Renewable and Sustainable Energy Reviews 144 (2021) 110973

Contents lists available at ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: http://www.elsevier.com/locate/rser

Coupling biochar with anaerobic digestion in a circular economy perspective: A promising way to promote sustainable energy, environment and agriculture development in China

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ARTICLE INFO

Keywords:
Anaerobic digestion
Biogas
Biochar
Circular agriculture
Sustainable energy
Environment

ABSTRACT

Promoting the biomass industry in China would provide significant ecological, economic, and societal benefits by ameliorating soil and groundwater pollution and providing a sustainable energy source. However, although there is great potential for expanding the biomass industry in China, it currently faces large obstacles, with respect to its utilization, low economic performance, and other limitations. This study conducts a comprehensive literature review on biomass utilization methods, and establishes a new recycling model that couples the use of anaerobic digestion (AD) and its byproducts with biochar. In the model, biomass waste is used for biogas and biochar production, and biogas residues are further cracked into biochar. Biochar is also used as an additive to modify the AD process and promote methane production. In addition to its single application, biochar is further used in combination with biogas slurry or residue as a soil conditioner to repair soil and promote the growth of crops. This circulation model differs from models in which biomass alone is used in biogas or biochar production, and it combines all processes to promote the use of biomass, increase its conversion efficiency and strength the applications of such processes in the agricultural sector. Previous reports have shown that each part of the model can play a positive role in improving overall efficiency. Therefore, the application of the combined circulation model would be beneficial in biomass utilization and as such is a promising method for promoting the sustainable development of energy and agriculture in China, while protecting the environment.

1. Introduction

China is at the beginning of an energy transition that involves building a sustainable energy system for the future. In 2017, the 19th National Congress of the Communist Party of China has emphatically confirmed that China will create a market-oriented green technology innovation system and promote the development of environmentally friendly and energy-efficient industries and clean energy production industries [1]. Therefore, it is necessary for China to accelerate the transformation of the current energy system to support an ecologically advanced society.

China is a vast agricultural country that produces massive amounts of agricultural waste, including crop and forest residues and livestock manure. The random dumping of livestock and poultry manure and the local burning of straw have become increasingly prevalent, and these practices pollute the atmosphere, soil, water, local ecosystems, and living environments [2,3]. Such practices are thus inconsistent with the ability to construct an ecological civilization and they are in opposition to sustainable development and rural revitalization strategies. It is thus necessary to find alternative usages for such biomass and different methods for disposal. There is an enormous potential for biomass that it can play a vital role in easing the imbalance between energy supply and

Abbreviations: AD, anaerobic digestion; SSA, specific surface area; EC, electrical conductivity; CEC, Cation Exchange Capacity; HTC, Hydrothermal carbonization; GHG, the greenhouse gas; IET, interspecific electron transfer; VFA, Volatile Fatty Acid; OLR, organic loading rate; AEC, anion exchange capacity; SOC, soil organic carbon; Mt, million tonnes.

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https://doi.org/10.1016/j.rser.2021.110973
Received 25 April 2020; Received in revised form 22 February 2021; Accepted 13 March 2021
Available online 30 March 2021
1364-0321/

demand, protecting the ecological environment, addressing global climate change, and as an essential component in the global transition to sustainable energy sources [4].

Biochar is a solid product that is rich in carbon, and it is produced by the pyrolysis of biomass (such as crop straw, rice husk, wood chips, etc.) under anoxic and hypoxic environment. It has high porosity, a large specific surface area (SSA), high stability, small bulk density, a strong adsorption capacity and weak electrical conductivity (EC), and it has thus been widely used in the fields of carbon sequestration, in pollution adsorption and soil improvement, and in other areas [5]. Biogas is a critical technology that provides renewable energy from processing a variety of digestible biomass types. Substrates such as straw, forestry residues, animal and poultry manure, and other organic wastes can be treated under anaerobic digestion (AD) to firstly produce biogas and then biomethane after purification and upgrading [4]. In this respect, AD technology enables the transformation of energy and assists in constructing an ecological civilization. The purified biomethane has a high energy level and can be integrated into conventional fossil energy supply systems to enable clean energy production, waste control, and ecological agriculture.

There is currently considerable potential for biogas technology to be developed as a renewable energy technology that addresses energy and environmental issues. However, the biogas industry faces many challenges, including a low gas production efficiency, a short biogas tank life, a high failure rate of biogas digesters, problems with utilizing biogas residues, and it currently provides limited economic benefits [6,7]. Therefore, to improve the development of biogas and highlight its role in correctly solving energy and environmental problems, it is necessary to develop new methods and technologies that extend the industrial chain, and to explore the use of new models that can promote commercialization. Based on the recycling model shown in Fig. 1, this study couples biogas and biochar technologies to develop a model that promotes the sustainable development of energy and environment in China. In this respect, the status of biomass utilization in China and the preparation and application of biochar are briefly described. The use of biochar in the AD process and the coupling of biochar and AD in ecological and circular agriculture are then reviewed, and a new recycling model is developed. Finally, the prospects and challenges of the new model (which couples biochar and AD) are discussed. The remainder of this paper is organized as follows: the characteristics and utilization of biomass resources in China are introduced in Section 2. In Sections 3 and 4, the production and utilization of biochar and the development status, opportunities, and challenges of biogas in China are presented, respectively. In Section 5, the coupling between AD and biochar is introduced from two aspects. In Section 6, an innovative circular agriculture model combining AD and biochar is proposed, focusing on the benefits, prospects, and challenges of the model. In section 7, the paper is summarized.

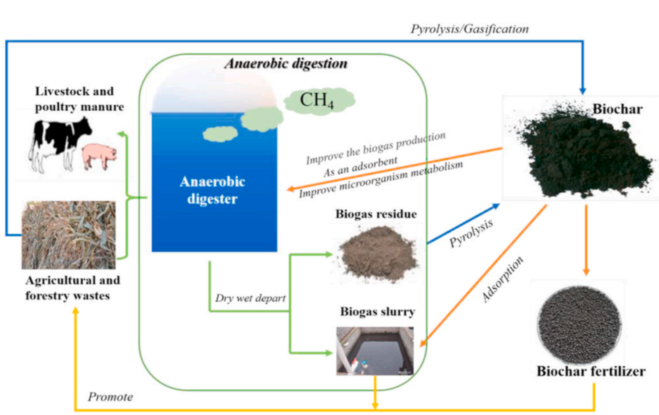

Fig. 1. A new recycling model coupling of biochar and anaerobic digestion.

2. Biomass characteristics and utilization in China

According to the statistics of the Energy Bureau, the total annual amount of biomass resources available for use as bioenergy in China is equal to 460 million tonnes (Mt) of standard coal [8]. In 2017, biomass energy was utilized at about 35 Mt of standard coal, and approximately 18 Mt of this was utilized commercially. The primary biomass resources used, as shown in Fig. 2, were straw, livestock and poultry manure, and other organic waste (which mainly included by-products from processing agricultural products (such as fruit and vegetable processing), agricultural waste, and rural organic living waste) [2,9]. According to statistics, there was a theoretical amount of 1040 Mt of straw resources in 2017, whereas the actual amount collected was only 900 Mt and the actual straw resource available for biogas production was only 180 Mt. In addition, the total theoretical output of livestock and poultry manure resources in China in 2017 was 2460 Mt; of this, approximately 1900 Mt was utilized, and 56% of this amount was used to produce methane. Furthermore, the total amounts of by-products from grain processing, waste from fruit and vegetables and from rural daily life were 210, 260, and 80 Mt, respectively, but the resources available for biogas production were only 20, 114, and 30 Mt, respectively.

China has made steady progress in utilizing biomass energy resources for energy. For example, industries have begun producing bio-natural gas and biomass molding fuel, and liquid fuel and biomass power generation industries are progressing [8]. Table 1 shows the current biomass resource utilization methods employed in China. With respect to biomass power generation in 2017, the installed biomass power generation capacity was 10.3 GW, of which 5.3 GW was generated by the direct combustion of agricultural and forestry biomass, 4.7 GW by waste incineration, and 0.3 GW from biogas, and approximately 52 GkWh was produced annually. In addition, with respect to biomass gas generation and usage, household biogas and large-scale biogas projects have been implemented. As of 2017, the theoretical annual output of biogas in China was approximately 14 Gm^3, and the annual gas output from approximately 100,000 large-scale biogas projects was approximately 5 Gm^3 [8]. Furthermore, the number of waste resources available for biogas production in China was approximately 1404 Mt, of which there were over 100 Mt of straw resources, over 1000 Mt of livestock and poultry manure, and 100 Mt of other organic waste, which could yield 73.6 Gm^3 of biogas. However, in actuality, China produced 19 Gm^3 of biogas in 2017, and replaced about 13.2 Mt of standard coal, which equals only 15% of the theoretical yield. It is thus evident that China has vast raw material resource reserves and enormous associated

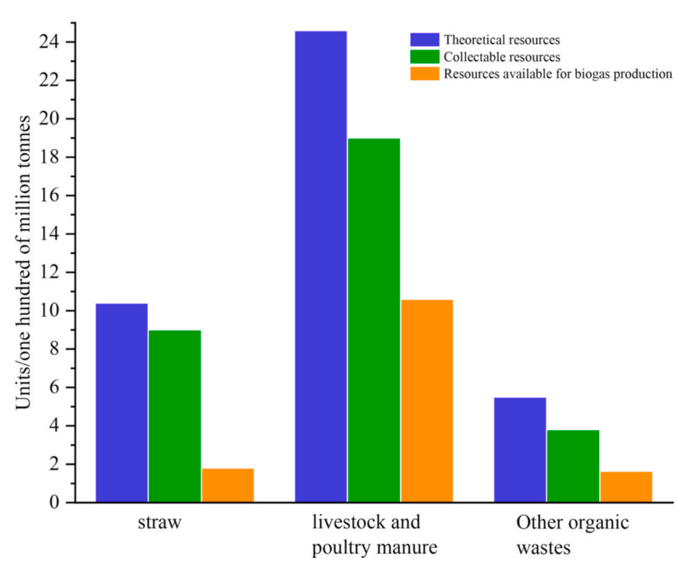

Fig. 2. Biomass resources in China in 2017(*Sources [2,9]).

Table 1
Status of biomass resources utilization in China.

Type	Scale		Annual output		Coal equivalent
	Total	Unit	Total	Unit	Mt/year
Biomass power generation	10.30	GW	52.0	G kWh	15.20
Household biogas	43.80	M families	19.0	Gm^3	13.20
large-scale biogas project	10	M plants	–		
Biofuels	8	Mt	–	–	4.00
Biomass-based fuel ethanol	–	–	2.10	Mt	1.80
Biodiesel	–	–	0.80	Mt	1.20
Total					35.40

*Sources [8]:

energy conversion potential, but the current biomass resource utilization level is very low.

3. Biochar production from biomass and associated utilization

3.1. Biochar raw materials and characteristics

Biochar is a solid product that is rich in carbon and develops under thermochemical transformations in hypoxia conditions [10]. Biochar can be produced from various raw materials, such as agricultural waste (including chicken manure, pig manure, wood chips, and straw), industrial organic waste, and urban sludge [11]. Biochar prepared from different biomass types has different physical and chemical properties in terms of its mineral elements, elemental composition, porosity, SSA, carboxylate, ash, aromatic, and aliphatic chain structures [12,13]. In addition to the raw materials employed, the properties of biochar are also influenced by its preparation temperature, heating rate, and ventilation conditions; of these, temperature is particularly important [14].

3.2. Biochar preparation technology

The main operational/control parameters for the preparation biochar are temperature, humidity, heating rate, and treatment time. The thermochemical treatment and typical conversion products employed under standard operating conditions are shown in Table 2 [12]. Four main carbonization processes are employed in the production of biochar: slow pyrolysis, rapid pyrolysis, gasification, and hydrothermal carbonization (HTC), and these are described in the following paragraphs.

Pyrolysis is the most popular method used to produce biochar [15]. The pyrolysis process can be separated into slow pyrolysis and fast pyrolysis according to the different heating rate employed. Slow pyrolysis is a thermal conversion process conducted at 400–600 °C, and this significantly improves the biochar yield [16]. On the other hand, fast pyrolysis has a high heating rate and a short residence time (less than10 s) that favors high yield of bio-oil (up to 75%) [15]. The heating rate of slow pyrolysis is typically 10–30 °C/min compared with fast pyrolysis where it is of the order 100–300 °C/min [12,15,17].

Gasification is a thermochemical process that provides a controlled amount of oxidant at high temperatures to convert biomass into gaseous mixtures [18]. The biochar yield by gasification is usually an average of about 10% (w/w) of the biomass employed [19,20]. Tay et al. (2013) studied the changes in char structure and reactivity during the gasification of Victorian brown coal at 800 °C in three different gasification atmospheres. Their results showed that the presence of steam decrease the relative ratio of small and large aromatic ring structures in char during gasification at 800 °C plays a decisive role in the evolution of char structure, and the retention rates of Mg and Ca ions were greatly improved [21].

The use of HTC is more suitable with biomass that has a high water content [22]. Biomass HTC is considered to be an economical and effective method of producing biochar, as it usually involves pressurizing the biomass in water at a low temperature of 180–250 °C [23]. The pressure, reaction temperature, water biomass ratio, and residence time are the main parameters used to determine product characteristics. Oliveira et al. (2013) determined that hydrolyzed carbon has excellent dehydration and drying properties. In addition, the water from the production process can be reused, which reduces the associated environmental impact and improves the energy efficiency [24].

3.3. Application of biochar

Biochar is currently applied in various fields, but particularly within agriculture and industry. Table 3 summarizes the advantages and disadvantages of applying it in these various fields. Biochar has the advantages being cheap and sustainable to produce, and it can be widely used to improve the retention of water and nutrients in the soil and to reduce fertilizer consumption and greenhouse gas emissions. Oxygen-containing groups on the surface of the biochar are beneficial for adsorbing pollutants in soil and water (such as organic pollutants in soil and heavy metals in water), and it can thus be used to alleviate environmental problems. Biochar has been proven to be an effective adsorbent of certain pollutants [25], but its effectiveness in organic/inorganic contaminant remediation remains uncertain.

Biochar also plays an important role in AD and composting. Pan et al. (2019) studied the effects of nine biochar species on AD in chicken manure. The results showed that the production of methane from AD increased when using all types of biochar: methane was produced from wheat straw, waste fruit trees and chicken manure, respectively, under pyrolysis conditions of 350, 450, and 550 °C [26]. In addition, it is known that a higher nitrogen retention rate and a better heavy metal stability can be achieved by adding biochar to composite materials [27]. Furthermore, organic matter can be effectively degraded by biochar produced during the slow pyrolysis of biomass [28,29], and this reduces total nitrogen loss and greenhouse gas (GHG) emissions during the composting process [30]. Biochar is also considered to be an ideal filler in nitrogen-rich compost processes, and it can thus reduce (or even prevent) emissions from the natural decomposition of agricultural waste within the soil. Dias et al. (2010) used biochar as a filler in poultry manure composting and found an obvious improvement in the degradation rate (as high as 70%). The addition of biochar also reduces odor emissions and the loss of nitrogen [31]. In this respect, Liu et al. (2017) showed that adding biochar to poultry manure during composting

Table 2
Typical parameters for biochar formation processes.

Treatment	Temperature [°C]	Residence Time	Heating Rate [°C/min]	Typical product yield [%]		
				Solid	Liquid	Gases
Slow Pyrolysis	300–650	5 min–12 h	10–30	25–35	20–30	25–35
Gasification	600–900	10–20 s	50–100	<10	<5	>85
Hydrothermal carbonization	180–260	5 min–12 h	5–10	45–70	5–25	2–5

*Sources [12]:

Table 3
The advantages and disadvantages of biochar application in different fields.

Application	Purpose	Advantages	Disadvantages	Reference
Adsorbents	Adsorption of organic contaminants and heavy metals present in soil and water	Low cost, abundant and sustainable resource, and oxygenated groups on biochar surface facilitate adsorption	Effectiveness of organic/inorganic contaminants remediation is still uncertain, and persistence of heavy metals	[25,33,34]
Precursor of catalysts	Catalyst support and carbon-based materials act as direct catalysts	Easy to recycle supported metal, large surface area abundant functional groups, co-catalyst, low cost	Relative low efficiency and low abrasive resistance compared with commercial catalyst	[35,36]
Soil amendment	Carbon sequestration, soil quality improvement	Low cost, sustainable resource, retain water and nutrient, reduce fertilizer consumption, reduce greenhouse gas emission and nutrient losses	Possible heavy metal and PAHs contaminant	[37–39]
Additives for anaerobic digestion/composting	Enhancing carbon mineralization, and changing the microbial community structure	Large specific surface area, porosity, and a large number of functional groups, reducing the emission of GHG and total nitrogen loss, high nitrogen retention and good heavy metals stabilization	The risk of introducing heavy metals and organic contaminants into soil	[40,41]
Electrochemical energy storage	Used as electrodes material or template	Low cost, renewable feedstock, high surface areas, rich porosity	Possible low performance	[15]

promoted the degradation rate and reduced the emission of various gases. They considered that the optimal addition amount was 10%, as the OM degradation rate was the highest, ammonia and GHG emissions were at their lowest, and the ideal cost was obtained [32].

4. Biogas development status, opportunities, and challenges in China

4.1. Biogas development status

The biogas industry in China has developed via the co-development of household biogas, a centralized biogas supply for households, and large-scale biogas. Biogas is used in various forms, such as a gas supply for rural households, in captive biogas power generation, in biogas cogeneration and grid-connected power generation, and it is purified and used as pipeline gas and automobile gas [42].

Rural biogas entered a new stage of rapid development from 2003 to 2015 under promotion by the central government. According to data released by *the National Development and Reform Commission of the People's Republic of China (NDRC)*, by the end of 2015, 41.9 million household biogas digesters had been built, which enabled 200 million people to benefit from using biogas. Of these, a total of 110,975 biogas digesters were built, including 103,898 small and medium-sized biogas digesters, 6737 large biogas digesters, 34 very large biogas digesters, and 306 industrial biogas digesters. Positive results were achieved by the integrated construction of natural gas pipe networks in cities with a large number of biogas digesters (within areas that have abundant and moderate natural gas resources). Furthermore, the number of rural service outlets reached 110,000, providing supplies to more than 74% of household methane users [43]. By 2015, the number of biogas power plants had increased to 32,070, which was 43% higher than in 2009. According to the *National* 13th *Five-Year Plan for Rural Biogas Development*, by 2020, the handling capacity of straw, livestock manure, and poultry manure would have been increased by 8.64 Mt and 71.83 Mt, respectively, by 2020, and 172 normal biogas plants and 3150 large biogas plants would have been built. The plan also includes the further development of household biogas digesters and small biogas digesters, to increase the production capacity of biogas to 22.77×10^9 m^3 and increase the production of biogas to 4.9×10^9 m^3 [2]. The above figures show that small biogas plants and rural household-based digesters represent the largest proportion of biogas facilities, and they are mainly used to supply gas in rural areas and are designed to have a non-profit operation mode [4]. However, the low efficiency of biogas projects, the low associated income, and the high investment into biogas has directly led to the failure of many biogas projects [4,6]. It is thus evident that China's biogas industry could be further improved.

4.2. Biogas development opportunities

In recent years, recognition that the global warming caused by excessive emissions of GHG is one of the most serious ecological challenges facing the planet, which has been endangering human health [44]. China's biogas industry has excellent development opportunities, as biogas projects can be focused on obtaining electricity and biological natural gas, which would help to alleviate environmental problems and ameliorate global climate changes. In this respect, if half of the straw and human and animal manure in China is used for biogas fermentation, then this can be used to produce 65×10^9 m^3 of biogas each year at a heat energy equivalent of 100 Mt of standard coal [44]. In addition, as air pollution and human health problems are promoting the development of biogas projects (air pollution caused about 911,000 deaths in China in 2015, and 240,000 of these were related to coal pollution [44–46]), it is thus critical to develop clean energy (such as biogas) to replace coal. Furthermore, the development of the biogas industry has been promoted by national policies and guidelines in China. For example, in 2016, the National Energy Administration issued the "13th *Five-Year Plan for Biomass Energy Development*", which determines that large-scale utilization and commercialization of biomass energy should be achieved by 2020 [8]. In 2017, the Ministry of Agriculture and Rural Affairs formulated the "13th *Five-Year Plan for National Rural Biogas Development,*" which integrated biogas into the national energy and ecological strategy [2], and in 2019, the National Energy Administration released "*Guidelines on Promoting the Industrialization of Biomethane*," which stated that China would be producing biogas on a greater scale than anywhere in the world by 2030, at an annual output of more than 30×10^9 m^3 [46]. As the biogas industry complies with the requirements of ecological civilization construction and can use agricultural waste to generate clean energy, it can effectively help alleviate climate change and environmental pollution. It is also important to note that the use of biogas will develop a circular economy: the combination of planting, breeding, and the lives of farmers are intrinsically related to biogas projects; as such a typical agricultural circular economic model can be realized in rural China [47].

4.3. Biogas development challenges

The current demand for environmental protection, the supporting associated policies, and improvements in technology have created unprecedented opportunities for the development of biogas. However, the development of biogas continues to face challenges that cannot be ignored, and these are outlined as follows:

First, changes in China's rural population (where rapid urbanization has led to a sharp decline in rural labor) and the energy structure have hindered the development of rural biogas. With economic growth and the expansion of the energy market, rural access to clean and efficient energy has become increasingly easy; therefore, many rural families are reluctant to engage in biogas production [6]. Second, China's biogas policy has limitations. China's existing subsidies for biogas projects mainly focus on the construction process of biogas projects, and subsidies to enable the daily operation of biogas projects are lacking, which results in biogas projects with low operational efficiencies (or they are terminated) [48]. Third, biogas engineering technology is not sufficiently mature. After years of development, biogas engineering continues to experience technical bottlenecks in China, for example with respect to the design of large digesters, heat exchange processes, and anaerobic co-digestion technology for multiple organic substances [6], all of which considerably hinder the development of biogas. Fourth, commercialization is lacking. As the industrial chain relating to most biogas projects is imperfect and the operation of projects is non-profit making, the sustainable development of the biogas industry cannot be guaranteed [4]. The lack of commercialization results in a lack of effective treatment for biogas slurry and residue produced by biogas projects. Due to imperfect supervision mechanisms within the environmental protection department, biogas slurry and residues are directly discharged, which causes secondary environmental pollution. Zhang et al. (2007) found that the amount of as in biogas slurry and slag collected in the Chongqing area exceeded Chinese standard by as much as 60%, and direct discharge into rivers or soil causes severe heavy metal pollution [49]. Nitrogen and phosphorus-rich biogas slurry is directly released into rivers and lakes, which causes eutrophication of water bodies. In addition, large amounts of nitrates and phosphorus are directly discharged onto soil, and they eventually reach both ground and surface water bodies [50]. Therefore, avoiding secondary pollution from biogas sludge is a considerable challenge that needs to be addressed to enable biogas industry development.

5. Enhancing bioenergy production via coupling AD with biochar

Biogas development faces multiple challenges, and numerous studies have been conducted to solve these. It has been found that biochar coupled with AD is a promising method that can address some of the associated issues relating to development. This section explains how this method can be used to meet such challenges in two respects (see Fig. 3), including the application of biochar in AD and the application of biochar produced by AD residues in AD.

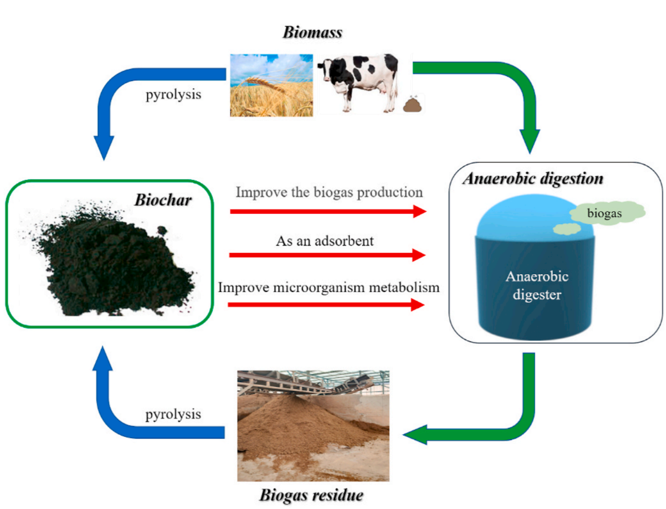

Fig. 3. A small cycle of biochar production and AD process.

5.1. Application of biochar in AD

Although AD is a mature technology used in organic waste management, associated problems have occasionally surfaced. In recent years, studies on biochar as an additive in AD have shown that biochar can be used to optimize operating conditions and modify the instability inherent in the AD process, including improving the biogas production and methane content, increasing the buffer ability, reducing amino acid inhibition, and improving microbial enrichment and interspecific electron transfer (IET) [51,52].

Table 4 summarizes the application of biochar in the AD process, which is crucial to improve biogas productivity and methane content, process stability, and slurry quality. Kumar et al. (1987) and Shen et al. (2020) treated cow manure at a medium temperature (35 °C) using an AD system in which biochar generated by powdered charcoal and rice husk at 600–900 °C was added, and the results showed increased biogas production of 17.4% and 9.86%, respectively [52,53]. In addition, the porous structure of biochar provides an environment that is suitable for microbial growth and inhabitation, and this is conducive to improving

Table 4
Application of biochar in AD.

Type of biochar	Substrate in AD	Application	References
Corn stover	Sewage sludge	Increased the methane yield; Changed the methanogenic community structure	[62]
Corn stover	Sewage sludge	Increased methane yield; Enhanced CO_2 removal; Improved process stability	[54]
Fruit woods	Sewage sludge	Lag phase was shortened; Increased methane yield; Overcome ammonia inhibition; Enhanced degradation rate of VFA	[55]
Sawdust	Food waste; sewage sludge	Mitigated VFAs accumulation; Improved microbial activities; Enhanced methane production	[58]
Fruit woods	Glucose	Lag phase was shortened; Enhanced the production and degradation of intermediate acids; Promoted IET	[59]
Fruit woods	Chicken manure	Increased methane yield; Alleviated ammonia inhibition; Remove hydrogen sulfide(H_2S)	[60]
Municipal solid green waste	Organic household waste; sewage sludge	Immobilized methanogenic archaea	[61]
Fruit wood	Food waste	Lag phase was shortened; maximum CH_4 production rate increased; enhanced degradation rate of dissolved organics; enhanced degradation rate of VFA; Immobilization of micro-organism; biofilm growth;	[50]
Coconut shell	Citrus peel waste.	Lag phase was shortened; slight increase in CH_4 production; increased colonies of methanogens; increased removal efficiency;	[63]
Sawdust	Food waste	Lag phase was shortened; increased methane production rate; high OLR with no immediate inhibition promoted; micro-organism adaptation to high VFA accumulation facilitated through pH adjustment with biochar; Promoted IET	[64]
Forest wood residue	Organic fraction of municipal solid waste	Increased CH_4 yield	[65]

the quality of biogas [54].

The inhibition of ammonia and the accumulation of acid during AD can be overcome by adding biochar. Due to local over-concentrations of a single substrate in AD, the nutrient balance between microorganisms and the raw material is damaged, which causes ammonia or acid formation to be inhibited. This phenomenon can inhibit the growth, reproduction, and metabolism of functional bacteria [55]. The addition of biochar enhances the tolerance of high ammonia concentrations in the AD system. In this respect, Giwa et al. (2019) found that the addition of biochar reduces the concentration of ammonia nitrogen in anaerobic digestion; in particular, the most apparent effect is seen when the ammonia nitrogen concentration is above 2450 mg/L, as this is conducive to the long-term biodegradation of volatile fatty acids (VFAs) [56]. Lu et al. (2016) also proved that adding biochar could effectively alleviate ammonia inhibition during AD when glucose was used as the substrate [55]. The rapid accumulation of VFAs in the AD process leads to acid inhibition, which results in a low pH value and the inhibition of methanogen activity, especially under a high organic loading rate (OLR) [57]. Wang et al. (2019) and Luo et al. (2015) also showed that biochar not only increased biogas production but also delayed the VFA accumulation time and degradation of intermediate acids [58,59].

Adding biochar can also improve microbial metabolism and IET in AD. One study showed that porous biochar enhanced the potential of microbial reproduction during AD and it was proven to be an effective method of enhancing microbial metabolism [55]. Biochar can promote the potential of anaerobic microbial co-metabolism through IET, and it can thus be considered a carrier material that can be used for microbial growth, reproduction, and metabolism [59–61].

5.2. *AD residues used for bioenergy recovery and biochar production through the pyrolysis process*

Biochar can promote AD, and AD residues can then also be used to prepare biochar by pyrolysis or hydrothermal carbonization. Biochar produced by biogas residue pyrolysis undergoes four stages: water volatilization, hemicellulose decomposition, cellulose decomposition, and lignin decomposition, water volatilization at temperature <220 °C; hemicellulose decomposition at 220–315 °C; cellulose decomposition at 315–400 °C; and lignin decomposition at temperatures >400 °C [66,67]. The HTC preparation of biochar can be divided into five hydrolysis processes: dehydration, decarboxylation, polymerization, and aromatization [24]. Yao et al. (2011) and Inyang et al. (2010) used biogas residue obtained from the AD of bagasse and beet residue to prepare biochar at pyrolysis conditions of 400 °C. The results showed that biochar made from biogas residue was better than that made directly from raw materials with respect to the pH, Cation Exchange Capacity (CEC), hydrophobicity, carbon yield, and adsorption performance [68,69].

Biochar produced by biogas residue pyrolysis has excellent physical and chemical properties and good application prospects. Bogusz et al. (2017) found that biochar produced by high-temperature pyrolysis of biogas residue had a higher adsorption capacity and ability to remove Cd and Ni ions from wastewater [70]. Stefaniuk et al. (2015) found the biochar prepared by biogas slurry pyrolysis had a high pH value, and it could thus be used to improve acid soils and reduce the use of lime [71]. Bruun et al. (2011) studied co-administered biochar and anaerobic digested slurry and found that it reduced soil N_2O emissions, enhanced soil microbial activity, and promoted soil nitrogen fixation [72]. Inyang et al. (2010) found that biochar produced from bagasse digestion residue had higher SSA, pH, and CEC values and a higher anion exchange capacity (AEC), hydrophobicity, and negative surface charge, which could be used to improve the soil quality and adsorb water contaminants [69].

6. Introducing innovative circular agricultural model that couples AD with biochar

6.1. *Traditional circular agriculture in China with a focus on AD*

China has developed different models that comprehensively utilize biogas (Fig. 4), including the “three in one” model in the south, the “four in one” model in the north, and the “five in one” model in arid regions of the northwest [73–75]. Although there are differences between these circulation models, they are generally interlinked in the following ways: the AD of agricultural waste produces biogas, biogas slurry, and biogas residue, and biogas is then used for lighting and cooking. Biogas slurry and biogas residue are subsequently used as biogas fertilizer, and the product is thus returned to the farmland, as shown in Fig. 5. In this way, agricultural waste is recycled. However, although these traditional ecological agricultural models realize the recycling of agricultural waste, increase the income of farmers, and improve the rural ecological environment, they also have certain limitations. For example, the conventional AD is unstable and inefficient and does not make full use of agricultural and domestic waste [76]. In addition, biogas fertilizer is directly applied to the soil, which has a weak effect on soil improvement and causes heavy metal pollution [48]. Therefore, to effectively make use of agricultural and domestic waste, it is necessary to optimize and improve the biogas utilization model.

6.2. *Innovative circular agriculture model based on a combination of biochar and AD*

6.2.1. *Description of innovative circular agriculture model*

Biogas engineering is a link in the chain of the circular agriculture system, as it transfers environmental waste into energy and fertilizer and successfully combines agricultural production and waste treatment. Compared with the traditional circular agriculture model, the innovative circular agriculture model can further improve the utilization rate of resources. Not only can agricultural waste be subjected to AD to obtain biogas, residue, and slurry, but it can also be directly used to prepare biochar. The biogas residue is then further cracked into biochar, which is used as an additive to modify the AD process and promote methane production. The biochar is subsequently used as an adsorbent that effectively enriches the nutrient components in the biogas slurry, or it is applied to crops together with the biogas residue. Finally, the biochar is used as a soil conditioner to repair soil and improve the growth environment of crops. In this process, methane gas is produced, and it can be used to generate power and heat. In this respect, the innovative circular agriculture model promotes the development of the circular agriculture and provides excellent technical strategies for treating biomass waste, biogas residue, and slurry (see Fig. 6).

Compared with the traditional recycling model in which biomass is directly applied to AD, the innovative model divides biomass utilization into two levels. First, the preparation of biochar and its use in AD; biochar added to AD can improve biogas production, as shown in a large number of studies [52,53,77]. Second, biogas residue is used to make biochar, and this is then returned to AD. A number of previous studies have shown that biochar prepared from biogas residue is superior to that prepared directly from biomass waste, with respect to improvements in pH, cation exchange capacity, hydrophobicity, the carbon yield, and its adsorption performance [68,69,78–80].

The innovation model not only reduces the cost of waste management, improves the utilization rate of biomass, and makes the production of bioenergy more sustainable and environmentally friendly, but it also improves the lives of farmers and provides certain economic benefits.

6.2.2. *Benefits of applying biochar in agriculture*

Biochar is used in anaerobic fermentation, but it can also be directly applied in agricultural practices in the following ways:

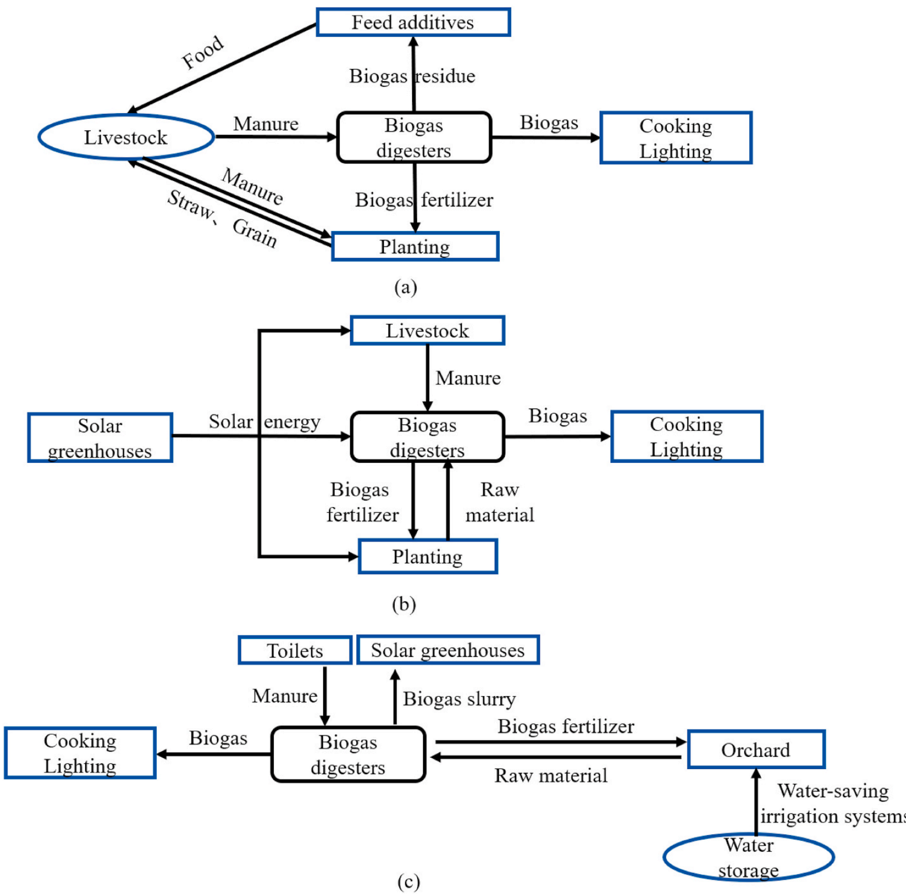

Fig. 4. Three Modes of Comprehensive Utilization of Rural Biogas in China:(a) "There in one" model; (b) "Four in one" model; (c) "Five in one" model.

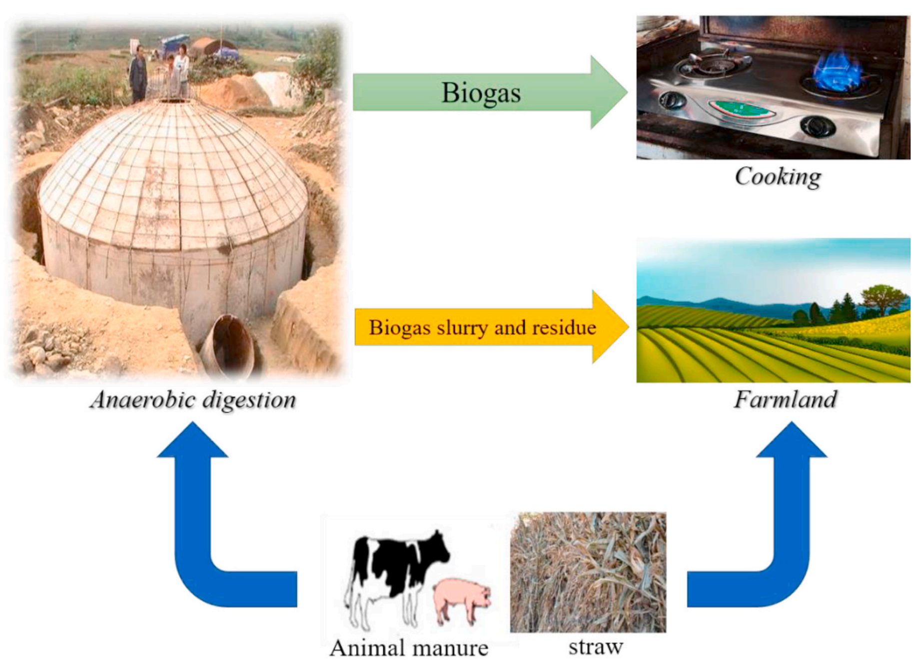

Fig. 5. Comprehensive rural biogas utilization mode in China.

(1) As a soil conditioner: biochar contains minerals and is alkaline when dissolved in water; it can thus be applied to modify acidic soil [81]. As biochar is rich in micropores and surface functional groups and has a large SSA, it can also improve soil water retention, the fertility permeability, and the cation exchange capacity, and reduce toxicity [82]. Biochar is highly stable, and it can be used to realize carbon sequestration and maintain the balance of soil ecosystems. Importantly, it can also be used to

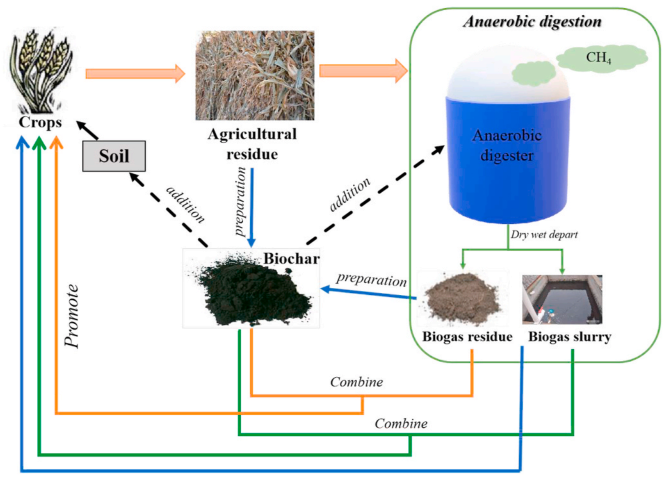

Fig. 6. Innovative circular agriculture mode.

regulate the soil microbial community structure and soil enzyme activity, and it enhances soil activity [83], which make it an excellent soil conditioner.

(2) As compost additive: biochar has high stability and porosity; therefore, adding biochar to compost material provides a higher nitrogen retention rate and superior heavy metal stability. It also speedily reduces the volume of the compost material and diversifies the structure of the microbial community [40]. All these characteristics indicate that adding biochar has a positive effect on compost.

(3) Improving the growth environment of crops: carbon is a basic element required for plant growth and development, and as a carbon-rich material, biochar can be appropriately applied to the soil as a carbon fertilizer. It can be used to promote the growth and development of crops by prolonging the fertilizer supply time, changing the structure and quantity of flora in the soil, and improving the disease resistance of crops [83].

(4) Improving environmental quality: the large amounts of factory sewage discharged causes soil pollution and heavy metal contents that exceed standards. The surface of biochar is rich in micropores and it has a large specific surface area: it thus also has a strong adsorption capacity and can adsorb pollutants in wastewater but heavy metals in contaminated soil [84].

6.2.3. Benefits of applying biochar combined with biogas residue or slurry in agriculture

6.2.3.1. Biochar + biogas residue (BBR). Greenberg et al. (2019) studied the effects of using biochar and biogas residue (BBR) and mineral fertilizers on the biomass of crops, the organic carbon components, and the mineralization of soil. The results showed that mixing 40 Mg of biochar ha^{-1} increased the C mineralization of soil by 16% and provided a SOC content that was 3.8 times higher than that of non-biochar treatment. Moreover, the soil organic carbon content of the biogas residue combined with 40 Mg biochar ha^{-1} was higher than that of mineral fertilizer combined with 40 Mg biochar ha^{-1}. These results indicate that the addition of BBR is highly effective for improving soil organic carbon [85]. Roberto et al. (2018) studied the short-term (100-day) effects of applying biogas residue, a single biochar application, and BBR to soil. The results showed that both the single application and BBR affected the soil pH to varying degrees. Following cultivation, it was found that BBR significantly reduced soil ph. When the highest doses of BBR were applied, the largest decrease in the pH value of the soil was obtained (pH = 6.5). The total organic carbon content in the soil was the highest (82.8% of the initial amount), which indicates that BBR not only effectively relieved the alkaline environment of the soil, but also increased the organic carbon within it. However, it was also found that biochar reduced the content of soluble organic compounds when applied with biogas residue, which thus limited the number and activity of microorganisms [86]. It is thus considered that it is necessary to conduct further studies in this respect, as only a limited amount of research has been conducted on the effects of applying BBR in agriculture to date.

6.2.3.2. Biochar + biogas slurry (BBS). Different effects on the growth of soil and crops have been found when adding biochar, biogas slurry, and biochar and biogas slurry (BBS). BBS can be applied in two ways: the simultaneous application of biochar and biogas slurry (BBS-A) and via the absorption of biogas slurry by biochar prior to application (BBS–B). Ge et al. (2016) and Wang et al. (2018) studied the effects of applying biochar, biogas slurry, and BBS-A on soil nutrients and active organic carbon, and their results showed that adding biochar significantly reduced soil nutrient loss, and BBS-A further improved soil active organic carbon content and soil fertility [87,88]. In addition, Zhang et al. (2015) studied the effects of applying biochar, biogas slurry, and BBS-A on the yield and quality of apple fruit. The results showed that the application of BBS-A was superior to using the other processes; in comparison to the single applications of biochar and biogas slurry, the following benefits were obtained: an average increased fruit yield of 58.5%, vitamin C content of 47.8%, and soluble sugar content of 17.4% [89]. The BBS-B method was applied in small rape planting, which improved crop yields by 10% compared to the direct use of biogas slurry [90]. Recent studies related to the effects of biochar, biogas slurry, and BBS on crop yield are summarized in Appendix 1.

6.2.4. Improving environmental benefits

In the traditional recycling agriculture model, the biogas residue produced by AD is usually directly applied to the field. Although it can promote the growth of crops, it may also cause secondary pollution via the excessive heavy metal content in the biogas residue. The preparation of biochar from biogas residue not only reduces the harmful substances in the biogas residue but it also improves the performance of the biochar compared to that of other biomass wastes [68,69,80]. Biogas slurry is an excellent organic fertilizer; however, as it is a liquid fertilizer, it easily permeates the soil after application. Therefore, organic substances in the biogas slurry are not well absorbed and groundwater becomes polluted [105–107]. Li et al. and Zheng et al. analyzed the adsorption performance of biochar on biogas slurry under different preparation conditions and found that the biogas residue had a better adsorption effect when KOH was used as the activator [108,109]. In this respect, not only was pollution effectively reduced from the direct discharge of biogas slurry, but nutrients in the slurry were better absorbed. These analyses show that the innovative circular agriculture model could be employed to solve the low utilization efficiency and secondary pollution problems associated with biogas residue and slurry.

6.2.5. Prospects and challenges

In recent years, an increasing amount of research has focused on using a combination of biochar and AD to solve the problems associated with the AD process. The advantages of applying biochar in the AD process are reviewed in the previous section. However, there are many existing challenges that hinder the development of this technology. The selection of raw materials is an initial significant factor affecting the preparation of biochar and the corresponding production of methane [110]***. Numerous types and vast amounts of biomass waste are produced annually*** [67]***, and it is necessary to define the types of biochar that match well with the different AD substrates. Furthermore, to ensure that biochar is better coupled with AD, it is necessary to further clarify the role and mechanism of biochar in the AD process.Another challenge is to determine how to combine AD by-products (residues and slurry) with biochar and apply them in agricultural production. Although previous studies have shown that the combined application of***

biochar with biogas residues or slurry can significantly improve crop growth, limited associated research has been conducted. In the actual application process, the two are simply applied at the same time, or they are mixed prior to application to achieve better functions, and this has not yet been adequately studied. Furthermore, it is necessary to conduct an economic evaluation of their coupling application. In summary, these challenges need to be addressed to enable the effective application of the innovative circular agriculture model proposed in this study.

7. Conclusions

A comprehensive literature review on the characteristics and utilization methods of biomass resources has been done, and establishes a new recycling model that couples the use of AD and its byproducts with biochar. The main obtained conclusions are the following:

(i) The development of biochar technology and AD technology is of great significance, which can solve the problem that China has abundant biomass resources but low utilization rate. (ii) Based on the combination of biochar and AD to form a small cycle that the biogas residue produced during AD can be made into biochar via pyrolysis and returned to the AD system, an innovative circular agriculture model has been established combined with biochar, AD and agriculture which can effectively use biomass resources, promote crop growth, increase crop yields, and increase economic benefits. (iii) The innovative circular agriculture model uses the coupling of AD and biochar at its core is a promising method that can provide new ideas for the sustainable development of energy, environment and agriculture in China.

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.

Acknowledgement

This work was financially supported by National Natural Science Foundation of China (41871205, 51508467), Scientific research plan projects of Shaanxi province (2020SF-356), Scientific research plan projects of Xi An (20NYYF0008), Youth Talent Cultivation Program Funding of Northwest A&F University, Special Research for Rural Vitalization Strategy (Z1090219062), Shaanxi Engineering Research Center of Circular Agriculture (2019HBGC-13).

Appendix 1

Table 5
Summarize on the effects of biochar, biogas slurry and their combination on crop yields

	Treatment	Control	Crop	Increased yield (%)	Reference
Biochar alone	Wheat straw carbon 30 g/kg soil	Conventional fertilization	Rice	34.20	[91]
	Rice straw carbon 30 g/kg soil		Rice	22.20	[91]
	Corn straw carbon 30 g/kg soil		Rice	55.20	[91]
	Corn straw charcoal 10 g/kg soil, 20 g/kg soil, 40 g/kg soil		Rice	11.7–33.21	[45]
	Peanut shell biochar		Wheat	8.46	[92]
	Peanut shell charcoal		Corn	18	[92]
	Biochar from wheat straw 1000 kg/hm^2, 5000 kg/hm^2, 10000 kg/hm^2		Corn	7.1–8.8	[93]
	Corn straw charcoal 10 t/hm^2, 20 t/hm^2,40 t/hm^2, 80 t/hm^2		Corn	1.16–42.13	[94]
	Corn straw charcoal 50 g/kg soil, 100 g/kg soil, 150 g/kg soil		Rape	37.3–64	[95]
	Corncob biochar 1500 kg/hm^2	Recycled straw + conventional fertilization	Peanut	41.7–58.3	[96]
Biogas slurry alone	The volume ratios of biogas slurry and water are respectively 1:4,1:6 and1:8	water	Tomato	13.22	[97]
	Duck dung biogas liquid, Pig manure and biogas fluid, Cow dung biogas slurry	Conventional fertilization	Tomato	55.9–232.8	[98]
	Synthetic fertilizer + cow manure biological fertilizer, synthetic fertilizer + poultry biological fertilizer	Synthetic fertilizer	Tomato	11–36	[99]
	Treatment	**Control**	**Crop**	**Increased yield (%)**	**Reference**
Biogas slurry alone	Pig manure and biogas slurry	No fertilization	Peanut	33.15–48.29	[100]
		Conventional fertilization	Peanut	10.24–22.77	[100]
			Peanut	10.8–19.4	[101]
			Rice	16.7–41.4	[102]
			Rice	8.90	[103]
			Wheat	18.40	[104]
			Wheat	15.7	[103]
			Wheat	23.47	[92]
			Corn	15.49	[92]
			Rape	16.6–60.7	[102]
BBS (applied simultaneously or biochar adsorbing biogas slurry)	Straw biochar 0.5 kg + biogas slurry 0.5 kg (applied simultaneously)	Conventional fertilization	Apple	58.50	[89]
	Straw biochar 50 g + biogas slurry (biochar adsorbing biogas slurry)		Rape	10	[90]

Funding

This work was financially supported by National Natural Science Foundation of China (41871205, 51508467), Scientific research plan projects of Shaanxi province (2020SF-356), Scientific research plan projects of Xi An (20NYYF0008), Youth Talent Cultivation Program Funding of Northwest A&F University, Special Research for Rural Vitalization Strategy (Z1090219062), Shaanxi Engineering Research Center of Circular Agriculture

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