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Upgrading argo-pellets by torrefaction and co-pelletization process using food waste as a pellet binder

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ABSTRACT:

This work presents a study on the properties of upgraded argo-pellets produced by torrefaction and co-pelletization process, where starch-rich food waste was used as a pellet binder. The impact of blending coal cutting waste (CCW), torrefaction pre-treatment and the use of starch binder on the mechanical properties of argo-pellets were examined. The resulting physical properties of argo-pellets were significantly improved. Thermal decomposition of corn straw (CS) can be divided into three stages. Co-pelleting with a small amount of CCW promote the combustion of CS, showing an improved thermodynamic characteristic of torrefied corn straw (TCS). Torrefaction reduces the difference of heat distribution, improving the adaptability of pellet fuel in operation. Both co-pelletization and torrefaction processes lead to reduced concentrations of NO_x and SO_2, increased calorific value of fuel and elevated CO_2 emission per unit mass, which are conducive to pollutants reduction and thermodynamic qualities of argo-pellets. Moreover, life cycle assessment (LCA) analysis indicated that more energy input was required for co-pelletization and torrefaction pre-treatment, consequently leading to an enhanced energy return ratio.

KEYWORDS: Argo-pellets, torrefaction, co-pelletization, starch binder, life cycle analysis, energy

1. INTRODUCTION

China has become the world's largest emitter of carbon, nitrogen and sulphur oxides due to its over-reliance on coal [1], which calls for the development of alternative energy conversion from biomass. This is further promoted by a series of policies made recently in China [2]. The wastes in agricultural production and wood industry are the main source of biomass [3]; however, direct utilization of biomass feedstock is often not practical, this is due to its low energy density, unstable combustion and difficulties in storage and transportation while biomass pellet presents a much higher bulk density, making it suitable for long distance transportation and storage. Together with its high calorific value in combustion [4, 5] and sustainable characteristics of biomass, application of biomass pellet is at the center of both scientific research and industrial practice.

To date, the majority of biomass pellets are produced from woody biomass due to its high calorific value and low ash content [6]. However, the production of wood-based raw material cannot meet the growing market in demand. It is thus necessary to search for alternative feedstock for biomass pelletization. For non-woody biomass, agricultural waste could be of significant use due to its wide availability and low price. This is especially the case in China, as approximately 0.24 billion tons of corn stover (CS) are produced yearly, accounting for 32% of the total crop residues [7, 8]. Agro-pellets made from corn stover usually have a lower calorific value, a lower density and a higher ash content than that of woody pellets, which can be categorised as a low-grade pellet fuel. There is thus a need to improve the quality of agro-pellets, which can be potentially achieved by pelleting various blends and torrefaction [9, 10]. Previous studies have shown that component complementarity and mechanical interlocking between particles were significantly enhanced through biomass blending [11], consequently leading to a better pelleting performance. For example, Djatkov et al. [12] showed that blending with spruce wood improves the mechanical properties of corn stover pellets. Similar results were reported by Stasiak et al. [3] and Harun et al. [13]. The calorific capacity of straw pellet was found to increase with the addition of higher calorific value pine sawdust. Blending can also reduce compaction energy and thus consumes less energy in the pressing process [13, 14]. In addition to

63 wood/agricultural residues blending, the mixing of various agricultural wastes [15, 16],
64 co-pelleting of agro-wastes with other residues, such as sewage sludge, pyrolysis oil
65 and hydrolysis lignin, has also been reported [17-20]. **Table 1** lists a short summary of
66 biomass blending reported in the literature.
67
68 **Table 1.** Blending biomass pellets: literature reviews

Biomass feedstocks	Research objectives	Experimental conclusions	Ref.
Spruce wood and corn stover	The influences on the mechanical-physical properties of corn stover pellets	Blending with wood improved the mechanical properties of corn stover pellets	[12]
Pine sawdust and wheat/ rapeseed straws	Mechanical and combustion properties of pellets made of pine sawdust mixed with straws	Pellet density、strength and calorific capacity increased with the addition of pine sawdust	[3]
Pine sawdust, spruce, timothy hay and switchgrass	The energy requirement for the pelletizing of agricultural biomass	Blending helped to improve the quality of pellets and lower the compaction energy	[13]
Rice husk and wheat straw	Physicochemical and energetic characterization of pellets produced with rice husks and wheat straws	Biomass blends improve the quality and combustion characteristics of the pellets	[16]
Pepper, perilla, rice chaff, and spent coffee	The possibility of pellets made from agricultural by-product	Perilla coffee pellets (PRCP) were determined as A-grade owing to higher durability.	[15]
Sewage sludge and biomass (Chinese fir, camphor and rice straw)	Energy consumption and properties of sludge mixed pellets and pure biomass pellets	Biomass-sludge mixed pellets reduced energy input, increased hardness and improved the combustion characteristics	[20]

69
70 Compared with wood and agricultural wastes and other residues, coal fuel has a
71 higher calorific value. Heat value and combustion characteristics of argo-pellets can
72 thus be improved by co-pelleting with coal or coal waste. As the main by-product of
73 coal mining, coal cutting waste (CCW) is characterised with a high-moisture content

and small particle sizes, presenting serious challenges in collection, storage and transportation [21, 22]. For example, in open areas, stored coal cutting waste may pollute ground water and transfer to far places by natural means. Large amounts of unburnt fine particles are released to the atmosphere in the conventional combustion systems, causing environmental problems if they are not controlled properly. In this regard, pelletizing or co-pelletizing allows for an effective use of coal cutting waste [23-25]. Atay and Ekinci [26] characterised the pellets made from lignite coal powder and pine bark. Experimental results showed that the added coal increased combustion temperature and decreased the maximum weight loss rate, leading to an improved combustion efficiency. Janewicz et al. [27] produced pellets from biomass-lignite blends and found that blending pellets present good combustion characteristics but poor water resistance. Both compressive strength and water resistance decreased with the increase of biomass proportion [28]. However, contrary conclusions have been drawn by Yaman et al. [29] and Ozyuguran et al. [30], where the mechanical properties of lignite briquettes were improved by adding biomass while excessive lignite is detrimental to briquetting by decreasing impact resistance and durability of pellets [9, 26]. It can thus be concluded that blending with coal or coal waste can improve the calorific value of biomass pellets but may not contribute to the mechanical properties of briquettes. For different biomass and coal materials, it is necessary to provide a guideline on the selection of blending proportion during the co-pelleting process. In this study, torrefied argo-biomass and fine coal particles (coal cutting waste) are selected, with their physical and chemical properties partially changed.

In addition to blending with coal cutting waste, torrefaction pre-treatment can improve calorific value and combustion characteristics of argo-pellets [31, 32]. During the torrefaction process, biomass feedstock is heated to the temperature of 200～300 °C with low oxygen concentration [33]. Dehydration breaks large numbers of hydroxyl bonds and reduces hydrogen bonding between biomass and water molecules, consequently improving the hydrophobicity of biomass [34]. With the removal of light volatile fraction that contains most of the oxygen in biomass, heating value of the remaining biomass is gradually increased. Decomposition of hemicellulose destroys the fiber structure of dried biomass, which benefits biomass crushing and improves grinding characteristics [35]. It is worthwhile to note that previous studies are mainly

106 focused on the underlying mechanisms of torrefaction [36, 37] and factors affecting its
107 performance [38] while less attention has been paid to the efficiency of energy
108 utilization for torrefied biomass. This can be readily achieved by life cycle assessment
109 (LCA). Adams et al. [39] assessed torrefaction process in wood pellets production, and
110 found out that torrefied pellets reduced energy consumption: the cumulative energy
111 per kg of modelled torrefied pellets is 18% lower than for the equivalent wood pellets,
112 with primary energy input for torrefied pellets is 43% lower than wood pellets. Rivera
113 et al. [40] identified the energy balance of torrefied microalgal biomass production
114 based on a life cycle approach. The energy analysis showed that torrefaction improves
115 the energy content of the microalgal biomass from 20.22 to 27.93 $MJ \cdot kg^{-1}$.
116 Torrefaction, among all other scenarios, had the least energy requirement by 20% and
117 less. Lin et al. [41] evaluated the energy return on investment (EROI) to determine the
118 sustainability of torrefied biochar. As the torrefaction temperature increased, EROI
119 index gradually increased from 13.1 to 22.9 for the Ananas comosus peel and from
120 10.9 to 21.9 for the Annona squamosa peel. Fantozzi and Buratti [42] presented a LCA
121 study on household heat from the combustion of wood pellets, where energy return
122 index was calculated. It was reported that energy return index was equal to 3.25 for
123 biomass chain, clearly above the breakpoint, equal to 1. However, for the pellet
124 production process, mass and energy flows were measured on an existing Italian
125 pelleting plant which was completely different from the domestic situation. A number
126 of articles have looked at LCA to quantify and compare the energy performance of
127 biomass pellets, but a wide variation of results can be found in the literature due to
128 the selection of methods, the source of data, the type of raw materials, etc. [43]. In
129 contrast there have only been limited studies which attempt to evaluate the energy
130 efficiency of this innovative argopellets. This paper addresses some of the existing
131 research gaps by performing LCA of upgraded pellets and comparing the results with
132 different processing conditions.

133 It should be noted that mechanical properties and energy efficiency of argo-
134 pellets are still not comparable with that coal particles or wood pellets if upgraded
135 alone by blending or torrefaction. Other processes to improve the utilization level of
136 agricultural waste are still necessary. One of the possible solutions is to combine
137 blending with torrefaction which not only enables a high calorific value and energy

efficiency, but it also improves mechanical properties of argo-pellets. Since pelleting is usually operated at high pressure and temperature, more energy is required to increase attractive forces between particles. Si et al. [44] showed that the addition of coal tar is able to improve mechanical durability of biomass pellets at ambient temperature and low pressure. Attempts have also been made to use food waste as binder in engineering applications [45, 46]. For example, starch-rich food waste contributes to granulation, increasing mutual adhesion between particles and consequently reducing energy consumption in pressing and improving the stability of pellets [47]. Although it is feasible in theory, the use of starch-rich food waste as a binder has not been verified experimentally.

The objective of this study is to investigate the properties of upgraded argo-pellets manufactured from torrefied corn stover and coal cutting waste, with the use of starch-rich food waster as binder. The effects of coal cutting waste blending, biomass torrefaction and food waste additive on argo-pellets were systematically examined, together with their mechanisms. Experimental conclusions drawn in this work can provide guidelines for argo-pellets optimisation and upgrading in the future.

2. Materials and methods

2.1. Materials

Corn stover used in this study was collected from Henan Province, China. Stover feedstock was washed with distilled water, dried for 36 hours at 105 °C and then ground to powders to ensure a particle size smaller than 2 mm. Torrefaction was conducted in a horizontal sliding tube furnace (TL1200, BYT). After torrefaction at 200 ℃ for 1.0 hour, the heating furnace was slid back and torrefied corn stover (TCS) was cooled in a quartz tube. Coal cutting waste from a coal processing plant in Anhui Province was used in this experiment. Starch-rich food waste was mainly from bread and pancake, which prepared for the binder after simple cleaning, drying, and grinding.

The contents of carbon and hydrogen were quantified by Liebig method (ISO 625:1996). Nitrogen concentration was determined by Semi-micro Kjeldahl method (ISO 333:1996). Infrared spectrometry (ISO 19579:2006) was used to quantify the sulfur content, and the remaining oxygen percentage was calculated by the difference according to $O* = 100 - (C + H + N + S)$. The proximate analysis was referred to GB/T

212-2008 standards [48]. According to method ASTM E 711, the calorific value was determined as the quantity of heat liberated by combustion of a unit quantity of fuel with oxygen in a standard bomb calorimeter. Three replicates were measured for each raw material. **Table 2** lists the measured properties of raw materials used in this study.

Table 2. Ultimate and proximate analysis of CS、TCS and CCW

Parameter	Proximate analysis (*wt* %, ad.)				Ultimate analysis (*wt* %, ad.)					$Q_{net,ad}$ (MJ/kg)
	M_{ad}	A	V	FC	C	H	O*	N	S	
CS	7.93	5.98	73.59	12.5	41.57	4.93	52.64	0.66	0.20	13.42
TCS	1.37	6.52	78.18	13.93	46.23	4.49	48.59	0.55	0.14	17.71
CCW	6.66	14.75	30.61	47.98	63.77	3.68	31.24	0.81	0.50	24.34

Notes: O*=100%-C-H-N-S; ad: air dry basis

2.2. Pelleting process

Corn stover and coal cutting waste were mixed with the set mass ratios of 9:1, 8:2, 7:3, 6:4 and 5:5. In order to maintain the basic characteristics of argo-pellets, the addition ratio of coal cutting waste was controlled below 50%. Mixture of each proportion was conditioned by adding a fixed amount of starch binder (5% mass), according to the limits of DIN 14961. To compare the effects of starch binder and torrefaction pre-treatment on biomass materials, the other two groups of argo-pellets (one group processed without starch binder and the other group torrefied corn straw) were manufactured at the same blending ratios. These pellets were produced using a laboratory pelletizing unit (PC-10T), made by Tianjin Jingtuo Instrument Technology Co., Ltd, with press channel of 10.0 mm and a diameter of 8.0 mm. The whole pelleting process is illustrated in **Fig. 1**.

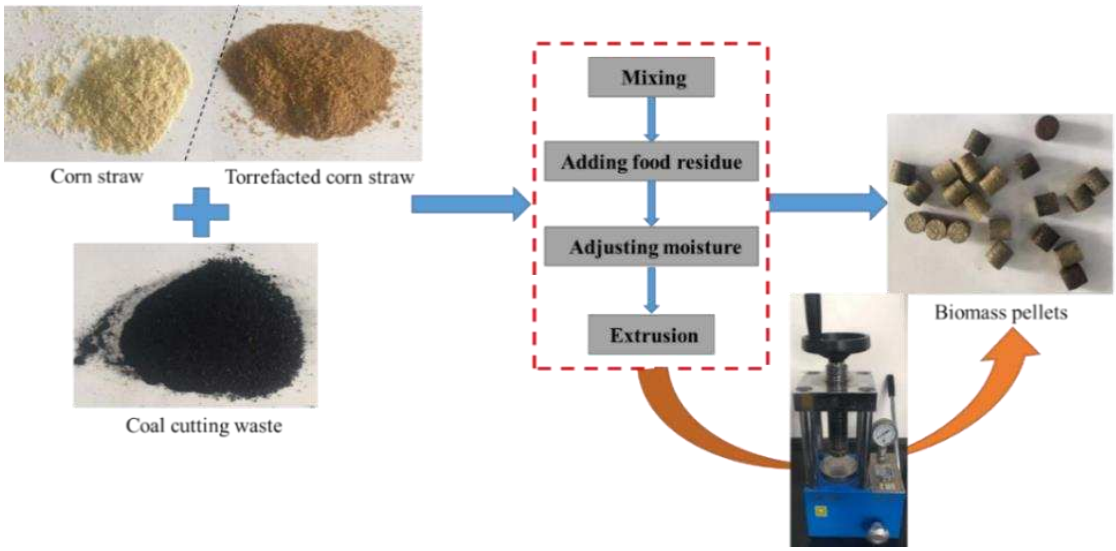


Fig. 1. The pelleting process for pellet samples

2.3. Mechanical properties of pellets

The properties of pellet samples were measured according to Pellet Fuel Institute (PFI) Standard (2011). **Fig. 2** illustrates the pellets preparation and analysis processes. The length (*L*) and diameter (*D*) of randomly selected biomass pellets were measured with a caliper (CD-56C, Aurora, III). Each measured *L* and *D* was recorded to the nearest 0.01 mm. True density (*TD*) was calculated by the ratio of mass to volume. Due to the cylindrical pellet shape, the volume formula was calculated according to the columnar particles. Pellet integrity was established according to the improved PN-G-04651 [49]. Pellet samples passed parabolic test (2 m height) and staircase test (14 standard stair face) successively to determine the loss of fine material and undamaged pellets. Then, the pellet integrity was calculated as the percentage of undamaged pellet weight to the total sample weight. Water absorbability was defined by the standard PN-G-04652 [49]. Pellet samples were taken out from water for two hours, and then placed in a natural indoor environment for a period of time (3 or 7 days). The difference between the sample weight and the initial weight made the measure of this parameter. The compressive strength and SEM analysis were completed by KC-3 digital particle strength tester and JSM-5610LV scanning electron microscope (SEM), respectively.

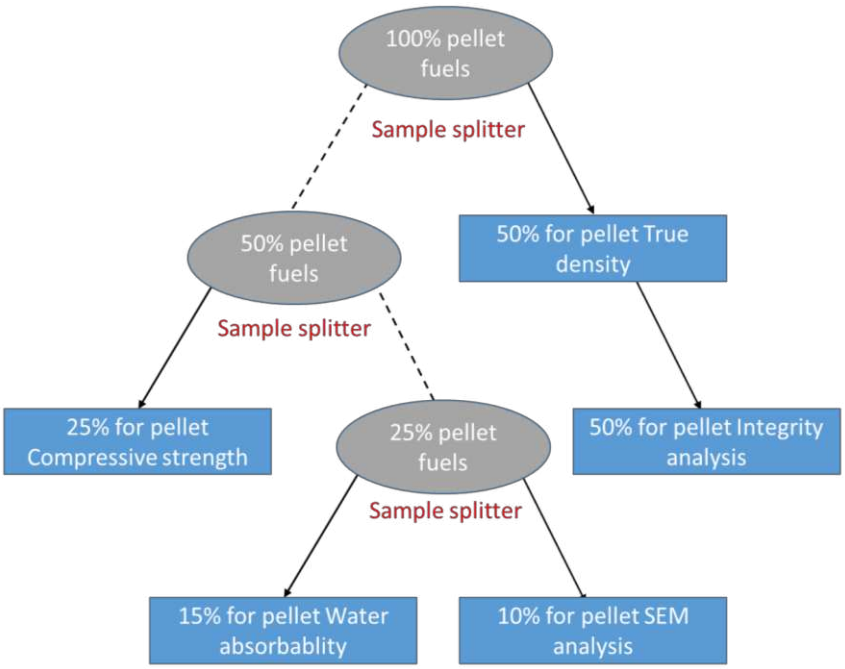


Fig. 2. The flow chart of pellet samples distribution and testing

2.4. Thermal behaviors of pellets

Thermogravimetric/differential thermal analysis (TG/DTA) experiments were performed on a Hitachi synchronous thermal analyzer (STA 449F5®) to characterise thermal properties of samples. In this experiment, 10±0.5 mg material was heated from 25 to 1000 °C at a heating rate of 20 °C/min under 40 mL/min air gas. However, thermogravimetric/mass spectrometry (TG/MS) technique were carried out on Netzsch thermal analyzer (STA 449F3®) coupled with Blazers mass spectrometer (Omnistar 200) to evaluate and characterise of combustion characteristics In each experiment, 100 mL/min of air gas flow rate and 20 °C/min of heating rate was used from ambient temperature to 1000 °C. Mass spectrometry system was operated under a vacuum and detected the characteristic fragment ion intensity of the volatiles according to respective mass to charge ratios (m/z). The number of m/z signals selected were: 18.03 (H_2O^+), 30.00 (NO^+), 43.94 (CO_2^+), 46 (NO_2^+), and 63.96 (SO_2^+). For each experimental process, it was done in triplicate for accuracy and the relative error among all the data was controlled within 5%. After all the experiments, thermal data was derived and processed by Origin 8.5 to obtain multiple thermodynamic curves to compare the combustion difference of different pellet fuels.

2.5. Life cycle assessment of pellets

This study aims to quantitatively assess the quality of argo-pellets by life cycle assessment (LCA) meanwhile exploring the feasibility of upgrading argo-pellets. Energy Return Ratio (ERR) is the ratio of the amount of usable energy obtained from a resource to the amount of energy expended to produce that net amount of energy [50]. The larger the ERR, the greater the net energy output and the higher the energy efficiency. In this study, the reference functional unit was selected as one-ton pellet fuel production. The corresponding calorific value was measured with a standard bomb calorimeter. Energy input of production equipment, plant, land and living facilities are not considered. The upstream production data of the material is also ignored, as raw material weight is less than 5%. In addition, waste and disposal are excluded from the boundary. **Fig. 3** presents the system boundary for pellets production, including three key stages which are: corn planting and coal mining (ST1); feedstocks collection and transportation (ST2); and pellet fuels production (ST3: chopping, drying, and co-pelleting).

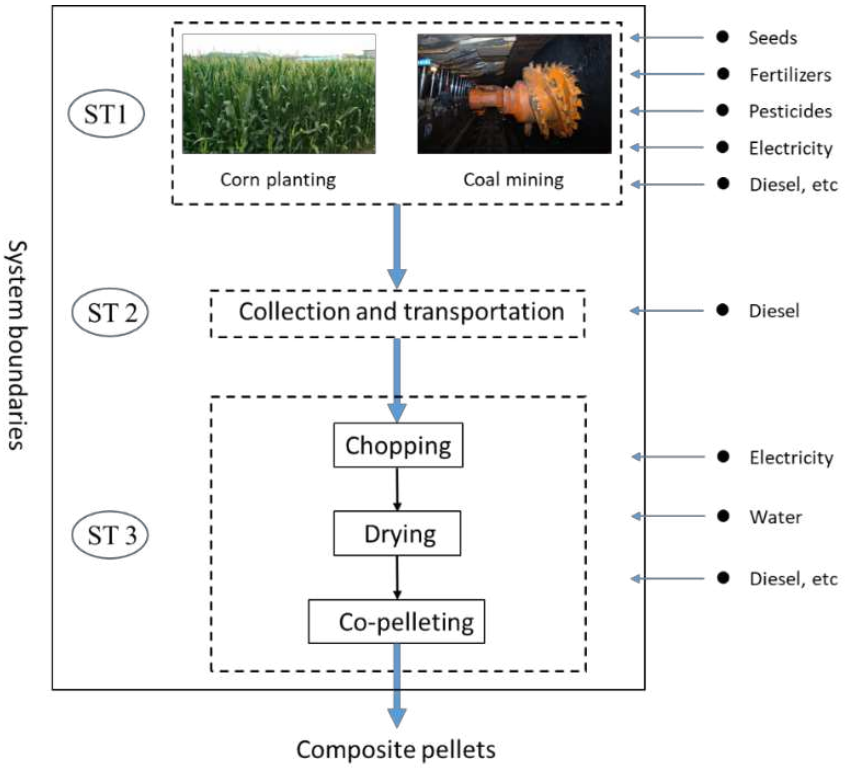


Fig. 3. The diagram of system boundary of pellets production

Table 3 presents the inventory data for corn planting stage per functional unit,

without considering output to technosphere. The consumption of raw materials is mainly collected from the calculated results of Henan statistical yearbook and literature [51]. Some necessary background data, such as the generation and transportation of raw materials, are derived from Ecoinvent 3.1 database, which is representative of China. As an agricultural waste, the yield of corn stover is 1.2 times of corn while its price is one tenth of corn. Based on the principle of economic distribution [51], the proportion of energy consumption in corn planting allocated to corn and corn stover are 90% and 10%, respectively. For coal mining, the relevant energy input data and background data are derived from similar reference [52]. The second stage (ST2) is the collection and transportation of raw material. According to the previous study [51], the average collection distance from fuel plant to corn planting area is set as 15 km, and the distance between fuel plant and coal area is about 50 km. Feedstocks are transported by the third-party trucks (Dongfeng, 8t load capacity), with average diesel consumption of 0.15 L/km and load factor of 50%. The last process (ST3) combines chopping, drying, and co-pelleting. Long corn stover is first crushed and dried, and then sprayed with water to adjust the water content approximate 10% before co-pelleting with dried coal cutting waste. Electricity is mainly consumed by drying machine, chopping device, and pelleting machine. For torrefied biomass, the power consumption includes vacuum torrefaction equipment. The natural loss and lifespan of various mechanical equipment are not considered in the LCA calculation. The consumed water, electricity, diesel, etc. are calculated according to actual pellets processing plant in Henan province, which are listed in **Table 4**.

Table 3.. Inventory data for corn planting per functional unit (1t pellets)-ST1

Item	Total Amount	Unit	Corn	Corn stover
Nitrogen fertilizer (N urea)	15.78	kg	14.2	1.58
Phosphate fertilizer (P_2O_5)	8.844	kg	7.96	0.884
Potassium fertilizer (K_2O)	6.722	kg	6.05	0.672
Pesticides	0.789	kg	0.71	0.079
Electricity	36.4	kWh	32.76	3.64

Diesel	10.9	L	9.81	1.09

Table 4.. Inventory data for pellet fuels production per functional unit (1t pellets) –ST2 and ST3

Item	Amount	Unit	Description
Corn stover	1100	kg	Used for raw material, loss rate of the whole process is about 10%
Diesel (Collection and transportation)	0.306	L	Corn stover collection and transportation
Electricity-Drying	32.7	kWh	JTSG1415，Electricity consumption during drying
Electricity-Chopping	2.5	kWh	HY-400, Electricity consumption during chopping
Electricity-Torrefaction	34.87	kWh	HY-8, Electricity consumption during torrefaction
Electricity-Pelleting	93.4	kWh	RGKJ560, Electricity consumption during pelleting
Water	22.77	kg	Water spraying in pellets production
Diesel (In-plant transportation)	0.12	L	For transportation of material and pellet fuels

3. Results and discussion

The photo of manufactured pellet fuels under different process conditions are presented in **Fig. 4**. As feedstocks are pressed by machine, the particle size of pellets is generally maintained, with a length of 8 mm and a diameter of 10 mm. Most of the briquette fuels exhibit excellent formability and integrity, and the color of pellets changes obviously: with an increase of CCW mixing ratio, the color of pellet fuels deepens. Moreover, the color of torrefied pellets (TCS+CCW+5%S) is obviously darker than that of pellets without torrefaction (CS+CCW and CS+CCW+5%S). Theoretically, the darker the color, the higher the calorific value, but this needs to be verified by combustion experiments. The effect of starch-food binder on pellets cannot be identified directly, hence further physical test and analysis are required.

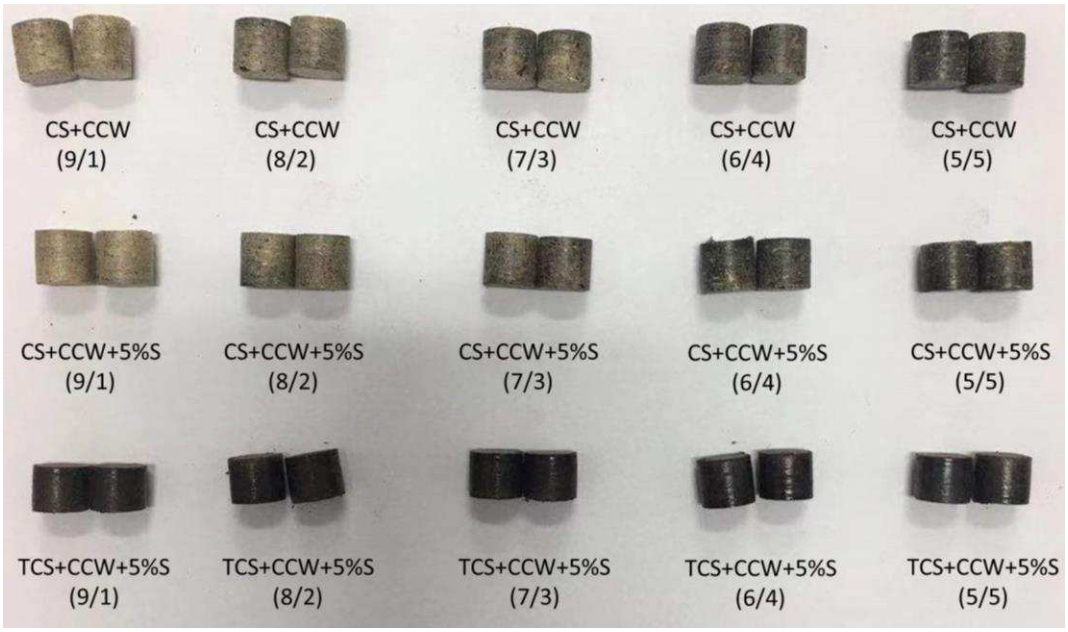


Fig. 4. Pellet fuels under different experimental conditions (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste; S: starch-binder)

3.1 Mechanical properties

3.1.1 True density

True density is an important parameter for pellet fuels, calculated as the ratio of mass to volume of single pellet. A large true density corresponds to a high transport efficiency and a small storage space [53]. **Fig. 5** presents the true densities of various pellet fuels. It is obvious that the pellet densities (CS+CCW and CS+CCW+5%S) increase with CCW percentage. The reason can be attributed to the higher density of CCW than that of CS. For torrefied corn stover (TCS), the advantage of high-density CCW is not that obvious. **Fig. 5** indicates that pellet density (TCS+CCW+5%S) remains nearly unchanged with the addition of CCW. However, the density of torrefied pellets (TCS+CCW+5%S) is much higher than that without torrefaction. Basically, all pellet fuels can meet the DIN 51731 standard (i.e., 1000 kg/m^3), except for CS pellets blending with 10% CCW. However, once 10% CCW is mixed with TCS, the pellet density is much higher than 1000 kg/m^3 as a result of torrefaction.

The addition of starch binder has a positive impact on pellet density, which is obvious when the CS fraction is less than 70%. The improvement on pellet density is more obvious with further decrease of CS proportion. In contrast, as the proportion of CS is 80% or 90% in **Fig. 5**, the addition of starch binder has little influence on true

density. Previous studies have shown that lignin was the main component of self-binding in stover, which combined with cellulose and hemicellulose to produce pellets by intermolecular attraction and entanglement [54, 55]. It may be speculated that the external starch binder plays the main role in palletization with less CS, and the internal lignin binder will gradually act with increasing CS. However, relevant inferences and assumptions need to be verified in future studies.

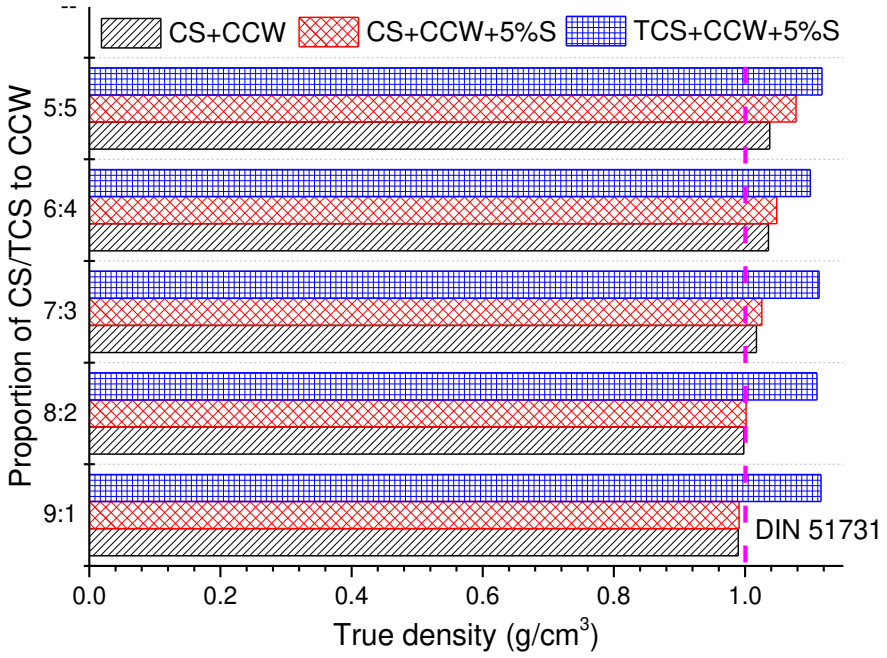


Fig. 5. Pellet true density as related to CS/TCS to CCW and starch binder addition (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste; S: starch-binder)

3.1.2 Pellet integrity

Fig. 6 shows that with increasing CCW percentage, the pellet integrity increases first and then decreases. The maximum value of integrity is found at a mixing ratio of (8:2) between CS and CCW. Blending different feedstocks can improve pellet integrity, and similar conclusions was also drawn in previous study [56]. Compared with CS, the particle size of CCW is much smaller. The addition of a small amount of CCW fills the gap between CS particles, which contributes to the improvement of pellet forming and integrity. This is especially so for CS without torrefaction. For instance, when CCW blending ratio increases from 10% (9CS:1CCW) to 20% (8CS:2CCW), the pellet integrity is improved by 1% (from 98.2% to 99.2%). In the presence of starch binder (CS+CCW+5%S), particle size is not the only factor affecting pelleting. The integrity of

mixed pellets (TCS+CCW+5%S) remains nearly unchanged while the physicochemical properties of torrefied biomass are modified, and the effect of particle size is significantly reduced.

Overall, the pellet fuels meet the DIN 1496 class-A requirement (97.5% for integrity). The pellet integrity after torrefaction (TCS+CCW+5%S) is higher than that without torrefaction (CS+CCW+5%S) and the pellet integrity with starch binder (CS+CCW+5%S) is better than that without starch binder (CS+CCW). Corn straw contains chemical extractives, such as low molecular weight fatty acids, waxes, sterols and terpenes [57]. When CS material is milled, these chemical substances migrate to particle surface and thus hinders the binding of CS molecules. This phenomenon is called passivation which reduces compression strength and integrity of pellets, as confirmed by Bergstrom et al. [58]. After torrefaction treatment, some of these chemical extractives from CS can be removed, thus reducing this passivation effect. This is the reason for the high pellet integrity of pellet fuels (TCS+CCW+5%S). Starch binder mainly includes dextrin, cellulose, free sugar, and pentosan, which plays an important role in pelleting process. The internal forces of pelleting includes attractive force, interfacial force, capillary force, adhesive and cohesive forces, solid bridges of particles [59]. Liu et al. [56] found that the predominant bonding was created by solid bridges during palletization. Adding starch additive can strengthen these solid bridges and promote pellet formation. Clearly, the effect of starch binder is dependent on the mixing ratio of CS and CCW. In **Fig. 6**, with the decrease of CS mixing ratio, the positive effect of starch on integrity is reduced. Even when the proportion of CS is 50%, the addition of starch binder reduces pellet integrity. This may be related to the reaction between starch binder and the components of CS or CCW, resulting in synergy or antagonism.

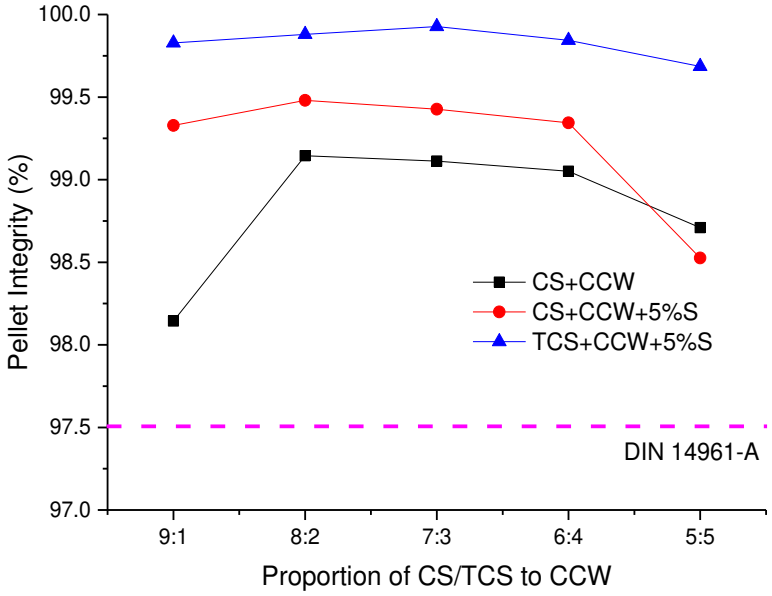


Fig. 6. Pellet integrity as related to CS/TCS to CCW and starch addition (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste; S: starch-binder)

3.1.3 Compressive strength

Similar to pellet integrity, the compressive strength of pellets rises first and then falls, as shown in **Fig. 7**. When the mixing ratio of TCS is 70%, the pellet fuel has the maximum compressive strength of 1897.67 N/cm^2. The addition of starch binder also improves the compressive strength from 1880.67 to 1895.33 N/cm^2. However, the effect of torrefaction is extremely weak, and the compressive strength remains largely stable, from 1895.33 to 1897.67 N/cm^2. SEM results in **Fig. 8** show that the surface of unroasted pellets has an obvious layered structure, mainly due to the spiral extrusion in channel. The material without starch binder (CS+CCW) has rough surface, loose texture and a small amount of gap and pore structures. After the addition of starch-binder, the layered structures (CS+CCW+5%S) are more obvious, orderly, and compact. This structural difference caused by starch-binder is the main reason for the improvement of pellet integrity and compressive strength. Torrefied granular fuel (TCS+CCW+5%S) still contains layered structures, but the layered structures are interspersed with partial melting and crosslinking, resulting in the reduction of intermittent space and the enhancement of compactness. This may be the reason why torrefied pellets have greater integrity and compressive strength.

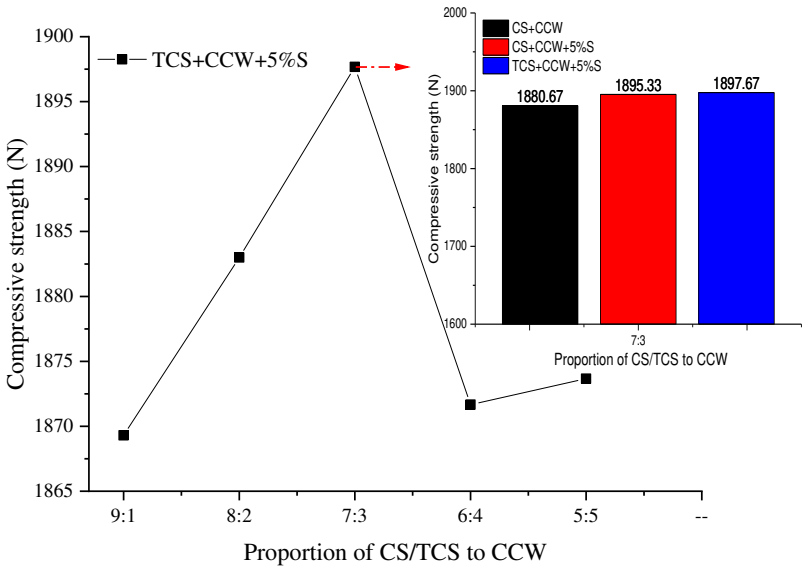


Fig. 7. Compressive strength as related to CS/TCS to CCW and starch addition (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste; S: starch)

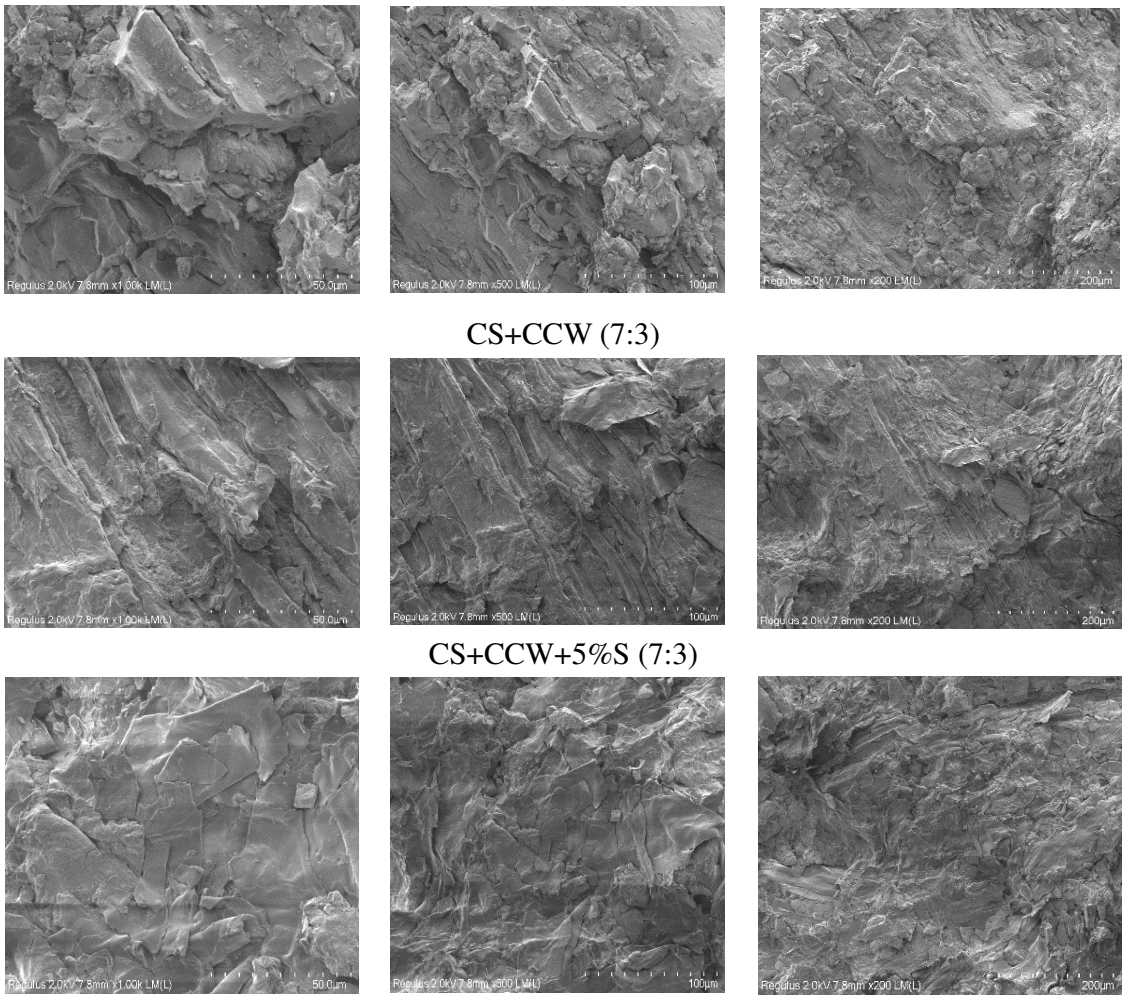


Fig. 8. SEM of pellets (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste; S: starch)

3.1.4 Water absorbability

Regardless of whether starch binder is added or not, pellet fuels without torrefaction decompose obviously after soaking in water for 24 hours as shown in **Fig. 9**, indicating that their water resistance stabilities are relatively poor. On the contrary, the torrefied fuel presents good water resistance and still has high integrity after soaking for a period of time. This is mainly related to the composition of CS material. Hemicellulose in corn straw is rich in hydroxyl groups, which are the main groups of water absorption [20]. After torrefaction, the hydroxyl groups of corn straw are decomposed by heating, which weakens the water absorption function. In addition, the surface of torrefied particles contains some hydrophobic organic matters [20], which can further reduce the water absorption.

The water absorbability of pellet fuels is closely related to the weak bond between particles and the corresponding pore structure. **Fig. 9** plots water absorbability of pellet fuels versus CS/TCS proportion. The water absorption decreases gradually with decreasing proportion of CS (from 90% to 50%). Moreover, water absorption of torrefied pellets (TCS+CCW+5%S) was much less than that of the non-torrefied pellets. The presence of starch binder (CS+CCW+5%S) leads to a smaller water absorption than that without starch (CS+CCW). Torrefaction heating destroys some hydroxyl groups; therefore, the water absorption is weakened. The addition of starch binder during pelleting process improves the mechanical strength and reduces the pore structures between particles. In general, the above two mechanisms of reducing water absorption are essentially different. The effect of time on the decrease of water absorption is further investigated. It was found that the water absorption of pellet fuels decreases differently with times from 3 to 7 days. Among them, the water absorption of pellets without starch-binder (CS+CCW) decreases the most up to 40～50%, and the decline seems to be related to the proportion of CS. When the CS percentage is the highest, the largest decline is obtained. Moreover, a smaller decrease is observed at a lower CS ratio, which is mainly due to the role of hemicellulose in CS. This difference shows that the hydroxyl weak bond is unstable and easy to be affected by external environment. Different from the function of hydroxyl bond, starch-binder mainly acts instantaneously. With the increase of time, water absorption

(CS+CCW+5%S) decreases, with a reduction range smaller than that without binder (CS+CCW) and a minor influence due to the ratio of CS. In comparison, pellet fuels with the smallest water absorption corresponds to a TCS proportion of 50%, implying that torrefaction greatly improves water resistance and stability of pellet fuels.

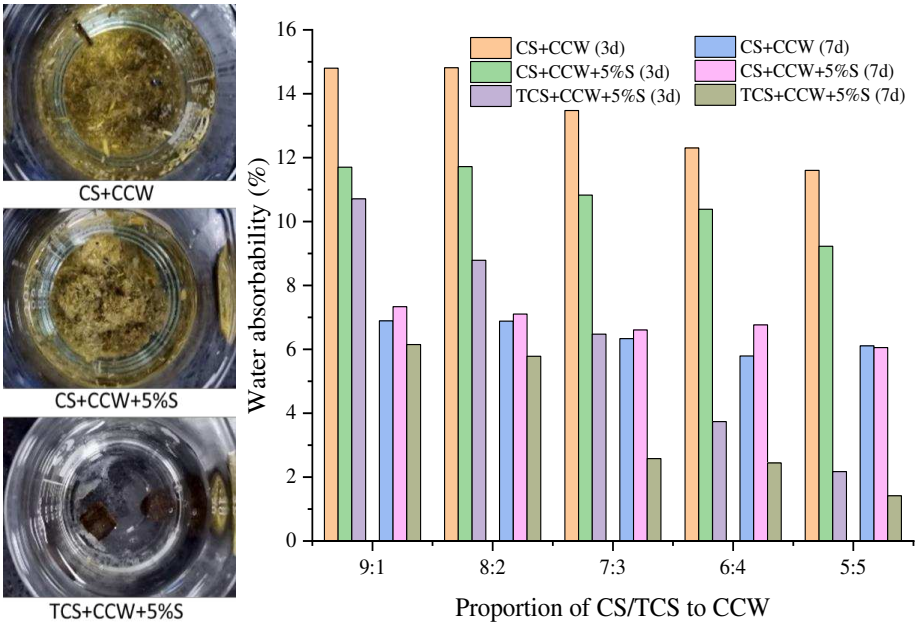


Fig. 9. Pellet water absorbability as related to CS/TCS to CCW and starch addition (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste; S: starch)

3.2 Thermal behaviors

3.2.1 TG-DTG-DTA

Thermogravimetric analysis is an efficient method widely used for the analysis of combustion process [60]. **Fig. 10** shows the thermal profiles of CS and CCW. The weight loss of CS can be observed in three main stages: first) the dehydration and drying stage ranged from 80 ℃ to 160 ℃; second) the pyrolysis of lignin and cellulose, and the release of volatile matter starting from 160 ℃ and finishing at 370 ℃ and third) coke combustion took place between 370 and 470 ℃. The first weight-loss stage is mainly due to the evaporation of water molecules. The second stage represents the loss of CO, and the third stage of weight-loss is mainly caused by volatilisation of CO_2. The final stage was characterised by a slight weight loss, which is mainly caused by the further cracking of C-C and C-H bonds. Three weightlessness peaks can be observed from the DTG curves, with the volatile release-combustion peak much larger than the peak of fixed char combustion. This is mainly because the volatile matter of CS is much

greater than its fixed char, thus showing a higher rate of release and combustion.

The weight loss of CCW combustion can be divided into two stages: dehydration (90 ~ 220 ℃), and volatile release and fixed char combustion (220 ~ 580 ℃). The first step is weaker than that of CS as the moisture content of CCW is lower than that of CS. The second weight-loss step is related to fixed char, where the weight loss increases with fixed char in CCW. There are two weightlessness peaks on the DTG curves: dehydration peak and the peak of volatile release-combustion and fixed char combustion. Due to the low volatile content and high volatile release temperature of CCW (390 ~ 500 ℃), the volatile release-combustion peak of CCW is not fully reflected. The volatile release and combustion are always accompanied by the combustion of coal char. There is only one obvious weight loss peak except the dehydration peak on the DTG curves.

By comparing the DTG curves of CS and CCW, it can be seen that the combustion process of CS is mainly concentrated in the volatile combustion stage while the combustion process of CCW is mainly concentrated in the fixed carbon combustion stage. Different from the sharp combustion peaks of CS, the volatile release and fixed char combustion peak of CCW shows a shoulder peak.

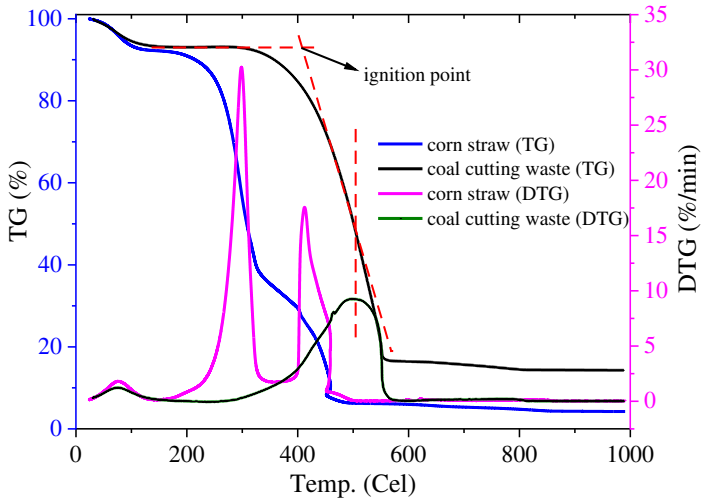


Fig. 10. TG and DTG curves of corn straw/coal cutting waste

It can be observed that TG and DTG curves of CS/TCS and CCW blends are arranged in order according to the proportion of feedstocks. As shown in **Fig. 11(a)**, the same first peak is obtained for both, this might be due to the dehydration of

moisture from 80 to 160 °C. Note that the dehydration stage is not analysed here. The combustion process mainly consists of two stages: volatile release-combustion and fixed char combustion. TG curves of blends basically coincide when the temperature is lower than 120 ℃ while differs with a temperature over 120 ℃. With the increase of CCW ratio, TG curves move towards the high temperature zone, and the ignition temperature (T_i) and burnout temperature (T_b) of the mixtures rise gradually, as shown in **Table 4**. In addition, the weakening of the second weight loss step is more pronounced due to a low volatile content of CCW, and the third step is gradually enhanced due to a higher fixed carbon content of CCW than that of CS. Comparing TG curves with or without torrefaction treatment, the second weight loss step is weakened after pre-treatment, indicating that torrefaction is conducive to pyrolysis and volatile precipitation. Torrefaction improves the fuel performance of biomass, mainly reflected in the improvement of moisture content, fixed carbon content, and energy density. Torrefaction had a significant impact on the distribution and characteristics of biomass pyrolysis products. The increase of pyrolysis carbon from torrefied biomass was mainly due to the large decomposition of hemicellulose and the sharp increase of lignin content, while the lignin pyrolysis products were mainly biochar.

According to DGT curves shown in **Fig. 11(b)**, the greater the CS proportion in the mixtures, the higher the volatile content, and the greater peak value of volatile release and combustion. Theoretically, the lower the CCW ratio in the mixtures, the lower the fixed carbon content and the smaller peak value of coke combustion. However, the coke combustion peak is the highest (D_{max}=23.5 mg/min) when the ratio is 7:3 (CS: CCW). Feedstocks with high carbon content can continuously provide higher heat for the combustion process, so as to promote the combustion of fixed carbon or residual samples. Meanwhile, raw materials with high ash content are easy to generate ash shell during burnout stage, which hinders the inward infiltration of oxygen and the outward diffusion of combustion products. The calorific value of CCW is large, but the ash content is also high. Therefore, only selecting the appropriate mixing proportion can help to improve the burnout characteristics of blending pellets.

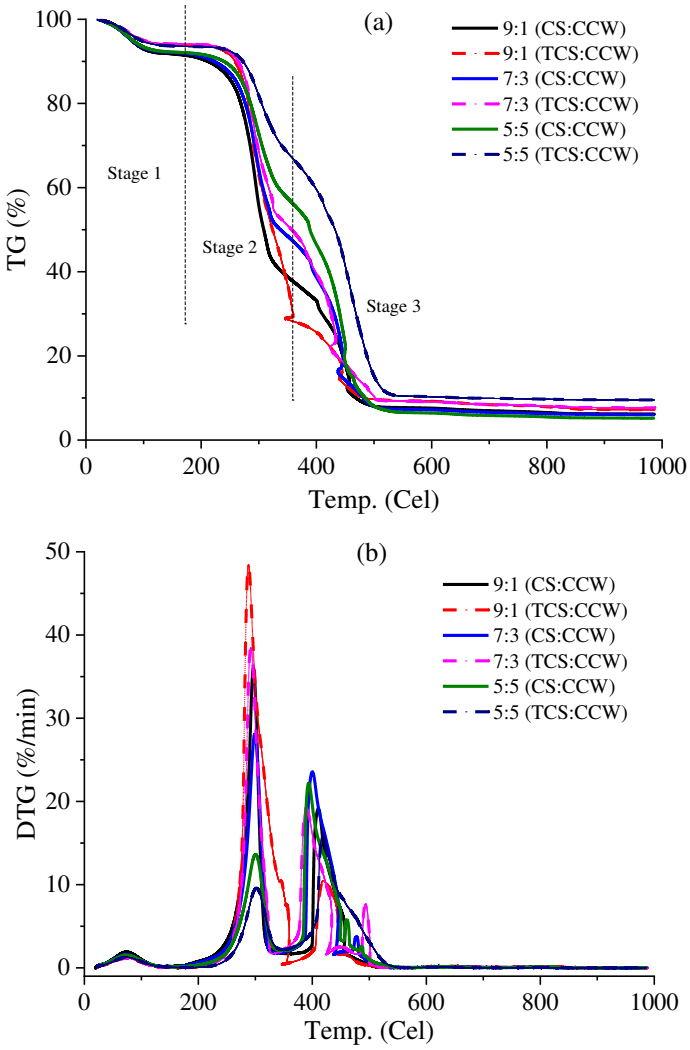


Fig. 11. TG and DTG curves of CS/CCW and TCS/CCW at different ratios (CS: corn straw; TCS: torrefied corn straw; CCW: coal cutting waste)

The peak values of volatile release and combustion increases after torrefaction as shown in **Table 5**. This is mainly due to the increase of calorific value of torrefied CS. Torrefied CS combustion preheats CCW and promotes the release of volatile matters, which is conducive to pellets ignition and burnout. However, when the CCW mixing ratio was larger (at 50%), the decrease of DGT peak is not conducive to the overall combustion, which is also related to the high ash content from CCW.

DTA curves with exothermic and endothermic areas are presented in **Fig. 12**, which are similar to mass loss trend of TG curves. By integrating the area under DTA curves, the heat release from the entire combustion process can be obtained to understand the thermodynamic behavior. Experimental results showed that the calorific value of pellet fuel can be improved by mixing with CCW (from 13540.8 to

19467.1 J/g) and torrefaction pretreatment (form 13973.8 to 18659.1 J/g, from 17271.6 to 19264.1 J/g, from 19467.1 to 23049.3 J/g). The corresponding interval of exothermic is devolatilization or combustion (stage 2) and fixed char combustion (stage 3). The endothermic peak is mainly caused by the endothermic volatilization of moisture.

Table 5. The combustion characteristics of pellet fuels

Samples		Volatile release and Combustion			Char combustion			
	T_i (°C)	T_p (°C)	D_{max} (mg/min)	M (%)	T_p (°C)	D_{max} (mg/min)	M (%)	T_b (°C)
CS	252	299	30.2	58.1	406	17.4	26.8	505
9:1(CS:CCW)	258	299	35.9	53.7	412	19.0	30.5	518
9:1(TCS:CCW)	261	290	48.2	47	420	10.4	18.9	529
7:3(CS:CCW)	261	300	28	43.4	402	23.5	42.3	534
7:3(TCS:CCW)	260	294	38.2	41	390	19	36.1	545
5:5(CS:CCW)	265	300	13.6	34.6	392	22	52.4	585
5:5(TCS:CCW)	274	302	9.5	25.4	420	16.3	58	599
CCW	408	—	—	—	502	9.2	76.5	650

Notes: T_i= ignition temperature; T_p= peak temperature; D_{max}= the highest combustion peak; M= mass ratio; T_b= burnout temperature.

Obviously, the heat peak of pure CS is higher at char combustion stage than that at the volatile combustion stage with a ratio of 2, indicating more heat is released per unit weight during coke combustion. Compared with CS combustion, the CCW peaks in the whole DTA curves are smoother in **Fig. 12(a)**. Similar conclusions have been drawn by previous studies [49, 61]. Theoretically, with the addition of CCW, the thermal profiles of CS/CCW will become smoother, as the desired result of combustion of fuel mixtures [48, 62]. **Figs. 12(a)-(d)** demonstrated that the addition of CCW smoothed CS thermal curves, but the effect was very limited. Moreover, when the CCW blending ratio is 30%, the DTA peak of CS/CCW reaches the maximum value of 380 uV, which may be related to the synergistic effect of CS and CCW [48]. Compared with original CS, the torrefied CS increased the DTA peak of volatile combustion and decreased the peak of char combustion, and the whole thermal curve tended to be smoother. In other words, the combustion of TCS/CCW presents relatively small

difference in heat distribution. The use of fuels with small differences in heat distribution curves can improve the availability of the boiler in operation.

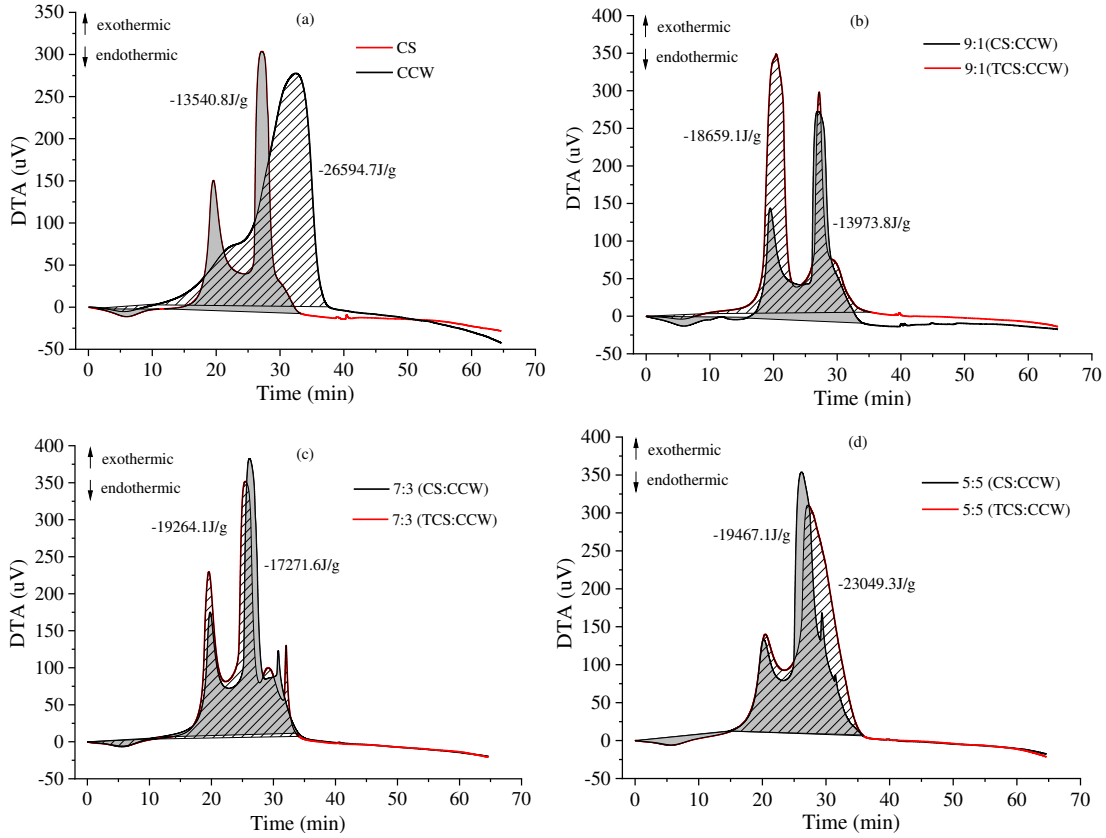


Fig. 12. DTA curves of CS/TCS and CCW at different ratios (a=CS/CCW; b=9:1; c=7:3; d=5:5)

3.2.2 TG-MS

Fig. 13 presents the variation of ion current of various gaseous components with temperature at the same heating rate of 20 °C/min. Moisture vaporised at about 100 ℃ and displayed a good ion peak as shown in **Fig. 13(a)**. The mass evaporation of water mainly occurred within the temperature range of 200 to 400 °C, corresponding to the volatilization stage[63]. For different pellet fuels, the largest H_2O peak is observed for pure CS, reaching 3.8×10^{-9} A/mg. Because the water content of CCW (6.66%) is slightly lower than that of CS (7.93%), when they are mixed up to produce the new blending pellets (0.7CS:0.3CCW), there is a slight decrease in H_2O peak. Similarly, after torrefaction treatment, the moisture ratio of CS decreases sharply (1.37%) and the H_2O peak of blending pellets (0.7TCS:0.3CCW) is the smallest. Biomass is mainly composed of cellulose, hemicellulose and lignin, and their contents affect the moisture release process directly. Hemicellulose begins to decompose and release water molecule

549 approximately at 200 ℃, while the temperature for cellulose and lignin is higher, about
550 250 ℃ [64]. However, most of water was lost after torrefaction, thereby the H_2O peak
551 is not very prominent compared with the case without torrefaction.

552 The CO_2 ion profiles are distributed in the entire combustion process, as shown
553 in **Fig. 13(b)**. Two prominent peaks of CO_2 can be found between 200 and 600 °C, which
554 are attributed to the decomposition of anhydrides, phenols, quinones, carbonates,
555 carbonyls and ethers [65]. Note that there is a slightly different peak in the range of
556 600 to 800 °C. This indicates that carbonates or covered carbon in ash is decomposed
557 at high temperatures during combustion. For different pellet fuels, it is obvious that
558 both blending CCW and biomass torrefaction shift the curve towards right, implying
559 that a higher burnout temperature is required for fuels burnout. This change is mainly
560 related to the increase of carbon content in pellet fuel, which would consequently
561 cause an increase in carbon dioxide emission.

562 Fuel nitrogen mainly exists in the form of gaseous nitrogen, tar nitrogen and char
563 nitrogen. The main products in volatilization stage are volatile nitrogen and char
564 nitrogen, in which volatile nitrogen is composed of gaseous nitrogen and tar nitrogen.
565 Gaseous nitrogen (NH_3, HCN, HNCO) can be further converted to NO, in which partial
566 NO is oxidised to NO_2, or N_2 is generated under the reduction of hydrocarbons and
567 coke. At high temperature, char nitrogen is further oxidised through combustion
568 reaction to produce NO and NO_2 [66].

569 Elemental analysis in **Table 2** shows that the content of nitrogen in CCW is greater
570 than that in CS. In theory, the combustion of CS mixed with CCW will release more
571 nitrogen compounds. However, when the temperature is low (<400°C), nitrogen
572 mainly exists in the form of gas-phase volatiles, and CS itself contains more volatile
573 components, thereby it presents a very obvious large NO peak in **Fig. 13(c)**. With the
574 increase of temperature, a relatively gentle NO peak appears on the CS curve, which
575 is because most of the nitrogen precipitates in the form of volatiles and a small amount
576 exists in char. In the temperature range of 400 °C - 650 °C, the content of char nitrogen
577 in CCW is higher, and presents an obvious NO peak compared with CS. When the
578 temperature reaches about 650 ℃, the ion current of NO and NO_2 increase briefly in
579 **Fig. 13(c) and(d)**, respectively. A portion of nitrogen-containing compounds exited as
580 the nitrate or covered organic in CS and CCW [63]. As the temperature rises, nitrogen-

containing compounds decomposes to form nitrogen oxides (NO_2 and NO). Meanwhile, NO_2 is reduced to NO on the coke surface at high temperature. It can be confirmed by **Fig. 13(d)** that the concentration of NO_2 decreases significantly at the same temperature. When the temperature is higher than 700 °C, the higher the carbon content, the more carbon reacts with NO_2 and the more NO is generated. Compared with the raw material CS, CCW blending and torrefaction both can improve the carbon content of pellet fuel.

During combustion, the formation of sulfur oxides is related to the occurrence form, temperature and composition of sulfur. In the devolatilization stage, sulfur oxides mainly come from organic sulfur, and sulfur oxides are mainly the decomposition of inorganic sulfur during char combustion. As shown in **Fig. 13(e)**, SO_2 gas is mainly produced in the volatiles release and combustion stage. Due to the low sulfur content in CS, there is only a weak SO_2 peak at 300 - 500 ℃, and the corresponding peak value is 5.0×10^{-13} A/mg. The sulfur proportion of CCW is 2.5 times that of CS, thereby the SO_2 emission peak of co-combustion of CS and CCW is very sharp (1.5×10^{-12} A/mg). Heating during torrefaction can remove some sulfur-containing organic matter in the form of volatiles, resulting in the reduction of sulfur content in pellet fuel, and the SO_2 released by combustion is greatly reduced (8.5×10^{-13} A/mg).

The total gas emission during combustion is calculated by integral method. The cumulative amount of different gas products is shown in **Fig. 13(f)**. As the main gas composition of all pellet fuels, the total amount of H_2O and CO_2 accounts for more than 99% of the total gas products. The co-combustion of CS and CCW leads to the increase of CO_2 and NO_2 emissions, which is mainly due to the high proportion of C and N in CCW. Although the emissions of CO_2 and NO_2 per unit mass increases, the emission of pollutants with the same calorific value will be greatly reduced due to co-combustion. After torrefaction treatment, the content of C element in CS increased, the content of N and S elements decreased, and the element compositions of raw material changed significantly. Meanwhile, the release of NO and SO_2 are reduced in the gas accumulation diagram. Although the CO_2 emission increased, the calorific value of pellet fuel (TCS+CCW) was improved. The results showed that torrefaction

technology was conducive to reduce the pollutant emissions and significantly improve the quality of raw materials.

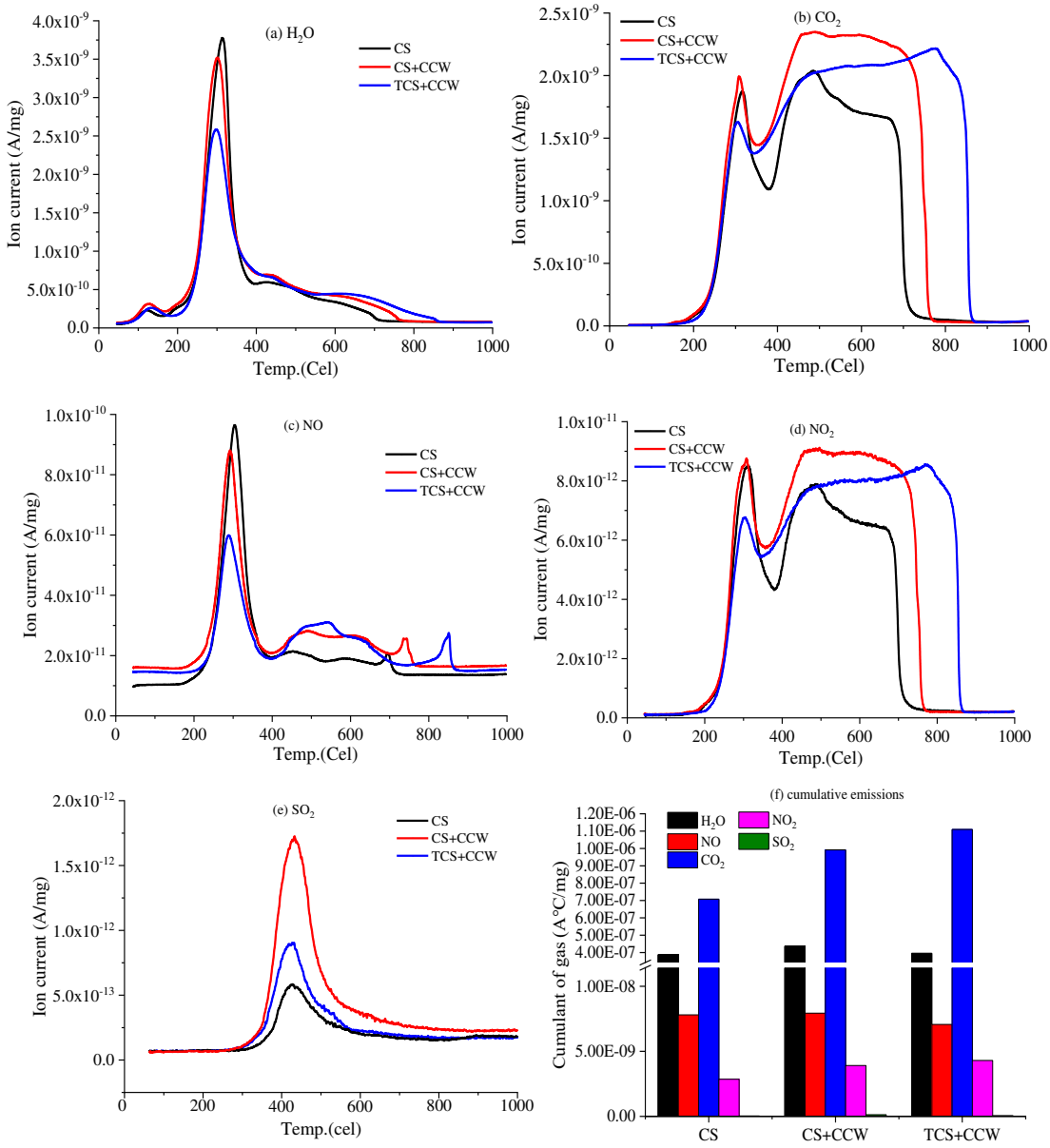


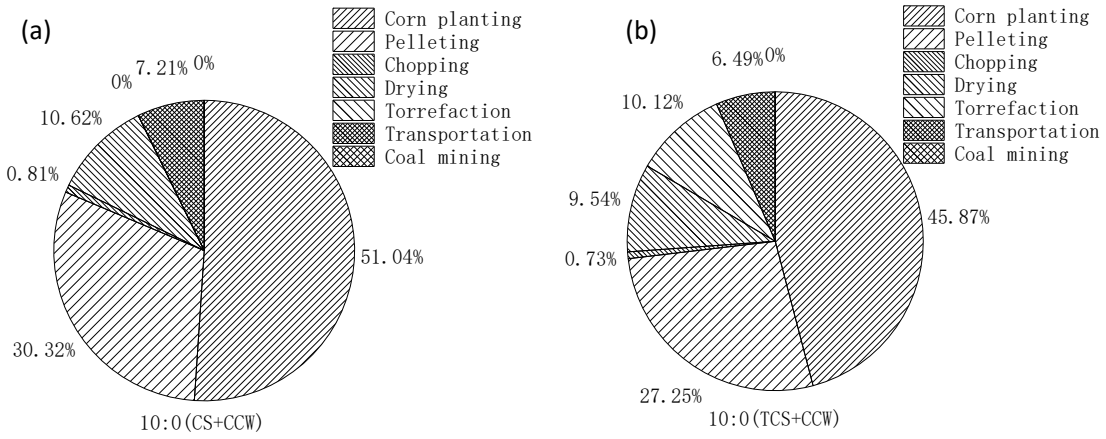
Fig. 13. Gas emission curves of(a) H_2O, (b) CO_2, (c) NO, (d) NO_2 (e) SO_2 and (f) cumulative emissions at the heating rate of 20 °C/min

3.3 LCA analysis

The energy input in the whole life cycle of pellet fuel manufacturing is shown in **Fig. 14**. For pure biomass pellet (CS and TCS), energy input is mainly used for corn planting, accounting for 51.04% (10CS:0CCW) and 45.87% (10TCS:0CCW), respectively; the second is the pressing processing of pellet fuel, with energy consumption accounting for 27.25 - 30.32%. These results are in line with those previously reported

by Hamedani et al. [50], where biomass cultivation and pelletizing were reported as the main contributors to energy consumption in olive pellet production. It is also clear that transportation and chopping are the least consuming energy activities. Torrefaction process is the main power consumption, accounting for about 10.12% of the total energy input, as shown in **Fig. 14(b)**. For biomass pellet fuel blending with CCW, the top three energy consumption are coal mining (37.78 - 41.17%), pellet pressing (22.09 - 24.08%), and corn planting (18.59 - 20.27%). Similar to biomass pellet, the composite pellet fuel consumes less energy in the transportation and chopping processes, consistent with previous studies [42, 50]. The mixing amount of CCW directly affects the total energy input. Therefore, a reasonable selection of CCW blending proportion is critical for energy efficiency of pellet fuel.

Fig. 15 shows that EER increases slowly with CCW blending ratio. Although increasing CCW leads to an increase in the total input energy, a higher calorific value of CCW implies a larger total output heat, thereby showing a better performance of EER. Moreover, the energy efficiency of pellet fuel improves with torrefaction, with the EER increases in varying degrees. However, this extent of improvement depends on the proportion of biomass material, which is more significant at large proportions of biomass.



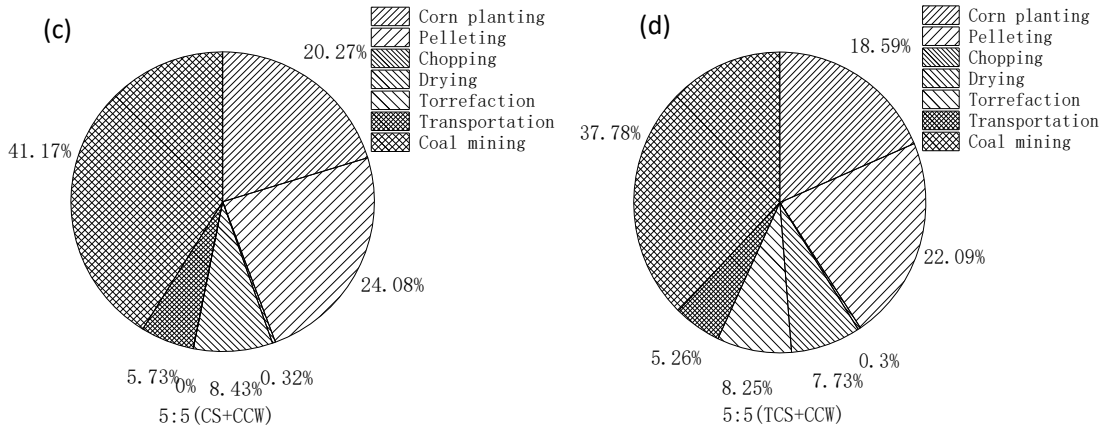


Fig. 14. Comparison of cumulative energy demand in pellet fuel production (a) 10CS:0CCW, (b) 10TCS:0CCW, (c) 5CS:5CCW, (d) 5TCS:5CCW

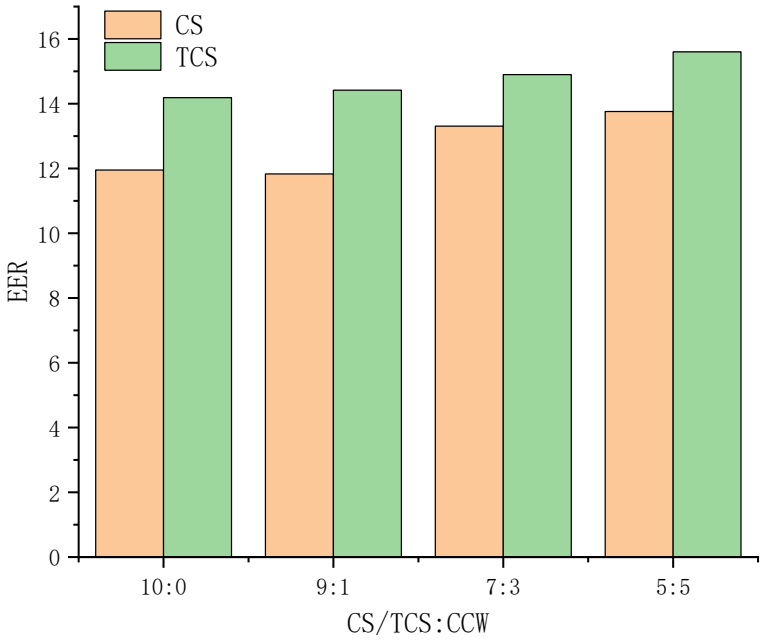


Fig. 15. Energy Return Ratio of different pellet fuel samples

3.3 Discussion and future perspectives

Combining the above-mentioned research findings, a summary of the physical and combustion characteristics of upgraded argo-pellets has been presented in **Fig. 16**. There are two feasible ways to improve the quality of argo-pellets: mixing CCW and torrefaction. Mechanical properties: with increasing CCW percentage, all the physical parameters increases, except that the pellet integrity increases first and then decreases. The addition and use of starch can further optimise this physical

performance. CS torrefaction can improve these mechanical properties in varying degrees on the basis of starch addition and CCW mixing. In terms of compressive strength, the compressive performance of argo-pellets first increases and then decreases, reaching the maximum at 30% mixing ratio of CCW. Combustion characteristics: CCW blending and torrefaction both improve the thermodynamic characteristic parameters, release more calorific value and present higher energy efficiency. However, C, N and S account for a large proportion in CCW, and the simple blending is not conducive to environmental protection. Although torrefaction treatment increases the proportion of C, it releases more net heat energy and reduces the contents of N and S. From the perspective of unit energy output, it is conducive to the emission reduction of pollutants.

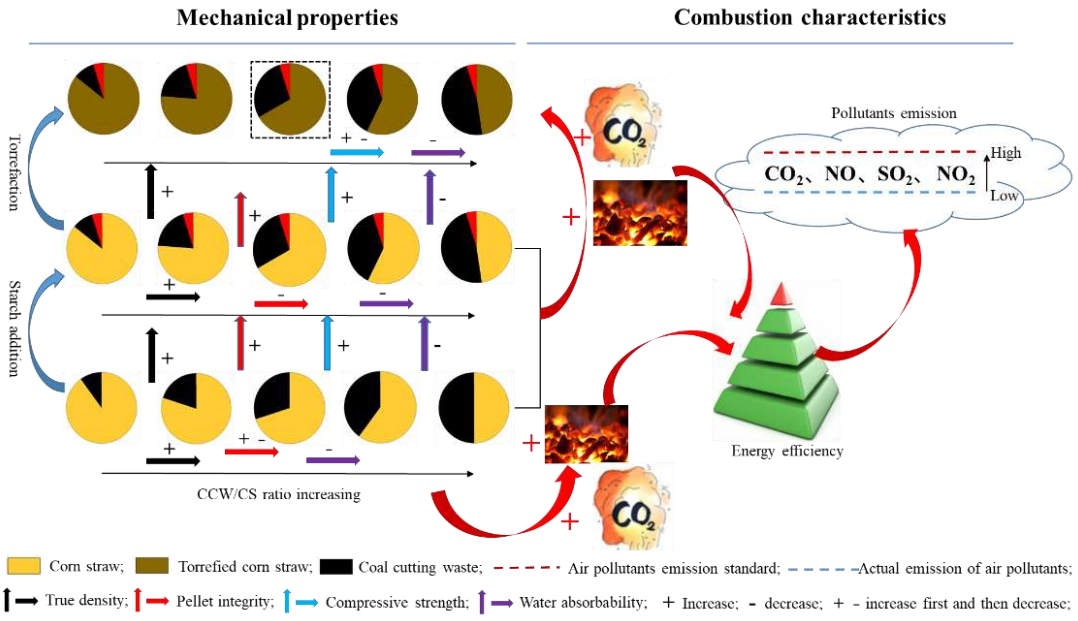


Fig. 16. The variation of mechanical and combustion characteristics of upgraded argo-pellets under different processing conditions

Considering that biomass itself belongs to clean energy, the blending of biomass and CCW to upgrade the quality of argo-pellets is still a feasible scheme on the premise of meeting the emission standards of air pollutants. However, in some places with high requirements for pollutant emission, the combination of torrefaction treatment and reduced proportion of CCW blending needs to be adopted. Based on the mechanical properties and combustion characteristic parameters of argo-pellets, the proportion

of CCW blending recommended in this study is 30%. For different sources of agricultural and coal waste, the recommended blending proportion may be changed. Meanwhile, the emission standards of air pollutants are often aimed at the emission of biomass or coal, and lack of relevant blending pellets. Therefore, future studies need to pay more attention to the variation of raw materials and the formulation of pollutant emission standards. In addition, LCA study in this work does not consider the main pollutant emission (CO_2) during the whole process, and the corresponding environmental impact analysis needs to be added in the follow-up, so as to analyse the pollutant emission in the whole process more comprehensively. CS and CCW are two different forms of waste; and upgraded argo-pellets show a good market prospect. The resource combined utilisation of these two wastes is a promising method with high economic benefits.

4. Conclusions

This work aimed to improve the quality of argo-pellets by blending with CCW, torrefaction treatment and adding starch binder. On the basis of the obtained results and discussion, the main conclusions can be drawn as:

- Mechanical experiments showed that co-pelleting of TCS and CCW with starch binder was technically possible. The qualities of the pellet samples including true density, pellet integrity, compressive strength and water absorbability have been improved to varying degrees.
- Thermogravimetric analysis revealed that the appropriate proportion of less CCW can not only promote the combustion of blending pellets, but also improve the thermodynamic characteristic parameters of torrefied pellet. Compared with CCW blending, torrefaction treatment can more effectively smooth the DTA curves, which improves the availability for the boiler in operation.
- Both CCW blending and torrefaction can reduce the concentration of NO_x and SO_2, improve the calorific value of fuel, but increase the CO_2 emission per unit mass. Comprehensive considering the increase of calorific value and CO_2 emission, co-combustion and torrefaction are conducive to pollutants

712 reduction and improve the quality of raw materials

713 • LCA analysis indicated that increasing the blending ratio of CCW and
714 torrefaction pre-treatment requires more energy input, but the energy
715 efficiency EER of pellet fuel will also be improved.

716

717 **Declaration of competing interest**

718 The author declares no competing financial interest.

719

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727

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