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Review Article

Different Pathways to Integrate Anaerobic Digestion and Thermochemical Processes: Moving Toward the Circular Economy Concept

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Abstract

As one of the most environmentally friendly and cost-effective method, anaerobic digestion (AD) has been widely studied and developed as a conventional technology to degrade biodegradable materials and produce biogas simultaneously. Various substrate sources are used in this process such as organic fraction of municipal solid waste (MSW), waste activated sludge (WAS), animal manures, agro-industrial wastes, energy crops, micro- and macro-algae and etc. With the aim of process optimization, several publications have recently studied different configurations to integrate AD and thermochemical processes such as pyrolysis and gasification. These linking technologies seeks to optimize the use of products or by-products of thermochemical processes interchangeably. In this regard, this paper aims to review different potential pathways of feasible integration and coupling. Five hybrid pathways including biochar-amended anaerobic digestion, digestate-derived biochar and hydrochar, anaerobic digestion of aqueous phase liquid derived from pyrolysis and gasification of digestate were reviewed and their schematic diagram were presented. Despite several studies to combine AD with thermochemical valorization processes, further studies at the industrial scale are needed to prove the energy efficiency and economic viability of these coupling pathways.

Keywords: Anaerobic digestion; Circular economy; hydrothermal carbonization; Pyrolysis

Introduction

There are different biochemical, thermochemical and physicochemical processes and technologies to covert biomass and waste to added-value materials and energy. As one of the most convenient biochemical conversion process, anaerobic digestion is an environmentally friendly technology not only to recover energy from different organic waste streams such as municipal solid waste (MSW), waste activated sludge (WAS), animal manures and etc. but also to reduce environmental pollutions and greenhouse gases emissions (Angelidaki et al., 2011; Appels et al., 2008). During anaerobic digestion, natural polymers such as polysaccharides (carbohydrates), lipids (fats/oils) and proteins are converted to methane and

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carbon dioxide during several biochemical stages including hydrolysis, acdidegenesis, acetogenesis, and methane generation, which are mediated by a number of different groups of microorganisms (Schnurer and Jarvis, 2010).

Biogas and digestate as the two main products of anaerobic digestion play an increasing important role in future of world circular economy or/and bioeconomy by improving resource recovery and nutrient recycling (Mohan et al., 2016). Produced biogas can be compressed and used as a transportation fuel or can be recovered in combined heat and energy (CHP) systems. It is expected that biogas has at least 25% share in growing global bioenergy market in future (Holm-Nielsen et al., 2009). Moreover, if managed properly, nutrient rich and stabilized digestate has a tremendous potential to be applied instead of chemical fertilizer in agriculture sectors. Although AD is known as a promising way for mitigating climate change and reducing dependence on fossil fuels, it seems that this technology still faces some limitations and challenges in operation and commercialization. The main obstacles and challenges for the development of this technology include: (i) presence of recalcitrance substrate and inhibitory compounds lower the rate of production (ii) non-stabilized digestate which is associated with lower quality because of defective operation (iii) instability of biological system performance over time. To overcome these drawbacks, a great deal of research has been conducted to promote biogas yield and biogas production rate. It is noteworthy, facilitate biogas production and increasing the content of methane of biogas also lead to increased quality of digestate.

Different thermochemical conversion processes such as gasification, hydrothermal carbonization (HTC) and pyrolysis have been developed. Unlike the anaerobic digestion process, thermochemical processes is able to convert recalcitrance biomass to energy and other value-added products in a short time without any concern about microbial community fluctuations. The moisture content of feedstock is the most important limiting factor in thermochemical processes, which should be less than 30%.

Gasification is a chemical process that transforms different organic matter into a combustible gases including CO, H2, and CH4 called syngas by indirect heating at elevated temperature from 800 to 1200 C° in a sub-stoichiometric condition. By-products of the gasification process include a solid carbonaceous called char and tar which is a mixture of different polycyclic aromatic hydrocarbons.

Pyrolysis is a thermochemical process which decompose biomass (such as wood, manure or crop residues) by heating in the absence or low concentration of oxygen. Different feedstocks such as woods, animal manures, agricultural wastes have been widely used in pyrolysis process for biochar preparation. Three main products of pyrolysis include pyrolysis liquid (50–60%), charcoal like solid named biochar (20%*30%) and non-condensable gas (10%-20%).

HTC is an appealing thermochemical conversion process specially concerning high moisture biowaste, which converts feedstock into hydrochar, varying temperature (190–250 ˚C), residence time (3 and 6 h), pH (5 and 7), and pressures (2–10 MPa) in the presence of liquid water.

In recent years, some studies have focused on coupling different biomass conversion technologies to optimize coupled pathway comprehensively. It is believed that this strategy may be desirable from an environmental and economic point of view especially concerning circular economy concept and biorefinery concept (Geng et al., 2013). Anaerobic digestion has extensive opportunities to combine with other biochemical, thermochemical and physicochemical processes such as microbial electrolysis cells (MECs) (Yin et al., 2016, Lee et al., 2017), microalgae or/and macro algae (Gonzalez-Fernandez et al., 2015, McKennedy and Sherlock, 2015, Ward et al., 2014), gasification (Li et al., 2016), pyrolysis (Salman et al., 2017, Fabbri et al., 2016) and hydrothermal carbonization. However, the focus of this study is on beneficial integration of anaerobic digestion with specifically thermochemical process including HTC and pyrolysis. The number of cited articles that studied one of the five aforementioned coupling pathways have been reported in figure 1, subdivided for different feedstocks.

Different pathways of integration:

Anaerobic Digestion and Gasification
Gasification of digestate

This coupling method is focused on energy recovery from digestate using gasification process. Kan et al., 2017 applied an AD technology as a pretreatment of lignocellulosic biomass to increase efficiency of gasification process. This hybrid system could not improve the quality and composition of produced syngas significantly, but gas production overally improved because of the high quality of biogas compared to syngas. Similarly, pretreatment of corn straw by AD in a short time (7-14 d) increased lower heating value (LHV) 8.7–10.69% at 600 °C, and 11.71–13.18% at 700 °C, and 5.49–8.06% at 800 °C, respectively (Chen et al., 2017). In another study, co-gasification of digestate with woody biomass was investigated numerically and experimentally which indicates the optimal mixing ratio of 20 wt% digestate with 30 wt% moisture content (Yao et al., 2017).

For digestate gasification, the digestate needs to be pre-dried. However, high moisture content and high ash content of digestate have undermined the economic viability of this approach. Techno-economic assessment showed that gasification of solid digestate from anaerobic digestion of food waste is economically unfeasible (Singlitico et al., 2017). However, this study stated that the establishment of a gasification unit could supply the required energy of the AD plant. Antoniou et al., 2019 showed that the heat excess generated from the AD process can be consumed for pre-drying of the digestate. Moreover, add gasification to the existing process increases electricity production that makes this combination economical. Figure 1 illustrates coupling pathway for gasification of digestate.

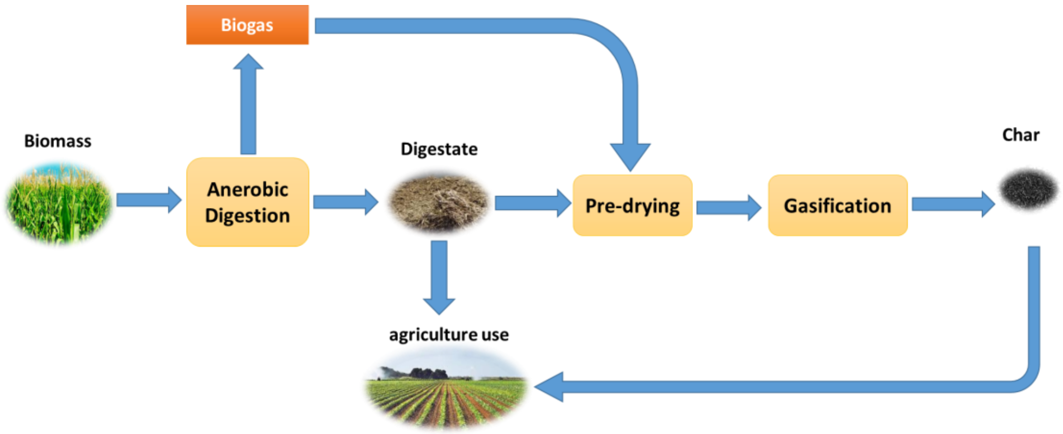

Figure 1. Coupling pathway for gasification of digestate

Anaerobic Digestion and pyrolysis
Biochar-amended anaerobic digestion:

Biochar, the carbon-rich solid, exhibited an extensive application due to the interesting features such as highly-porous structure, functional groups and large specific surface area (Lehmann and Joseph, 2015). Biochar usage in soil is well recognized as a promising way to sequester CO2, increase crop yield and improve nutrient availability. Besides that, other applications of biochar and its performance in water purification, air decontamination, detoxification have been investigated in recent studies.

Several publications have recently explored possible effects of biochar supplementation on AD process. Many important researches are recently devoted to find out the main governing mechanisms of biochar addition to anaerobic digestion. Mumme et al. 2014 showed hydrochar has capability not only to enhance biogas yield by 32% but also to prevent ammonia inhibition. To the best of our knowledge, four governing mechanisms in biochar-AD interaction to improve biogas yield were suggested: (I) adsorption of inhibitors, (II) improvement of buffering capacity of the digester, (III) immobilization of microbial cell, and (IV) reinforcement of direct inter-species electron transfer (DIET) (figure 2) (Fagbohungbe et l., 2016; Luo et al., 2015).

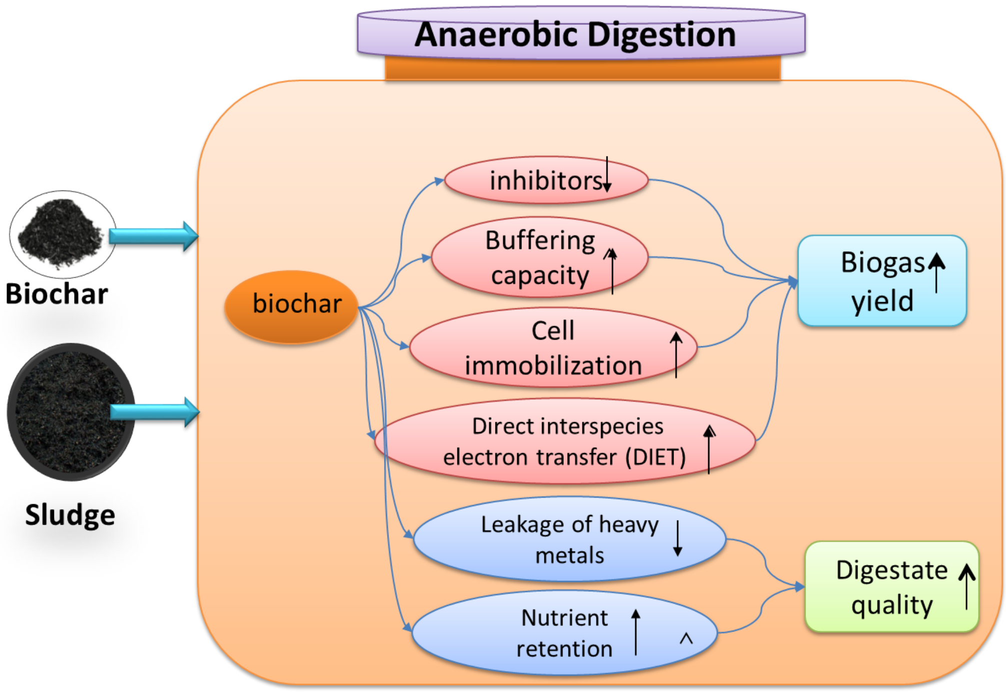

Figure 2. Schematic diagram of governing mechanisms in biochar-amended anaerobic digestion

Anaerobic digestion of nitrogen-rich matters, such as livestock manure, results in high concentrations of ammonia. Although, ammonia is a nutrient for bacterial growth, in high concentration it could cause the anaerobic digestion fails. Accumulation of ammonia-nitrogen in digesters can limit methane production via toxicity to microorganisms, and the digester may require energy intensive treatment downstream to avoid adverse environmental effects from nitrogen release (Chen et al., 2008, Rajagopal et al., 2013, Yenigün and Demirel 2013). Management of ammonia liberated during the degradation of organic matter containing nitrogen is a particular concern during anaerobic digestion. Some kinds of biochar have metal oxides (MgO, CaO, CrO, Fe2O3) on their surface that increase ammonia absorption. Taghizadeh et al. 2012 showed that biochar has excellent capability to absorb ammonium and prevent its emission to atmosphere, while slowly releasing ammonium for plant consumption in soil environment. Lu et al. 2016 highlighted that the use of biochar obviously reduces inhibitory effect during the anaerobic digestion of glucose at a concentration of 6 g / l and 7 g / l of ammonium. Fagbohungbe et al., 2016, studying the effect of lemonin inhibition on orange residue, indicated that the addition of biochar has a positive effect on biogas production rate and lag phase reduction of the process.

Biochar could improve the buffering capacity of the system. Luo et al., 2015 considering two glucose anaerobic digestion reactors with and without biochar addition reported that production of methane in the first system was 86.6% higher, while acidity decreased. Likewise, Sunyoto et al. 2016 indicated that biochar supplementation not only improved hydrogen and methane yields by 31.0% and 10.0%, but also decreased the lag phases in the two steps by 36.0% and 41.0%, respectively. Immobilization of microbial cell in anaerobic digestion is very critical, because it can facilitate syntrophic associations between acetogenic bacteria and hydrogen-consuming methanogenic archaea. It seems that biochar can serve as an excellent support carrier to provide anaerobic microorganism growth and thus promote the biofilm formation (Zhang et al., 2017). According to Cooney et al., 2016, biochar has ability to act as a packing material in order to facilitate the growth and retention of methanogenesis-rich biofilms resulting high performance of anaerobic digester.

Besides, studying the use of cotton and carbon cloth on the activity of bacterial symbiosis in the anaerobic digestion process showed that the latter case clearly had a positive effect on the syntrophic bacteria and archaea activity and methane production, attributed to electron transfer capability of carbon materials (Chen et al., 2014). Lee et al., 2016 showed that granular activated carbon (GAC) has the ability to provide condition for direct interspecies electron transfer (DIET) through receiving an electron as an anode-reducing microorganisms and transfer it to the cathode-oxidizing microorganisms (methanogenic archaea) due to its conductive feature, thus led to increase methane production up to 1.8 times. Despite the low conductivity of biochar, it was documented that biochar can stimulate direct electron transfer as a biological circuit, that facilitates syntrophic interactions and methane production (Zhao et al., 2016).

Leaching digestate of anaerobic digestion can lead to serious environmental impacts such as eutrophication and acidification as well as adversely influence human beings. On the other side, considering the high content of valuable macro and micronutrients such as phosphate, ammonium, micronutrients and metal in digestate, nutrient retention capacity improvement is a sustainable solution to develop naturally closed loop concept. In recent years, positive effect of biochar on soil nutrient availability has been repeatedly proven in several studies. It is also expected that biochar improve digestate quality by increasing nutrient retention capacity. According to Shen et al., 2015, compared to control condition, the biochar addition significantly enhanced concentration of K, Ca, Mg and Fe in digestate up to 4435%, 134%, 183%, 95%, respectively, thus can be applied as a valuable agricultural bio-fertilizer. Likewise, Shen et al., 2016 showed that the nutrients (P, K, Ca, Mg, Fe) of digestate increased by up to 33 times through adding corn stover biochar to anaerobic digestion.

Digestate-derived biochar (pyrolyzing of digestate):

A novel method of using digestate is pyrolyzing it to produce biochar, which could be used in various application such as soil amendment, water purification, air decontamination and etc (Atkinson et al., 2010, Hung et al., 2017, Peng et al., 2016). This approach could significantly decrease nutrient leakage in soil as well as reduce the N_2O emissions. A comparison between anaerobically digested bagasse biochar and bagasse biochar characteristics indicated more suitable characteristics of the latter forz various applications such as fertilizer, soil ameliorant and adsorbent, makes it economically and environmentally viable (Inyang et al., 2010). Biochar produced from digestate showed the highest adsorption capacity of methylene blue dye compared to other feedstocks, which achieved an adsorption efficiency of 9.60 mg g^{-1} (Sun et al., 2013). In another study, biochar characteristics produced from solid separated and unseparated digestate were investigated. Results indicated that unseparated digestate does not have suitable parameters as a soil amendment because of the low specific surface area and carbon content as well as the highest salinity. Therefore, separated digestate is a good option for biogas residue valorization (Stefaniuk and Oleszczuk 2015). Yao et al., 2011 characterized two biochars were produced from anaerobically digested and undigested sugar beet tailings. Digestate biochar had higher surface area compared to undigested biochar, as well as showed the highest phosphate removal capacity. Figure 3 suggests a more sustainable way for digestate handling through chemically charring process (pyrolysis or HTC) from the viewpoint of circular economy. Figure 3 suggests combination pathway for digestate-derived biochar.

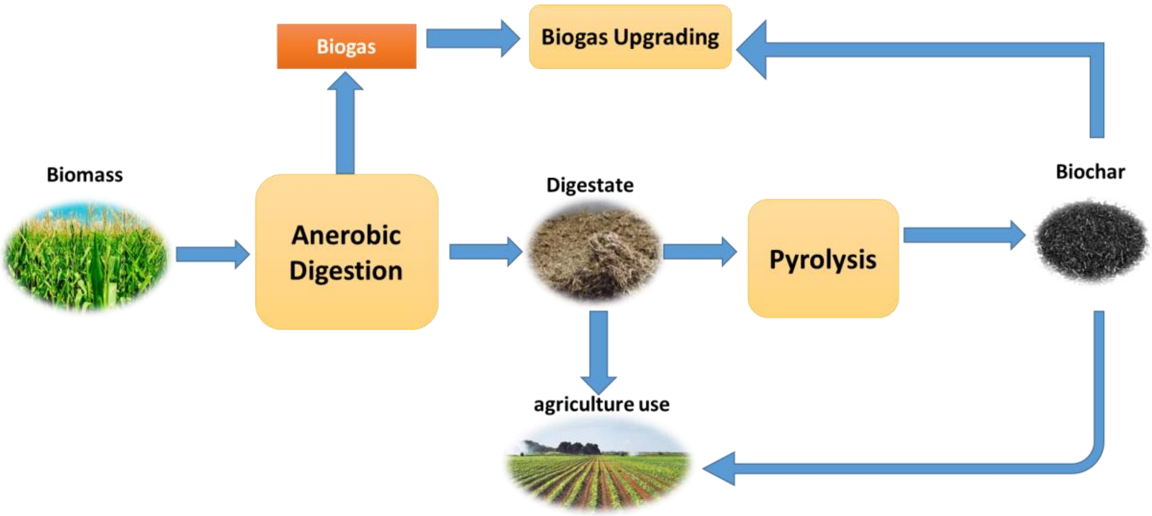

Figure 3. Coupling pathway for digestate-derived biochar

Anaerobic digestion of aqueous phase liquid derived from pyrolysis

As mentioned earlier, one of the main product of pyrolysis technology is a liquid phase containing different organic substances including C_2–C_6 sugars, hydroxyacids, oligomers, phenols and furans (Hubner and Mumme, 2015). Despite the low heating value of aqueous phase liquid (APL), some research has recently focused on recovering APL because of its high contribution in pyrolysis products (about 50%). Several publications have applied anaerobic digestion as a sustainable approach for APL conversion process to stabilize different organic compounds. However, presence of some toxic substances such as water soluble phenols may inhibit and microorganism activity and slows down biological process, which should be noted to find solutions. Yang et al., 2018 showed that temperature of pyrolysis process is a main factor for toxicity of APL, as the higher the pyrolysis temperature, the more toxicity. Acclimatization of microbial population of AD process is necessary to make the use of APL reasonable. This study also expressed that although use of APL is considered as a method of improving energy recovery, this performance improvement is not high enough to make the building a biological unit attractive.

According to Torri and Fabbri 2014, anaerobic digestion of APL showed a low biomethane yield and nutrient supplementation did not have a significant effect to improve process, but biochar addition could increase biomethane yield (60 ± 15% of theoretical) compared to APL (34 ± 6% of theoretical). On the other side, investigating different APL from pyrolysis of digestate at three temperatures (of 300, 400, and 500 ˚C) showed remarkable biogas yield especially at lower temperatures (of 300 and 400 ˚C) without any additives, whilst high initial COD dosage (30 g/l) created some inhibitory effects. Figure 4 offers a coupling pathway for anaerobic digestion of APL (Hubner and Mumme, 2015).

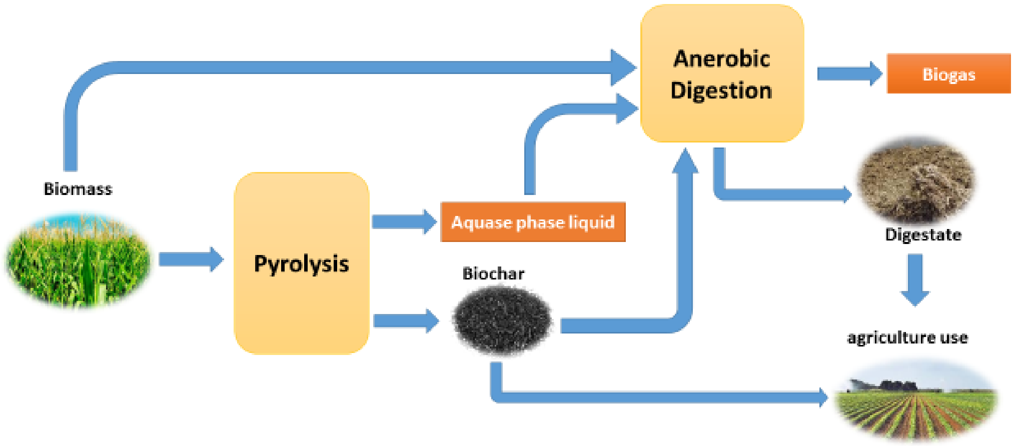

Figure 4. Coupling pathway for anaerobic digestion of APL derived from pyrolysis

Anaerobic Digestion and hydrothermal carbonization
Hydrothermal carbonization of digestate

Hydrothermal carbonization process is a good choice for energy recovery from wet feedstocks such as digestate. In comparison with unprocessed digestate, digestate-derived hydrochar could considerably sequester carbon in soil media over long period of time (Reza et al., 2015). Considering high content liquid phase of digestate, dewatering and separation process of solid phase of digestate is energy intensive, led to move toward HTC as an attractive option. Applying digestate in HTC process has been recently investigated by several researches in order to improve energy recovery and fertilization characteristics. Mumme et al., 2011 prepared hydrochar from hydrothermal carbonization of digestate from two stage thermophilic anaerobic digestion of maize silage. This study indicated higher temperatures enhanced the hydrochar's carbon content but decreased its surface area. In another study, investigating the hydrochar yields of anaerobically digested straw and straw showed higher yield for the latter, which can be attributed to its higher lignin content (Funke et al., 2013). For recalcitrant biomasse such as straw, an important part of original energy remains in residue after AD process, can be recovered by applying HTC process. Furthermore, produced hydrochar could recovered majority of nitrogen (60–65%) and phosphorus (77–80%). In another research, investigating HTC of sewage digestate showed that hydrochar yield drops with temperature (Aragón-Briceño et al., 2017). In another research, investigating HTC of sewage digestate showed that hydrochar yield drops with temperature (Aragón-Briceño et al., 2017). Correa et al., 2017 focused on generating activated carbon from digestate hydrochar to utilize as a gas adsorbent. Chemical activation of hydrochar have significantly enhanced absorption capacity due to its high surface area, named activated carbon, which shows a great capacity for gas adsorption specifically for CO2, thus can be applied for upgrading biogas to biomethane process. Although the specific surface area of produced hydrochar increased dramatically during the activation process from 8–14 m^2g^{-1} to 930–1351 m^2g^{-1}, high cost of activation process and very low yield were the two main limiting factors. Applying HTC for microalgae digestate improve its biodegradability by 4 times, which resulted to increase methane yield of digestate to about 200 L_{STP} CH_4/kg volatile solids (Nuchdang et al., 2018). Some hydrochar properties including low surface area, presence of polycyclic aromatic hydrocarbons (PAHs) and phenolic matter have relatively limited its usage as a soil amendment or wastewater treatment adsorbent (Nakason et al., 2018). With the aim to overcome these, Garlapall et al., 2017 reported that pyrolyzing the produced hydrochar from HTC process (pyro-HTC) could significantly modify various characteristics, for example surface area increased compared to hydrochar but was still lower than biochar, pH is more basic than both previous approach as well as the amount of PAHs recognizably decreased compared to hydrochar and biochar. Figure 5 illustrates the combination pathway for HTC of digestate.

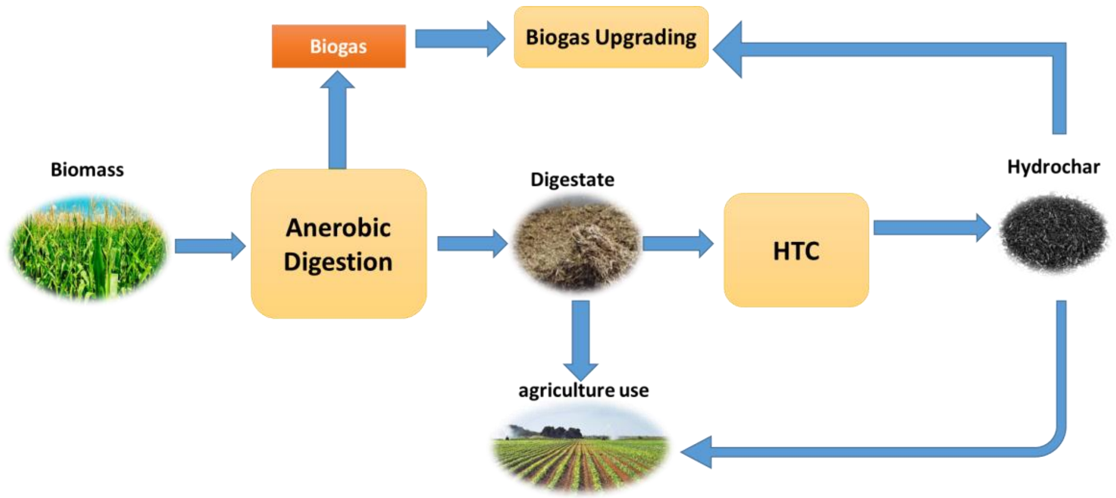

Figure 5. Coupling pathway for HTC of digestate

Conclusion

It is believed that integration of different biomass conversion processes may be desirable from an environmental and economic point of view and play an important role for extending the concept of circular economy in future. This approach attempts to optimize the use of products or by-products of different biomass conversion processes interchangeably. In this regard, this study aimed to briefly provide new prospects of potentially different pathways of AD-thermochemical integration in order to couple technologies and develop naturally closed loop concept. Five hybrid pathways including biochar-amended anaerobic digestion, digestate-derived biochar, anaerobic digestion of APL and gasification of digestate were reviewed and their schematic diagrams were presented.

Applying biochar to optimize AD process and improve biogas production has been studied extensively in recent years, which has shown improvement in process performance. For gasification of digestate, pre-drying of digestate as an energy consuming process is necessary, that have undermined the economic viability of this approach. However, AD is an effective pretreatment process for lignocellulosic biomass gasification which significantly improve LHV of syngas. Although pyrolyzing of digestate well improves its properties for the use as soil ameliorant, pre-drying of digestate is a limiting issue. Some properties of digestate-derived hydrochar such as low surface area, presence of polycyclic aromatic hydrocarbons (PAHs) and phenolic matter have relatively limited its usage as a soil amendment or wastewater treatment adsorbent. Concerning the anaerobic digestion of APL, low biomethane yield has made this combination less attractive but biochar addition is recommended to increase biomethane yield. Finally, despite several studies to combine AD with thermochemical valorization processes, more studied is needed regarding the energy efficiency and economic viability of these methods.

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