \times 10^{11}$ kWh of electrical energy produced by biogas in 2012, and contributing only 0.2–0.4% to worldwide production of electricity [31]. However, in recent years, it becomes a developing field of research for converting the organic waste including biomass produced from municipal waste, industrial waste; domestic waste, farm waste and agricultural waste, into beneficial products such as a renewable energy (CH_4) and soil fertility enhancer in the form of digestate to improve crops productivity. However, a wide variety of inhibitory substances and challenges are the primary causes of anaerobic digester upset or failure since they are present in substantial concentrations in the wastes. In the course of AD process, the substrate-induced inhibition stands a prominent anaerobic instability that occurs due to the metabolic intermediate products of substrates. These metabolic intermediate products pose a major hurdle, by disturbing the operational stability of the AD process because of acidification [32], which can retard the microbial activity (breakdown of the organic waste) and associated methane yield resulting in low energy output [33]. To improve the AD efficiency, several strategies such as substrate pretreatment, co-digestion, additive supplementation, parametric optimization (temperature, pH etc.) have shown excellent aptitude in improving the AD conversion process. However, the additive supplementation offers numerous advantages over the rest of the aforementioned, such as ease of application, no requirement of infrastructure modification and competitive operation cost [34].

To improve the AD process, the current technical challenges that are a great hindrance in its extensive adoption (i.e., slow startup, substrate-induced inhibition, and low energy output), can be handled by introducing various additives in AD systems. The detail discussion on additives in the AD system has been comprehensively carried out in the next section.

1.2. Additives in anaerobic digestion

Considering great practical applications, the AD of organic waste has been enhanced its performance by using various techniques including co-digestion, substrate pretreatment, additive supplementation, parametric optimization (temperature, pH, etc.). Among of these strategies, the introduction of accessible and cost-effective additives into AD systems has been proven a successful way to improve the AD performance, which could lead to enhance the gas production, favorable digestion conditions, and superior organic degradation [31,35,36]. There are many types of additives, such as microbes, enzymes, microelements, biological metabolites, adsorbents, a defoaming agent, chelating agents and so on for AD systems [37], and these additives can accelerate the decomposition of solids and volatile solids in AD reactor.

Of them, the enzymes and the microorganisms have been widely investigated as an alternative to physicochemical pretreatments of wastes before AD. However, their direct introduction into the digester has received less attention. Table 1 summarizes the effects of various types of additives, such as milk gel, polyvinyl alcohol, activated carbon (AC) [38], pectin, kaolin and silica gel [39], aluminum powder, talc, zeolite powder [40], bentonite [41], fly ash [42], plant ash [43] and so on, on AD systems have been tested, and their effects on the AD performance were summarized in Table 1. It should be noted that the carbon-based materials are also extensively being used in other fields of energy, like water purification, carbon fixation, environment, energy storage, fuel cell, gas sensors and CO_2 sequestration [44–46]. However, its use in AD system gets very fame now [47].

1.3. Latest development in anaerobic digestion with bio-based carbon materials

To enhance bioenergy from different biomass wastes, the carbon-based materials are widely used in AD systems due to their eco-friendly nature and low-cost fabrication from biomass wastes. Several research groups enhanced the bio-methanization with addition of bio-based carbons. For example, research group of Shen enhanced the cumulative CH_4 yield (46.9%) and maximize its production rate (181.6%) from AD digester of waste activated sludge amended with biochar [48]. Lü' research group enhanced the stimulatory mechanism with addition of biochar prepared by corn stover and conductive graphite. They found that biochar prepared from corn stover enhanced the methane production (46.9%) that was higher than the methane produced by graphite (38.3%), during the AD of waste activated sludge. Lü' research group also recommended that graphite and biochar increased methane via acetoclastic pathway. In CO_2 reducing pathway, biochar was superior to graphite due to the functional groups to deliver electron for methanosaeta. However, under higher H_2 partial pressure the graphite surpasses biochar by 42.4% in terms of methane production. Moreover, they demonstrated that when DIET mechanism become infeasible during methanogenesis stage, both biochar and graphite can enhance the methane production by acetoclastic pathway [49].

Yun' research group also enhanced the methanization with the addition of bio-based carbons in AD system. They developed a good synergistic effect between bio-based carbons and mixed substrate in AD system, enhanced the cumulative biogas production (580.9 mL/g VS), chemical oxygen demand (COD) reduction (79.37%) with adding 2.16 g/L aloe peel-derived bio-based carbons. They prepared the bio-carbon by using three methods including two-step activation, microwave pyrolysis, and hydrothermal carbonization [50]. Yun' research group also enhanced the biogas production (576–585 mL/g VS), and proposed the enhanced DIET on the bases of first-principle density functional theory with addition of biomass-derived carbon-based composites (Co/C, CoO/C, and Co_3O_4/C) [11]. Lü et al. applied different particles size of biochar for enhancing the biomethane. They found that powdered biochar (<5 μm) strengthened and aggregated more microorganism than granular biochar (0.5–1 mm), and also observed an increase CH_4 enhancement by 13.3% under thermophilic conditions. However, the granular biochar increased 32.5% of CH_4 yield in mesophilic digester, indicating most favorable perspective for energy recovery [51].

2. Carbon-based additives in anaerobic digestion

As discussed in section 1.3 that the strategy on the use of cost-effective additives in AD system has gained significance because of

Table 1
Additives in anaerobic digestion.

Additives	Substance	Effect	Ref.
Microbes	Hydrolytic bacteria	Increase the microbial diversity towards a better hydrolytic or methanogenic activity (bioaugmentation).	[154]
	Acid-producing bacteria	Accelerate the degradation rate of cellulose in the AD raw materials, shorten the fermentation cycle.	
	Methanogens	Increase the number of microorganisms in the methanogenic stage.	[118]
Enzymes	Cellulose Hemicellulose Protease Lipase Amylase Saccharification enzyme	Appear hydrolase activity at the peak of gas production, and the hydrolase activity was similar to that of biogas production. However, the amount of protease added should not be too high. Adding enzymes can facilitate hydrolysis of the particulate compounds.	[155]
Nutrients	Nitrogen Phosphorus Potassium Calcium	Require three essential nutrients such as C, N, and P for the growth of biogas fermentation microorganisms. Organic nitrogen in organics provides N-nutrients from bacteria, and P-based nutrients need to be added exogenously.	[43]
Microelements	Iron Cobalt Nickel Selenium	Require a sequence of nutrients for *methanogens*, such as N, S, P, Fe, Co, Ni, Mo, Se, vitamin B_2, vitamin B_{12}, etc..	[5, 156, 157]
Adsorbents	AC Bentonite Pectin Aluminum powder	Increase the biogas content and CH_4 content in different degrees, and decrease the BOD and COD content with the increase of the adsorption dose.	[158]
Foaming agents	Polyether defoamer Organosilicon defoamer	Eliminate foam during anaerobic digestion.	[159]
Chelating agents	Ehime blue Serine-cobalt complex	Not only provide a carbon source for anaerobic digestion microorganisms, but also improve the availability of other inorganic nutrients, thereby increasing the growth rate of the *methanogens* and the stability of the population, and ultimately increasing the gas production of anaerobic digestion.	[160]
Inhibitor	Heavy metal ions Acids Cyanide	Promote the biological activity of the system at very low concentration; when its concentration is high, it begins to produce inhibition; and the higher the concentration, the stronger the inhibition.	[161]
Accelerants	HfC, SiC, TiC, WC	Increase the biogas production (463–499 mL/g TS) and chemical oxygen demand (COD) and the buffer capacity, improve the degradation rate (58.62–78.90%) and total nitrogen concentrations (905.0–1077.0 mg/L) as compared with control check. Also, shorten the duration of	[162]

(continued on next page)

Table 1 (*continued*)

Additives	Substance	Effect	Ref.
		acidogenesis stage, the high-solids hydrolysis stage, and wet methanogenesis stage.	
	$Nb_{3.49}N_{4.56}O_{0.44}$, NbN, NbO_2	Improve the cumulative biogas yield (437.1–522.7 mL/g VS) and chemical oxygen demand removal rates (56.08%–65.19%) as compared with the control sample (409.2 mL/g VS and 29.55%).	[163]
	TaO, WO_3, $WO_{2.72}$, W_2N, NbO_2	Improve the COD degradation rate and cumulative biogas yield, WO_3 and $WO_{2.72}$ increase the cumulative biogas production by 38.8% and 92.8%, respectively, as compared with the control group.	[164]
	Steel slag	Increase the cumulative biogas yield, methane yield, and COD degradation rate by 507.29 mL/g VS%, 274.70 mL/g VS%, and 58.62%, respectively, as compared with the control group.	[165]
	Vermiculite	Increase biogas yield (295.14–353.96 mL/g VS), improve COD rates (45.53%–71.03%), VS and. TS reduction rate (50.70%–52.76%) than reference group (234.08 mL/g VS, 39.38%, 45.10%).	[166]
	Urea, bentonite, active carbon, plant ash	Increase biogas yield (485.7–681.9 mL/g VS), methane content (63.0–66.6%), TS,VS and COD (29.7–55.3%, 50.9–63.0%, and 46.8–69.1%). Also increase the fertilizer nutrient content (4.95–5.66%).	[43]
	$(Fe_2(SO_4)_3$, $Fe(NO_3)_3$, $FeCl_3$, $FeCl_2$	Increase the biogas yield (505–568 mL/g VS), reduce the digestion time (15–21 days), improve COD reductions (41.4–69.3%)	[36]

advancements in scientific knowledge and applications of new materials. Carbon-based materials (AC, graphene, carbon cloth, carbon nanotubes (CNTs), biochar, maghemite, and magnetite carbons) as an additive can improve the AD process due to their good properties, such as superior chemical stability, good electrical conductivity, and porous structure. The carbon-based materials can play key roles in enhancing AD process by promoting microbial growth and its aggregate, providing required nutrients to microbes, expediting interspecies electron transport, improving enzyme activity and buffering capacity, thus resulting in the faster VFAs formation and utilization, faster H_2 and CH_4 production, shorter lag phase, higher CH_4 contents, and better digestate quality. The carbon-based materials can also help to microbial immobilization by providing better habitation, a facility for bioelectrical influences among the cells, and some important elements for anaerobic microbes [52]. The several studies have been revealed that these carbon materials can improve the efficiency and stability of the AD process. It also increases CH_4 yield and digestate utilization [53–58].

In addition, these findings demonstrated that carbon-based materials in the AD systems could stimulate the methanogenesis by improving the DIET mechanism between *methanogens* and syntrophic bacteria. However, most of these studies were carried out with well-defined co-cultures or inside such anaerobic bioreactors served semi-solid slurry, or wastewater with pretreatment and functioned in both modes like semi-continuous and continuous [23,59,60].

2.1. Use of graphene in anaerobic digestion

The graphene has great impacts on anaerobic microbial communities during AD process. Due to great electric conductivity, high mechanical strength, and large surface area, graphene can improve the stability and efficiency of AD [55,61]. Tian et al. demonstrated that methanogenesis of glucose was improved by adding graphene during long-term AD under low-temperature range such as 10–20 °C [62,63]. The potential microbial network of the major bacteria and archaea involved in the AD of glycine with and without graphene was evaluated. In the AD of glycine without graphene, glycine is degraded via acidogenic microbes at first, which consist of *Aminobacterium* (17.6%) and *Levilinea* (18.0%), then glycine acidification produced intermediates products such as hydrogen and acetic acid. In the end, acetic acid is consumed through *methanosaeta species* (31.2%) to produce CH_4 and CO_2. The CO_2 may then be reduced to CH_4 via *Methanobacterium* (61.7%) and *Methanosarcina* (6.6%) through the hydrogenotrophic pathway. However, AD process with graphene gives significantly change the syntrophic associations between methanogenic archaea and acidogenic microbes, and increase the CH_4 production rate of 28% via *Methanobacterium* (71.1%) and *Methanosarcina* (11.3%) through DIET [64]. Moreover, some studies described that the CNTs effectively stimulated the methanogenesis phase of the AD process in syntrophic co-cultures as well as in complex environmental conditions [65].

Regardless of the great potential of CNTs and graphene as good promotor of DIET mechanism, their utilization in field-scale developments might be very difficult because of their high cost. The cost for adding 1 g of CNT/L was approximately $100,000/m^3 of the anaerobic digester, which is much higher than the cost for adding 50 g of AC/L ($15–150/m^3) [20]. Indeed, carbon-based materials take a significant part in adsorbing colloids and larger organics molecules, which can reduce the resistance of biomass cake, consequently enhancing the COD degradation [66,67].

2.2. Use of bio-based carbons in anaerobic digestion

The bio-based carbons such as AC, biochar, and carbon cloth have a great advantage over the other known additives in AD process because these can be fabricated from biomass waste at very low cost. Several studies demonstrated the positive effect of bio-based carbon materials on the biogas yield, COD degradation, the stability and the fertilizer utilization of the digestate [57,68–71]. In this way, these substances have the potential to be used in two ways (i) by reducing organic wastes, the potential source of indirect environmental pollution and public health risk. (ii) act as accelerants for AD optimization to enhance the production of bioenergy energy, as shown in Fig. 1. Moreover, the various types of AD reactors have been working for wastewater treatment and also for producing bioenergy such as UASB and VTBR. These reactors can be optimized for biogas generation and wastewater treatment via bio-based carbons and being recommended for future studies.

The bio-based carbons such as biochar in digestate can act as catalyst and nutrients retention enhancer. Additionally, biochar reduce leaching of pollutants and heavy metals via organics absorption (physically and chemically) such as ammonium, phosphate, nitrate, metals, nitrite and CO_2 [33,72]. The enhanced quality of digestate with biochar directly related to its properties including surface functional groups, existence of metals, large surface area and high porosity and ash content [50,73]. Hence, the bio-based carbons not only improve the efficiency of AD process but also enhance the quality of digestates as fertilizer utilization and protect the environment from contaminants spreading.

3. Fabrication of bio-based carbons

The microwave pyrolysis (MP) and hydrothermal carbonization

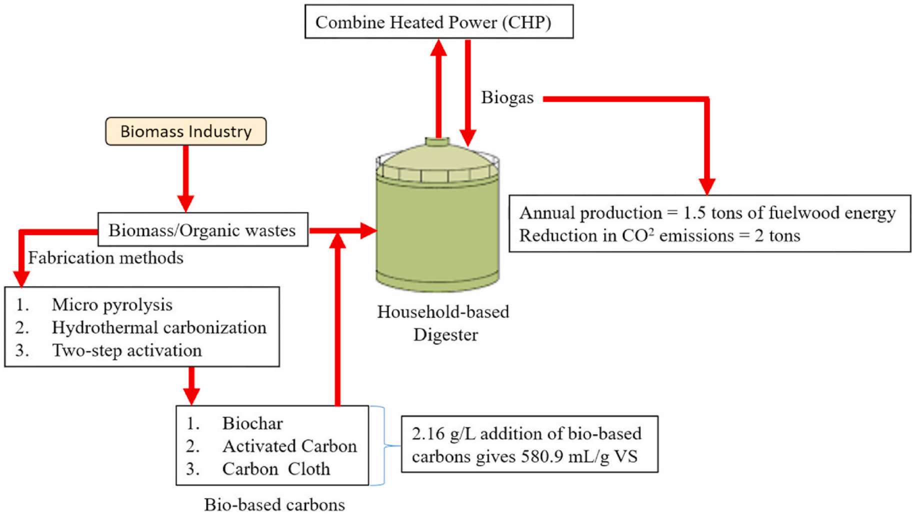

Fig. 1. The production biogas from household digester with additiob of bio-based carbons.

(HTC) methods are commonly used to prepare biochar and hydrochar from biomass, respectively, at lab scale. More favorable properties such as thermal stability, Brunauer-Emmett-Teller (BET) surface area, ash and oxygen contents, etc., of biochar, are higher than hydrochar due to a higher temperature in the MP than in the HTC [74]. Furthermore, the two-step activation method has been widely used for the preparation of AC from various materials like coconut shells, lignite, wood, and peat [75]. This method is a novel technique to prepare porous activated carbon with higher surface areas. The detail discussion on the following three fabrication methods is given below.

3.1. Microwave pyrolysis

The MP mainly consists of primary and secondary. Primary MP covers de-volatilization (dehydration, dehydrogenation, and decarboxylation) of the main constitutes, while secondary MP involves the thermal or catalytic cracking of heavy compounds or char into gases such as CO, CO_2, CH_4, and H_2 [76].

The MP method has been applied to pyrolyzed various lignocellulose feedstock, such as pinewood sawdust [77], peanut shell, maize stalk [39] and rice husk [78]. In this regard, Yun et al. used sawdust, walnut shell, waste carton, corn cob as a precursor, and H_3PO_4 as activating agent to prepare the AC materials via one-step MP with activation process, and products get larger BET specific surface area of 580–824 m^2/g and uniform pore volume of 0.54–0.64 m^3/g [79]. Schematic illustration of the developed one-step MP with activation process is shown in Fig. 2a. The corn grown on-farm was threshed, and then the corn cobs were obtained. The obtained corn cobs were cut into very small pieces and further converted into cob powder after grinding. The corn cob powder was mixed with concentrated H_3PO_4 (85 wt%) by mass 1:3, and then this mixture was put in vacuum drying and resulted in black viscous charcoal. After washing with HCl and drying with a microwave oven at 500 W for 6 min, finally, the black viscous porous charcoal was transformed into required porous AC (C_c).

Advantages: The MP has many advantages as compared to conventional pyrolysis (CP) method, (i) accelerating heating rate, and selective heating, (ii) improving reaction rate, (iii) increasing energy efficiency, (iv) quick start-up and stopping, (v) uniform internal heating of large biomass and easy control, (vi) better accuracy and efficiency due to volumetric and uniform heating [80–82].

Disadvantages: The MP has some disadvantages including (i) Large scale reproducibility/inhomogeneneities problems in yields, (ii) Finances of the process scaling-up, (iii) Temperature measurement [82].

3.2. Hydrothermal carbonization

HTC is a well-known thermal conversion method that has been displayed to be a lot of energetically and environmentally beneficial for the wet biomass conversion into hydrochar, bio-oil, and biogas. The HTC conversion process of the biomass is achieved with water by applying high temperatures (180–250 °C) at elevated pressure (2–10 MPa) for several hours [41,50,83].

The HTC process usually undergoes decarboxylation, hydrolysis, dehydration, aromatization, and re-condensation simultaneously. The hydrolysis process breaks down the chemical structure of biomass by the help of ester cleavage and either the bonds of bio-macromolecules through water molecules. The hydrolysis process also creates saccharides (oligo-), and lignin trashes that go into the liquid phase [84]. Then the fragments of lignin are rapidly hydrolyzed into phenols compounds, but on the other hand, the saccharides can choose some other chemical ways to initiate and products during the process of the HTC. These products produced through the other mechanisms could undergo hydrolysis process [85].

During the dehydration process, water is removed from the biomass matrix, eliminating the hydroxyl groups. Decarboxylation is responsible for the CO_2 removal from the biomass, which causing the carboxyl groups to be eliminated from the process [84]. The aromatization takes place due to the decarboxylation and dehydration process [86]. The furfural compounds produced via these two processes (mechanisms) then undergo hydrolysis process, which further breaks down into phenols, aldehydes, and acids. The produced acids catalyze inorganic elements that are released from the biomass [87]. That compounds produced during the earlier mentioned mechanisms undergo re-condensation due to its high reactivity. Highly reactive fragments in lignin condense rapidly and easily, along with aromatized polymers after the degradation of cellulose [88]. The hydrochar is formed after the re-condensation of HTC degradation products [84]. Wang et al. used sustainable sunflower stalk as raw material via the hydrothermal carbonization process successfully to prepare hydrochar, followed by the pyrolysis and KOH activation, three kinds of bio-based carbon materials (such as hydrochar (HC), pyrolytic carbon (PC), and activated carbon (AC)) were obtained, as illustrated in Fig. 2b [83].

Advantages: The advantages of HTC over other thermochemical processes (i) cut down the cost of prior drying processes, (ii) high solid yields, (iii) excellent final product quality including conductive behavior, high calorific value, adjustable surface functionalities, existence of natural binders, and ash composition, (iv) ease of operation, (v) low cost and energy efficient [89,90].

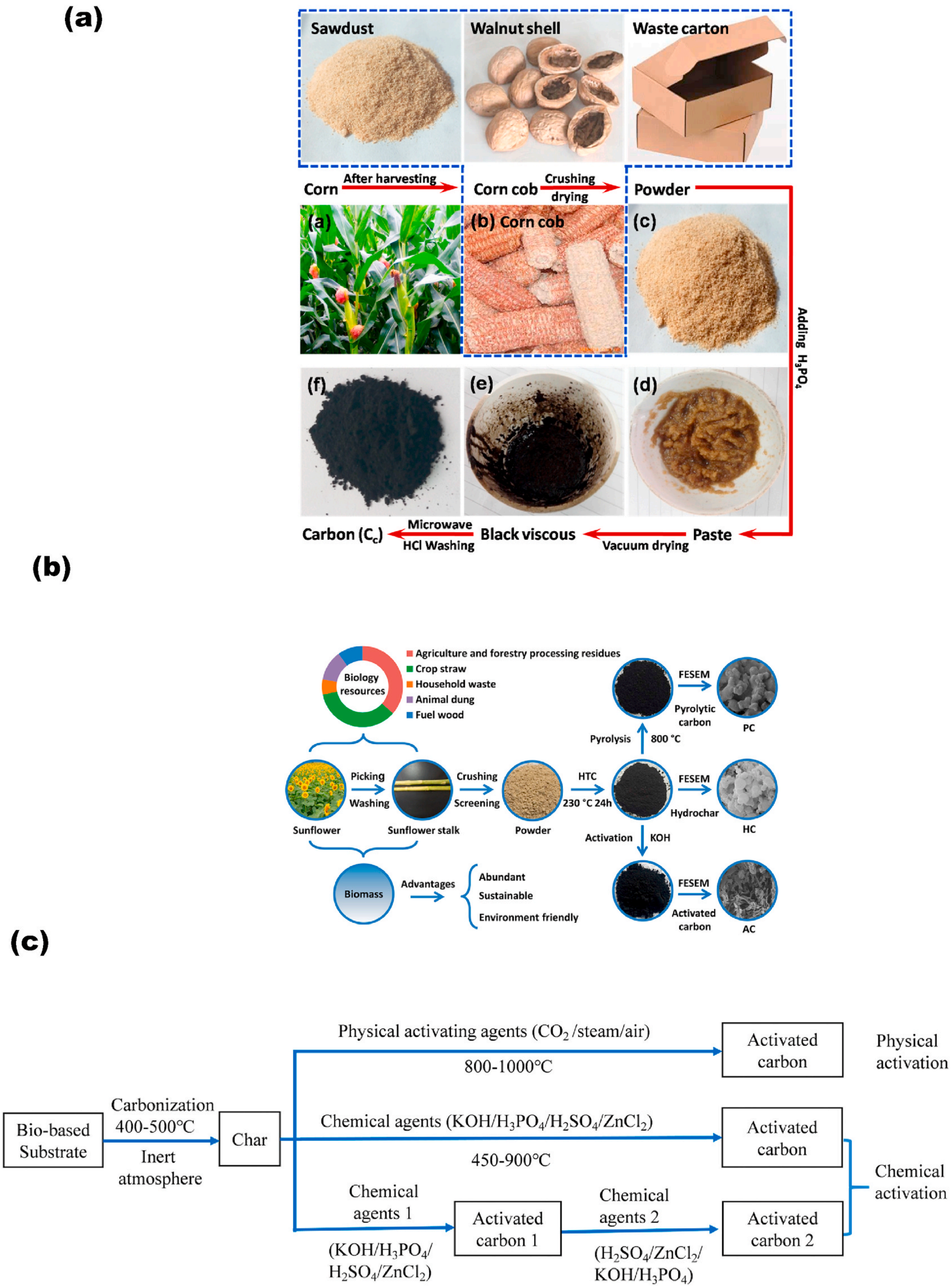

Fig. 2. (a) Schematic illustration of the preparation process of microwave pyrolytic activated carbon (Cc) derived from waste biomass resources sawdust, waste carton, walnut shell and corn cob [68], (b) Schematic illustration of preparing bio-based carbon materials derived from sunflower stalk [83], (c) Schematic diagram of physical and chemical activation.

Disadvantages: (i) Generalize the dynamics of rate limiting steps, (ii) HTC cannot offer adequate informations for full-scale facilities, there application limited to similar feedstock and experimental conditions.

3.3. Two-step activation

Generally, two-step activation can be divided into physical two-step activation and chemical two-step activation, as shown in Fig. 2c. Usually, some activation agents, such as alkali (KOH, K_2CO_3, NaOH, and Na_2CO_3), alkaline earth metal salts ($AlCl_3$ and $ZnCl_2$) and some acids (H_3PO_4 and H_2SO_4), are used in chemical activation method [91].

For physical two-step activation, in the first step activation, carbonization is carried out by pyrolysis of the feedstock in an inert environment at 400–500 °C, resulting in the formation of a non-porous carbon material or char. In the second step activation, by introducing an oxidizing gas, such as CO_2 or steam or air at the 800–1000 °C, the char transfers into the AC with microporous structure [92].

By contrast, chemical two-step activation is carried out by two different methods. Firstly, like physical two-step activation, the raw material is heated at 450–500 °C in an inert environment for carbonization to get non-porous biochar in the first step, and then the char is chemically activated by mixing activation agents (H_3PO_4/H_2SO_4/KOH/$ZnCl_2$) [93] in the second step and finally transferred into the activated carbon [94].

In a second way for the chemical two-step activation, the carbonization and activation steps are carried out simultaneously with the addition of dehydrated chemical agents, such as H_3PO_4, H_2SO_4, KOH, and $ZnCl_2$, and the mixtures are heated at 450–900 °C in an inert atmosphere and obtained the activated carbon. This method is a novel technique to prepare porous activated carbon with higher surface areas.

Advantages: (i) Two-step process includes carbonization and activation, (ii) it can enhance carbon content and porosity, (iii) it can be used for non-uniform sized particles so that save prior milling stage.

Disadvantages: (i) It needs long time and consumes more energy than other two process, (ii) it increases production cost due loss materials of during transferring and reloading, (iii) the produced AC by this process is not more effective AC [95].

Moreover, Bio-based carbons production cost depends on biomass waste type and pyrolysis process. Chiappero et al. reviewed that the production cost of activated carbon through pyrolysis (0.6 and 20 USD/kg) is quite higher than biochar (0.2–0.5 USD/kg), clearly indicating that biochar more economical for AD process [73].

4. Use of bio-based carbon materials in anaerobic digestion

4.1. Biochar in the anaerobic digestion system

Generally, the biochar has been potentially used in greenhouse gas reduction, for carbon sequestration, and as a soil enhancer in several agricultural and environmental applications [96]. It is well known that biochar possesses unique properties, such as adsorption, alkalinity, porous structure, large surface area and great ion-exchange ability [97], which can effectively prevent various type of environmental pollutants, such as organic and metal pollutants, and also reduce their ecotoxicity and bioavailability [98,99].

Recently, it was also reported that the biochar as an additive could improve the efficiency of the anaerobic process through mitigating ammonium (NH_4^+) inhibition, accompanied by encouraging the development of *methanogenic archaea* [100], consequently the lag spell was reduced in 21.4–35.7%, and thus H_2 production potential was improved from 14.2% to 31.0% [66]. Several other studies have also indicated that the biochar in AD system as an additive can reduce lag phase length by improving the CH_4 yield [101,102]. It can also enhance the methanogenic conversion of organic waste materials by stimulating the syntrophic relation between methanogenic and acetogenic microbes [9]. Biochar also supports the DIET mechanism, which is responsible for enhancing the CH_4 production in the AD of wastewater [103].

The practices concerning the effect of biochar on the AD system as an additive in recent years are given in Table 2. Enhancements in the degradation rate and increase in the methane yield have been described by many research groups [104,105]. The addition of the biochar in AD system as an additive is proposed to decrease different inhibitory phases and help the AD process stability, and also favorable to adsorb toxic compounds, enhance buffering capacity and support the immobilization of anaerobic microflora [33].

4.2. Bio-based activated carbon in the anaerobic digestion system

AC is widely used as a supporting media for biomass retention in several bioreactors [106], a redox mediator in several environmental applications, an adsorbent in wastewater treatment, and an electrode in electrochemical system due to its remarkable properties such as large specific surface area, rich porosity, and high electrical conductivity [107]. In recent years, the use of AC in the AD process is getting popularity due to its interesting properties, availability, and economical formation.

Table 2
Effects of bio-based carbons on AD process.

Additives	Substrate	Major results	Ref.
GAC	Synthetic wastewater	Improve DIET mechanism; Enhance methane production rate; Increase the methanogens.	[134]
GAC	Sewage Sludge	Improve syntrophy; Enhance the DIET; Enhance rate of sludge reduction.	[70]
GAC	Wastewater	Develop the syntrophy and improve DIET; Increase methanogens.	[114]
GAC	Wastewater	Increase the methane yield; Reduce the redox potential; Promote the DIET.	[167]
GAC	Sewage Sludge	Enhance syntrophic metabolism; Enhance DIET and methane yield.	[113]
AC	Poultry blood	Improve the methane production; Adsorb the ammonium.	[112]
AC	Food Waste	Improve the methane production. Enhance the process stability.	[111]
AC	Food Waste	Improve cumulative and specific methane yield; Tolerate high OLR.	[109]
AC	Dairy manure	Improve the cumulative biogas yield; Enhance the digestate stability.	[79]
AC	Dairy manure	Improve the cumulative biogas yield and COD removal rate	[22]
AC	Food Waste Poultry shits	Improve the biogas yield; Increase the OLR.	[108]
Biochar	Glucose	Shorten the methanogenic lag phase, improve the methane production rate (38–86.6%) of glucose.	[100, 168]
Biochar	Food wastes	Shorten the methanogenic lag phase and enhance the methane production rate by 123%. Proceed at a rate up to 5 times by conversion of VFAs higher.	[104, 169]
Biochar	Swine manure	Improve the methane yield (39%) and enhance the proteins degradation.	[105]
Biochar	Sewage sludge	Produce near pipeline-quality biomethane (>90% CH_4 and <5 ppb H_2S), and increase alkalinity and mitigated NH_3 inhibition, as well as increase maximum methane production rate by 27.6%.	[170]
Biochar	Municipal Solid Waste	Increase about 5% methane yield.	[171]
Biochar	Citrus peels (Orange)	Enhance the methane production about 56%, sport the electro-active microbes consortia developing a better syntrophic metabolic rate between archaeal and eubacterial populations.	[172]

Several studies reported that the AC-amended AD system exhibited great possibility to increase the microbial resistance against the high organic load and facilitate the microbes to settle down in colonies, which were beneficial to enhance the syntrophic links between *acetogens* and *methanogens*, thus resulting in more CH_4 yield [108,109]. Additionally, AC can adsorb many pollutants (antibiotics and ammonium) and reduce the VFAs concentration that greatly inhibits the microbial metabolism in AD systems [68,110,111]. The use of AC (powder and granular) as additives in the AD of poultry blood residue can reduce the ammonium and VFAs inhibition by enhancing the specific CH_4 production (216 ± 12 mL/g VS) [112].

Recently, Yun et al. used four different types of bio-based activated carbons in AD systems. It was noted that bio-based carbon-amended AD systems exhibited two characteristic peaks, and enhanced cumulative biogas yield (380–502 mL/g TS), indicating the process stability and superior digestion environment for biogas production in the AD system. Moreover, these bio-based carbons enhanced the digestate stability, and total weight loss of digestates range from 55.28% to 65.38%, lower than that of the control group (70.54%) [79]. In another work, Yun et al. has investigated the role of different dosages of granular activated carbon (GAC) in the AD of sludge, and their effect on cumulative CH_4 and sludge reduction. Notably, the addition of GAC can enhance the CH_4 production by 17.4%, and the sludge reduction (total suspended solids (TSS) removal) rate increased about 6.1% from 39.1% to 45.2% [57]. In addition, some researchers used GAC along with magnetite or trace element to enhance the CH_4 yield. Peng et al. observed that CH_4 yield can be improved by adding GAC, magnetite, and magnetite-GAC by 20%, 13.1%, and 20%, respectively, during the AD of sludge [113].

To enhance the CH_4 production rate by AC and trace elements simultaneously in AD systems as additives, is an encouraging technique. Recently, Capson-Tojo et al. reported that the AD systems amended with GAC and trace elements simultaneously showed higher CH_4 production rates approximately 545–719 mL d^{-1} than the AD systems amended with GAC and trace element separately (397–419 mL d^{-1}). Moreover, the addition of GAC favored food waste acclimation by improving acetic acid consumption, and trace elements speed up the propionic acid consumption. It was very clearly observed that a strong synergy was developed when trace elements and GAC were added simultaneously [53]. The most recent information on the use of AC in AD systems in order to improve the performance of the AD process is summarized in Table 2. AC can also facilitate the syntrophic metabolism of microorganism and DIET mechanism [108,109,112]. The DIET mechanism will be discussed in Section 5.

4.3. Bio-based carbon cloth in the anaerobic digestion system

Carbon cloth is another bio-based carbon material which has been generally used to enhance the AD performance by stimulating the DIET mechanism [54]. In addition, the carbon cloth has a great ability to enhance COD removal during the syntrophic conversion of ethanol to methane. However, the CH_4 production rate of the AD systems with bio-based carbon cloth is lower than that of the AD systems with biochar and GAC additives, which is the major reason for limiting its use in AD system [20].

Previous works illustrated that the carbon cloth could greatly improve the organic loading rate (OLR) and biogas yield. Dang et al. reported that the carbon cloth and GAC can improve the CH_4 yield rate of approximately 109.6 ± 3.0 and 106.6 ± 9.4 mmol/d in AD systems, respectively, allowing high OLR (6.7 kg COD/(m^3 d)), and stimulating fast retrieval of soured reactors [59]. Zhao et al. improved the CH_4 production about 59%, higher than the control group at 5 pH level with 750 mL d^{-1} production rate by adding the bio-based carbon cloth in the anaerobic digester [114]. Lovely et al. reported that the carbon cloth could also enhance the conversion of solid wastes to CH_4 and allow higher OLR [115]. Furthermore, Lei et al. increased the production rate about 11.5 L d^{-1} than reference 8.9 L d^{-1} by adding bio-based carbon cloth in the anaerobic reactor. They also found that carbon cloth-amended reactor can be stably operated with high OLR 49.4 kg

Table 3
Summary of different bio-based carbons used as additives for promoting methane production in AD systems.

Additives[a]	Substrate[b] (Digestion time/day)	Digestion mode[c]	Optimal dosage	Synthesis method for bio-based carbon	CODt removal/%	Biogas yield	Ref.
Biochar	APL (49)	Batch	0.8 g/mL	Pyrolysis at 400 °C	–	5 g/L	[173]
Biochar	Sludge (45)	Batch	4 g/L	Pyrolysis at 800 °C	–	16.6 6 mmol/g	[168]
Biochar	Organic wastes (20)	SC	5.0 g/L	Gasification	98	127.7 ± 2.7 mL/h	[174]
Biochar	Sludge (30)	Batch	4.97 g/g TS	Purchased	31.8	107.08 ± 3.35 mL/day	[23]
Biochar	Sludge (80)	Batch	5 g/L	Pyrolysis	–	150 mL/h	
Biochar	Sludge (319)	SC	0.5 g/d	Purchased	32.5	0.342 L/g	[175]
Biochar	Sludge (25)	Batch	10% w/w	Purchased	–	200 mL/g	[176]
Biochar	Sludge (32)	Batch	10 g/L	Pyrolysis	–	332 ± 16 mL/g	[116]
Biochar	Sludge (–)	Sequence	2.49 g/g TS	Pyrolysis at 450 °C	28.6 ± 5.5	0.329 ± 0.001 (mL CH_4/g COD degraded)	[101]
AC	Poultry blood(20)	Batch	3.3 g/L	Purchased	–	1052.7 mL	[112]
AC	Seed sludge(52)	Batch	14.5 g/L	Purchased	–	4.38 g VSFW/L^{-1} d^{-1}	[57]
AC	Sludge(20)	Batch	10.27 ± 0.18 g/L	Purchased	–	104 ± 3 μmol/d	[133]
AC	Dairy Manure	Batch	0.27 wt%	Two-step activation	70.95	525 mL/g VS	[22]
GAC	–	Batch	25 g/L	Purchased	–	Decreased lag phase. Increase methane production rate (2.5 times)	[177]
GAC	–	Continuous	1 g/L	Purchased	–	Increase methane production rate (1.8 times)	[178]
GAC	–	Batch	–	Purchased	–	Improve methane production rates over control	[144]
GAC	–	Batch	50 g/L	Purchased	–	Improve production rate (18 times)	[110]
GAC	–	SC	50 g/L	Purchased	–	Improve methane production rate	[179]
GAC	–	Batch	10 g/L	Purchased	–	Improve production rate (2.68 times)	[153]
GAC	–	Batch	0.5–5 g/L	Purchased	–	Improve production rate (17.4% 5 g/L times)	[57]
Carbon cloth	Sludge	Batch	5.2 g/L	Purchased	–	800 mL d^{-1}	[114]
Carbon cloth	–	SC	500 cm^2/L	Purchased	–	Increased methane production by 59%	[114]
Carbon cloth	–	SC	1465 cm^2/L	Purchased	8.5	Increased methane production 13 at high OLR 8.5 kg	[179]
Carbon cloth	–	Continuous	833 cm^2/L	Purchased	–	Increased methane production (1.3 times)	[60]
Carbon cloth	–	Batch	300−600 cm^2/L	Purchased	–	Increased methane production rate &improve ethanol degradation	[180]
Bio-based carbon	DM (35)	Batch	1.8 g/L	Microwave pyrolysis	51.39–67.81	Improve biogas yield (380–502 mL/g TS)	[68]
Bio-based carbon	DM/ASW	batch	2.16 g/L	Hydrothermal carbonization	79.37	Improve biogas yield 580.9 mL/g VS	[50]

[d] Up-flow anaerobic sludge blanket (UASB); AW:acorn slag waste; ASW: acorn slag waste.
[a] C: charcoal; PAC: powdered activated charcoal; CSB: coconut shell biochar; AC: activated carbon; GAC: granular activated carbon.
[b] CD: cattle dung; WH: water hyacinth; PW: poultry waste; CW: cheese whey; CS: cattle slurry; M: maize; MS: maize silage; APL: aqueous pyrolysis liquid; CPW: citrus peel waste; DM: dairy manure; WAS: wasted activated sludge; FW: food waste.
[c] SC: semi-continuous; AMB: anaerobic membrane bioreactors.

COD/(m^3 d), and increased the sludge conductivity 9.77 μS/cm [60].

Table 3 evaluates the previous works conducted on bio-based carbon-amended AD system. These studies give comprehensive information about the AD systems supplemented with bio-based carbons, including their digestion period and mode using different substrate. These studies also give the information about synthesis methods of bio-based carbons and their optimal dosages in AD system as additives, and their effects on COD removal rate and biogas yield [116,117]. Generally, bio-based carbon materials are highly recommended for achieving higher COD removal rate (mean complete digestion of organic waste) and for decreasing lag phase in AD reactors, resulting in a greater amount of cumulative biogas yield by absorbing many contaminants [68]. Among them, the biochar and AC are widely used in AD system to improve the process stability and its performance due to their low cost and environmental friendliness. It can be concluded from Table 3 that the AC is more efficient than the biochar because of its larger specific surface area and reliability resource [33]. However, the addition of AC increases the operating charges by almost 50% (3000 dollars per ton AC) in the AD process as compare with biochar. If the AC could be retained for reuse, it will help in lowering operating cost. By contrast, the biochar is cheaper to demand, there is no need to recycle it afterward the AD, and this could recover digestate value and quality of biogas [33].

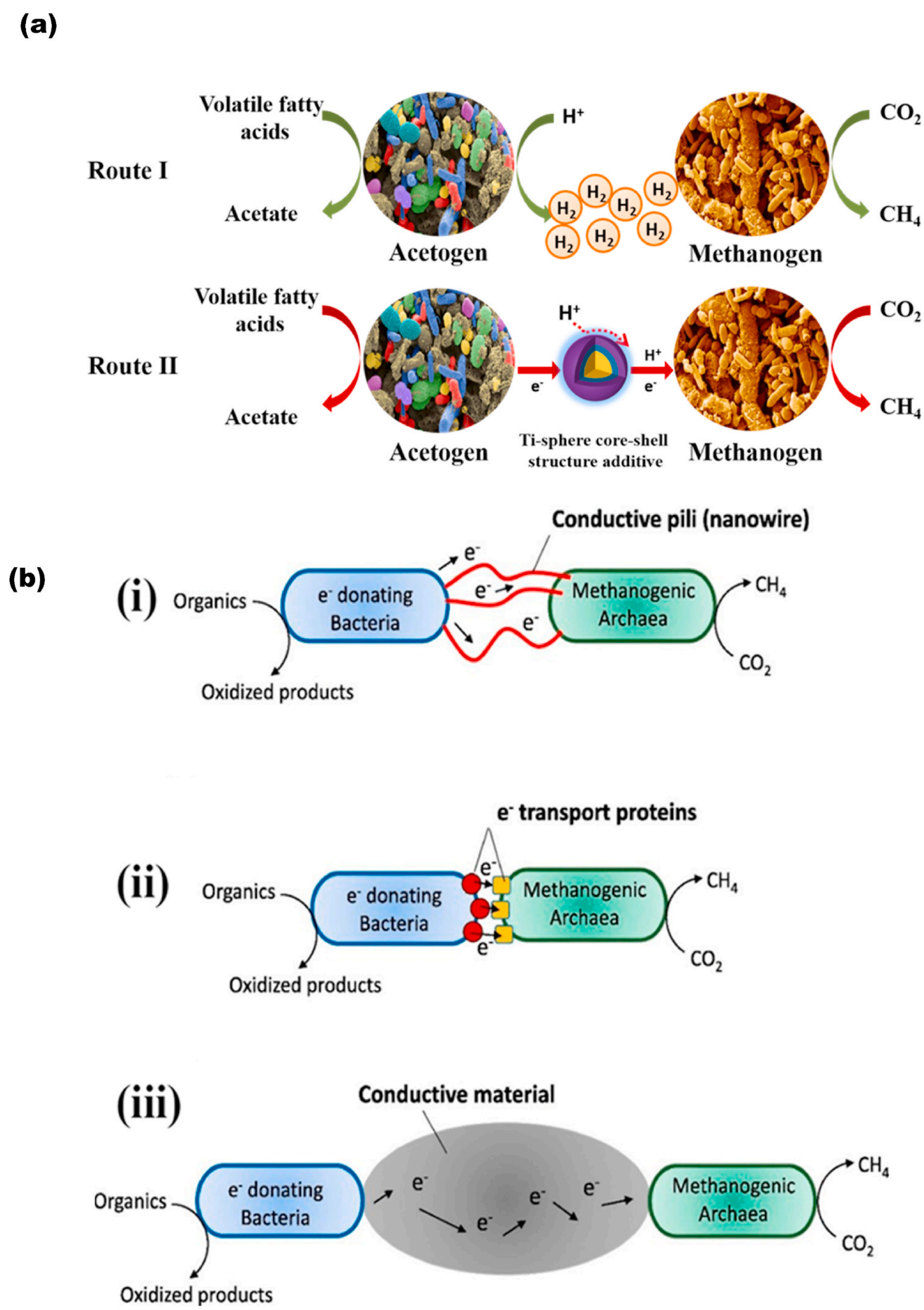

Fig. 3. a) The interspecies electron transfers in AD system via (Route I) IHT, and (Route II) DIET [21], (b) Three different DIET mechanisms concerning methanogenic archaea and organics-oxidizing microbes, DIET via (i) conductive pili. (ii) membrane-bound electron. (iii) conductive material [124].

5. Mechanism of bio-based carbon materials in anaerobic digestion

5.1. Direct interspecies electron transfer (DIET)

Microbes play an important role in the AD system that is a biological-microbial process to produce valuable CH_4. Interspecies electron transfer (IET) between syntrophic partners, which mainly contain Proteobacteria, Bacteroidetes, and Firmicutes, are responsible for converting the alcohol (ethanol) and short-chain fatty acids (SCFAs, i.e. propionate and butyrate) into formate, acetate, methanol, CO_2, and H_2 [118]. The balanced syntrophic affiliation provides a thermodynamically ideal environment to degrade the carboxylic acids and leads to stable AD process. Generally, the syntrophic relation is balanced by indirect interspecies electron transfer (IIET) via electron transporters like formate or hydrogen. Any disruption in this relation can cause the accumulation of VFAs along with an increase in the partial pressure of hydrogen; these can decrease the AD efficiency [119]. Moreover, a syntrophic connection between the *acetogens* and *methanogens* is established for the achievement of a stable AD environment. In the methanogenesis, the acetate is consumed by *acetate-consuming methanogens*, and H_2 as an electron carrier between the *acetogens* and *methanogens* (IHT) as shown in Fig. 3a (Route I), is utilized by *H_2-consuming methanogens* for the production of CH_4. Remarkably, the *H_2-consuming methanogens* could maintain the AD environment with a very low partial pressure of H_2, which is beneficial to the growth of fermentative microorganisms [120,121].

However, recent published scientific works proposed that several microorganisms can directly transfer electrons to *methanogens* [122]. This unique cell-to-cell electron transfer mechanism, known as the DIET, which provides a new methanogenesis pathway and contributes to the effective CH_4 production [123], as shown in Fig. 3a (Routr II).

Howevere, the DIET mechanism possesses considerable biochemical importance and wide application value. In AD system, DIET exhibits more efficient electron transfer with the existence of conductive materials than IHT. In general, there are three different DIET mechanisms used in AD system, including conductive pili-based DIET (Fig. 3b (i)), membrane-bound proteins-based DIET (Fig. 3b (ii)), and conductive materials-based DIET (Fig. 3b (iii)).

Summers et al. firstly proposed the idea of the DIET mechanism that was established in a co-culture of *Geobacter metallireducens* and *Geobacter sulfurreducens*. They found that metabolizing ethanol can promote the development of multispecies groups. These groups are highly electrically conductive and beneficial for improving *c-type cytochrome* production. This *c-type cytochrome* greatly involves extracellular electron transfer and also quickens the development of multispecies groups [125]. The *c-type cytochrome* is well-known to promote DIET between the two groups of microorganisms to be connected via conductive pili (nanowires), as shown in Fig. 3b (i). The physical links were detected in a methanotrophic archaeon co-culture by a *sulfate-reducing bacterium* [126], which was supposed to facilitate a membrane-bound proteins-based DIET [127].

The DIET by membrane-bound electron transportation proteins (Fig. 3b (ii)), the multiheme outer-surface cytochrome (OmcZ), could be a good mechanism for CH_4 production. Up to now, no descriptive reports on this phenomenon are presently available. This proposes that the DIET through membrane-bound electron transportation proteins is very rare for methanogenesis. Ha et al. reported that prosthecochloris aestaurii can accept electrons released from G. sulfurreducens in two bacterial co-culture through the transmission electron microscopy [128]. The OmcZ can easily transfer electrons to anodes [129]. Therefore, OmcZ could be involved in this type of DIET [127]. Mcglynn et al. also observed similar physical associations in a co-culture of a methanotrophic archaea with a sulfate-reducing bacterium [126].

Apart from the conductive pili (Fig. 3b (i)) and membrane-bound proteins (Fig. 3b (ii)), the conductive materials, bio-based carbon materials such as carbon cloth, biochar, GAC, have significant potential for enhancing the DIET process, thus promoting CH_4 production, as presented in Fig. 3b (iii). Remarkably, compared to other conductive materials, the bio-based carbon materials attract more attention owing to the superior physicochemical property and the low-cost preparation. To date, several studies were conducted on the addition of bio-based carbon materials in AD systems. Liu et al. verified that GAC-based DIET could exert a more positive effect on the CH_4 production, in comparison with conductive pili-based DIET considering its higher electrical conductivity, than that of the conductive pili [130]. Furthermore, other several studies reported that bio-based carbon materials such as GAC [131], biochar [23] and carbon cloth [58,59] can serve as electron mediator to stimulate DIET between Geobacter species and methanogens for quickening the syntrophic methanogenesis metabolism [130,132]. Recently, in Yun' group, Chen et al. enhanced the methanogenesis with three different bio-based carbon composits (Co/C, CoO/C, and Co_3O_4/C), and these bio-based carbon composits acted as mediators and enhanced the DIET by efficiently transferring elctrons to methanogenic archaea, which increase the CH_4 yield, as shown in Fig. 4a [11].

5.2. Role of activated carbon in DIET mechanism

As mentioned above, DIET can also easily and efficiently occur through bio-based carbon materials as nonbiological conduits [103]. In bio-based-amended AD system, anaerobes do not make strong physical contact via pili between cells; they only tightly attach to the surface of the bio-based carbon materials. Recently, Zhang et al. found that bio-based AC can improve the methane and lipid metabolism during the AD of food waste, and also encourages the propagation of syntrophic microbes [109].

The electron transfer rate in the GAC-based DIET is much faster than that of the H_2/formate-based electron transportation [134]. Liu et al. proposed that GAC can strongly attach to the microbial cells and improve the DIET mechanism [130]. The conclusive results indicated that the direct electron transmission was occurred by GAC. Moreover, Zhang et al. confirmed that the GAC adsorbed on the surface of the biomass waste play the role of *c-type cytochromes* in transferring electrons, thus establishing the enhanced methanogenesis [109] (Fig. 4b).

Additionally, the microscopic analysis of GAC-amended co-culture demonstrated that both Methanosarcina barkeri and Geobacter metallireducens species were attached on the surface of GAC without any electrical network induced by conductive pili, thereby indicating that the GAC could play the role of conductive pili [130]. In this regard, Peng et al. provided the experimental proof that the GAC improved the syntrophic metabolism among methanogens and iron-reducing microbes because of its large surface area and remarkable electrical conductivity. In addition, the addition of magnetite-GAC speeded up sludge hydrolysis process and the CH_4 production resulted in better AD performance of sludge [113].

5.3. Role of carbon cloth in DIET mechanism

The carbon cloth is an excellent bio-based carbon material for fabricating the electrode used in bacterial and electrochemical systems [47,135–139]. Up to now, few works have conducted on the use of carbon cloth to enhance the AD performance by improving the DIET [140]. For instance, Chen et al. investigated the feasibility of using carbon cloth to improve the DIET kinetics, and exposed that *c*-type cytochromes and *Geobacter* strains lacking pili enable the electron exchange with *methanogens* (*methanosarcina barkeri*) species in the existence of carbon cloth [54]. In another work, during the AD process of sludge, Zhao et al. found that the sludge attached to the surface of carbon cloth was rich in *Methanosaeata* and *Geobacter*species, which were well-known species to take part in the DIET process, and the major mode of the interspecies electron transfer might be changed from IHT to

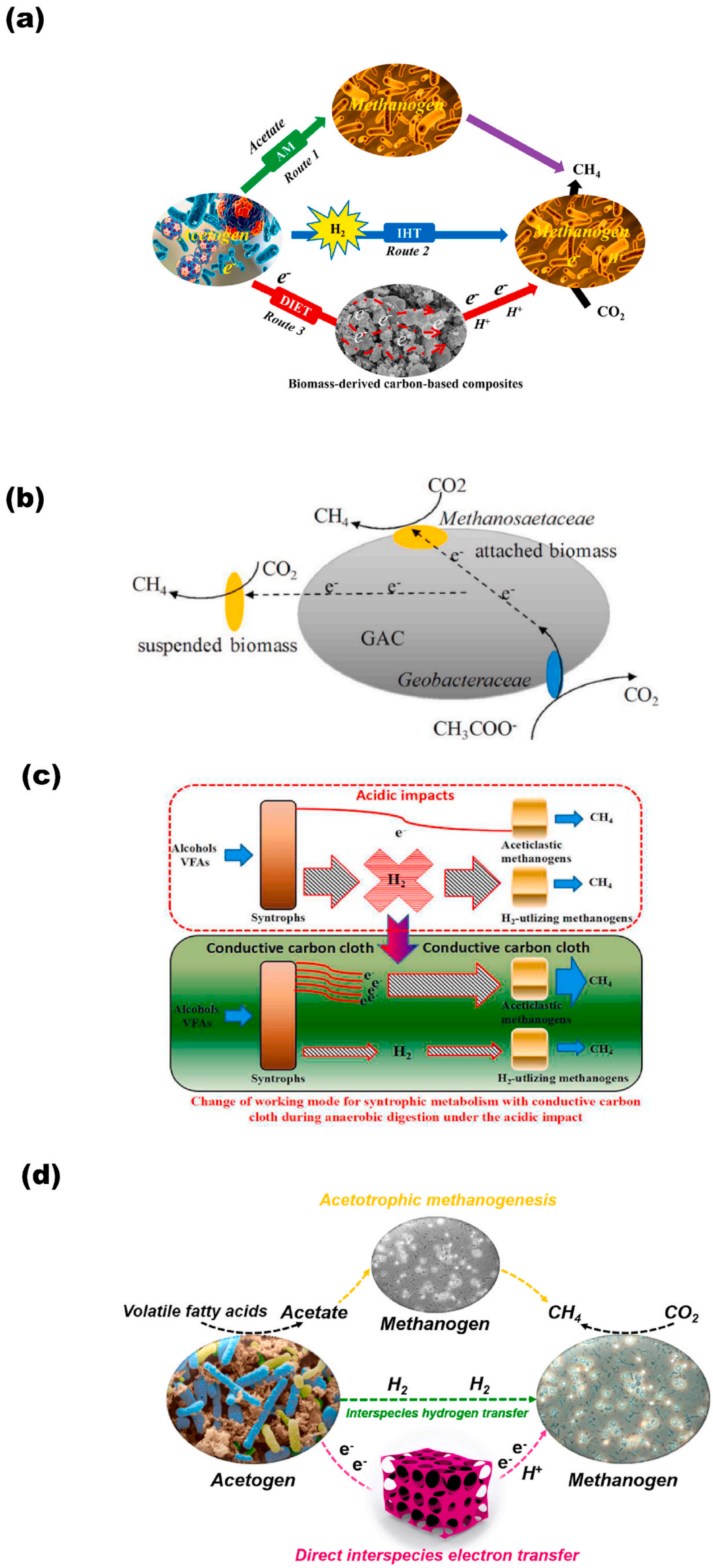

Fig. 4. (a) Schematic illustration of enhanced methanogenesis pathways with bio-based carbon composits [11], (b) Promotion of methanogenesis via GAC-based DIET [133], (c) Change of working mode for syntrophic metabolism with carbon cloth during AD under the acidic impact [114], (d) Methanogenesis pathway of the BC-enhanced AD systems based on DIET mechanism, pathway I (yellow dash line): acetate-dependent methanogenesis; pathway II (green dash line): H_2–dependent methanogenesis; pathway III (pink dash line): electron-dependent methanogenesis [50]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

DIET during the acidogenic stage, as shown in Fig. 4c. In addition, the DIET appeared on the surface sludge of the carbon cloth can assist the IHT mechanism for maintaining the syntrophic metabolism, thus achieving the stable methanogenesis.

5.4. Role of biochar in DIET mechanism

Biochar is another high-performance bio-based carbon material [141]. Several investigations were carried out on the utilization of biochar in AD system to decrease the methanogenesis inhibitions, which were caused by the accumulation of acid and NH_3 [100,142]. The enhanced biochar-based DIET between the *Methanosarcina barkeri* and *Geobacter metallireducens* was demonstrated by Chen et al. and Zhao et al. [58]. They observed that the biochar enhanced the 86% electrons retrieval as CH_4 production from ethanol, equivalent to 77% electron recovery noted with AC [130]. Moreover, Chen et al. recommended that the biochar-based DIET might be comparable to the GAC-based DIET by using microscopic analysis [54]. Wang et al. investigated the effect of bio-based carbons on the co-digestion system and proposed the DIET mechanism based on the bio-based carbon, as presented in Fig. 4d, in which the electrons obtained from the decomposed VFAs could be directly transferred to specific methanogenic archaea via the well-conductive bio-based carbon materials when the acetogens metabolize. Three methanogenesis pathways, namely, acetate-dependent methanogenesis, H_2-dependent methanogenesis, and electron-dependent methanogenesis, existed in the co-digestion system after bio-based carbons were supplemented. It was concluded that the establishment of an enhanced biological interspecies electrical connection by adding the bio-based carbons accelerants in AD could effectively promote the consumption of VFAs, thus alleviating the inhibition of acid accumulation and creating a favorable environment for methanogenesis [50].

The DIET mechanism of bio-based carbon materials (biochar, AC, and carbon cloth) in AD systems are summarized in Table 4, and the effects of bio-based carbon materials on AD performance are also demonstrated. These works illustrated that bio-based carbon materials could serve as mediators (conduits) for directly transferring electrons among the syntrophic microorganisms, and thus promoting the capacity of methane production in AD systems. The bio-based carbon materials can facilitate the syntrophic VFAs oxidation and CH_4 production from complex organics via DIET, [53,143]. For example, AC has been applied as an additive in AD systems for improving DIET by providing effective microbial attachment due to its relatively large surface area [144]. The ability of biochar for improving DIET [12,145] is similar to that of AC, but its conductivity is about 1000 times less than that of AC [136]. This suggests that DIET can be stimulated with very low electrical conductivity. In another study, Dang et al. confirmed that the GAC could better improve the DIET mechanism than carbon cloth owing to its larger surface area [59]. Hence, the GAC is more efficient and effective additive for DIET and AD performance due its higher conductivity and larger surface area than biochar and carbon cloth.

It can be found from Table 4 that the electron-donating bacteria or electron donor (*Geobacter species*) transfer electron to an electron acceptor (*methanogens*) through bio-based carbon materials [103,124]. These syntrophic partners (*Geobacter species and methanogens*) easily enrich on the large surface of bio-based carbons materials and use them as electrical mediators for electron transferring [58]. It was a strong evident from these works that bio-based carbon materials can enhance methanogenesis (CH_4 yield and production rate) by promoting DIET. Moreover, in the presence of an inoculum (co-culture or mixed culture) *Geobacter species, Thauera, Corynebacterium, Spirochaeta, Clostridium, Coprothermobacter, and Syntrophomonas species* donate electrons to bio-based carbon materials [60,134]. The bio-based carbon materials transfer these electrons to methanogens (electron acceptor) including *Methanosaeta* and *Methanosarcina* species, *Methanospirillum*, *Methanobacterium*, *Methanolinea*, and *Methanoregula* species to enhance

Table 4
Use of various bio-based carbons in AD systems for DIET mechanism.

Additives	Inoculum	Enriched bacteria (electron donor)	Enriched archaea (electron acceptor)	Role and effect of bio-based carbons	Ref.
GAC	Co-culture[a]	Geobacter metallireducens	Methanosarcina barkeri	Reduce significantly lag phase.	[181]
GAC	Mixed[b]	Geobacter metallireducens	Geobacter metallireducens	Increase methane yield rate increased by 2.5 times.	[182]
GAC	Mixed[b]	Geobacter species.	Methanolinea species.	Improve methane production rate by 1.8 times.	[178]
GAC	Mixed[b]	Geobacter species.	Methanosaeta species. Methanosarcina species.	Improve butyrate and propionate degradation and methane production rates over control.	[144]
GAC	Mixed[b]	Sporanaerobacter species	Methanosarcina species.	Improve methane yield rate by 18 times.	[110]
GAC	Mixed[b]	Sporanaerobacter species	Methanosarcina species.	Enhance methane yield rate was about 13 times at OLR (8.5 kg COD/m^3 d).	[59]
GAC	Mixed[b]	Coloramator species.	Methanosaeta species.	Increase methane yield rate by 2.68 times.	[153]
Biochar	Mixed[b]	Bacteroidales	Methanosarcina barkeri	Improve methane yield rate improved by 30–45%.	[58]
Biochar	Co-culture[a]	Geobacter metallireducens	Methanosarcina barkeri	Observed no methane production without biochar.	[183]
Biochar	Mixed[b]	Syntrophomonas species. Clostridiaceae	Methanosaeta species. Methanosarcina species.	Shorten the lag phase improved methane yield	[168]
Biochar	Mixed[b]	–	–	Enhance methane yield rate by 1.15 times.	[12]
Carbon cloth	Mixed[b]	Bacteroides species. StreptococcusSyntrophomonas	Methanosarcina species. Methanospirillum	Enhance organic loading rateabout 34.2% over control.	[60]
Carbon cloth	Mixed[b]	Geobacter species.	Methanosaeta species.	Enhance methane yield rate by 59%.	[114]
Carbon cloth	Mixed[b]	Sporanaerobacter species	Methanosarcina species.	Enhance methane yield rate by 9.7 times.	[110]
Carbon cloth	Mixed[b]	Sporanaerobacter species	Methanosarcina species.	Increase methane yield rate was ~13 times at great OLR (8.5 kg COD/m3 d).	[59]
Carbon cloth	Mixed[b]	Bacteroides species.	Methanosarcina species.	Increase methane yield rate by 1.3 times.	[60]
Carbon cloth	Co-culture[a]	Geobacter metallireducens	Methanosarcina barkeri	Increase ethanol degradation and Methane yield rates.	[54]
AP-BC/AS-BC	Sewage sludge	–	–	Increase the biodegradation and biogas yield	[50]

[c]AP-BC: bio-based carbon derived from aloe peel waste.
[d] AS-BC: bio-based carbon derived from acron waste.
[a] Co-culture of Geobacter metallireducens and Methanosarcina barkeri.
[b] Anaerobic digester sludge.

biogas production by promoting DIET [124].

6. Drawbacks of bio-based carbons in anaerobic digestion

Bio-based carbons can also impose negative impacts on biogas yield due to composition of biomass from which they made. Indren et al. investigated the effect of biochar on AD performance made from wood pellets, wheat straw and sheep manure. It was noted that the biochar from wood pellets enhanced the methane yield about 32%. But in contrast, the biochar from wheat straw and sheep manure has harmful effects on AD process [146]. The main reason was the biochemical composition of wood pellets contained lower ash content (0.3 $\pm$ 0.4% of TS) than in wheat straw (14 $\pm$ 2.0% of TS) and sheep manure (58 $\pm$ 0.2% of TS). Thereby the wood pellet had the least sodium (Na), magnesium (Mg), Sulphur (S), potassium (K) and calcium (Ca). The fact about detrimental effect of these elements on AD process had been exposed by previous works [77,147]. Moreover, Liu et al. exposed the cytotoxicities of biochar (adding 10 mg/L) to microorganism. They found that original biochar has less toxic effects on gram positive bacterium *Streptomyces species* which are responsible to secrete antibiotics. In contrast, the milled biochar together with graphene and multi-walled nanotubes can sever toxicity, and damage the cell badly that limits its survival rate [148].

Moreover, bio-based carbons are fabricated by lignocellulosic biomass wastes (wood, animal manure, waste activated sludge). Hence, the bio-carbons have major issue of environmental pollution related to higher risk of heavy metals and organic compounds contaminants such as PCBs, dioxins and PAHs [99]. However, Lian et al. exposed that the addition of pesticides such as HCHs in CSTR showed no negative influence on conventional reactor parameters and methanogenesis at the concentration range found in biomass grown on contaminated areas [149].

7. Current challenges and future outlooks

The current challenges related to bio-based carbons, and their future outlooks for AD are discussed.

- It has been proved that the bio-based carbon materials would not properly stimulate DIET for complex organics, specifically municipal sludges. For developing such microbes, it is essential need to develop favorable conditions for them to support healthy growth over several competing microorganisms. In this regard, such type of reactor is constructed that can produce more alcohols and fatty acids for inspiring electron-donating bacteria, such as *Geobacter species*, [124]. The constructed reactor being responsible for two-stage AD [150] could provide favorable conditions to enrich electron-donating bacteria [114].
- The long-term stability of AD system amended with bio-based carbons in field applications needs clarification. The potential environmental risks of digestates containing bio-based carbons should be measured in future research. Detailed techno-economic analysis of bio-based carbons including their fabrication should be achieved to observe the total advantages and drawbacks on large scale.
- Even though several studies have proposed many conceivable models to clarify the enhanced fermentation performance in AD system amended with bio-based carbons, significant proof on the electrical structure, features related to surface adsorption, and electrical properties of bio-based carbons in AD systems, are still blurred. Therefore, advance logical research, including first-principles density functional theory calculations and ab initio Car-Parrinello molecular dynamics similations performed on the interface of solid/liquid in AD system [70,136], electrical properties, adsorption properties, etc, will be critical for complex and changeable AD system to identify and confirm the specific mechanisms of carbon materials which improve the AD performance.
- The revolutionary new technologies of generating useable energy from sustainable and non-polluting sources must have the flexibility to meet the energy demands of both producers and users while producing fewer carbon emissions than traditional energy systems. Regarding connections between carbon materials structure and various operational conditions of the AD system play a critical role in order to enhance the AD performance and overcome the related critical challenges. Continued research in this direction will focus on enhancing the CH_4 production and digestate nutrient in the future. Besides, more attention should be paid to explore the interaction of carbon-microbe and the effects on continuous-feed digesters based on both experimental and theoretical investigations using the first-principles density functional theory calculations [70,136,137,151–153].

8. Conclusion

The effect of additives addition expressly bio-based carbons on AD system are summarized in this review. The bio-based carbons as additives can increase COD removal rate, biogas yield, and enhance the stability of AD process. The effects of adding bio-based carbons on enhanced DIET mechanisms in AD performance are also commented briefly. This review also puts light on the fabrication methods of bio-based carbons, and also exposes the advantages and disadvantages of these methods. Moreover, the key challenges related to bio-based carbon addition in AD system are highlighted, and their future outlooks are also advised. Hence, the application of bio-based carbons in the AD system can encourage the biogas industry, in order to meet future energy challenges.

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.

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