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Combustion behavior and pollutant emissions of the pressurized fluidized bed oxy-fuel co-firing of coal and poultry manure

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ABSTRACT

Pressurized oxy-fuel combustion is a next-generation and low-cost carbon capture technology with industrial application potential. This work presents an innovative research exploration—coupling coal pressurized fluidized bed oxy-fuel combustion technology with energy utilization of poultry manure as a renewable and carbon-neutral fuel, in order to capture CO_2 and solve the problem of poultry manure treatment simultaneously. In this study, a stable co-combustion of coal and chicken manure in a laboratory-scale pressurized fluidized bed under typical oxy-fuel condition (30%O_2/70%CO_2, *i.e.*, Oxy-30) is achieved. The key parameters including the combustion pressure (0.1–0.5 MPa) and chicken-manure proportion (0% to 100%) and their impacts on fundamental combustion efficiency, carbon conversion, nitrogen and sulfur pollutant emissions, and residue ash characteristics have been investigated. The result show that pressurization favors an increase in the CO_2 enrichment concentration and fluidized bed combustion efficiency. During co-combustion under 0.1 and 0.3 MPa, the CO_2 concentration in the flue gas is the highest when the chicken manure blending ratio (M_{pm}) is 25%. Although the NO emissions fluctuate and even increase as M_{pm} increases, the co-combustion of coal and chicken manure exhibits a synergistic effect in reducing NO conversion rate (X_{NO}). The effect of pressurization on reducing NO emission is significant, X_{NO} at $M_{pm} = 25\%$ decreasing from 15% to 5% as the pressure (P) increases from 0.1 to 0.5 MPa. As P increases from 0.1 to 0.5 MPa and M_{pm} increases from 0% to 50%, the SO_2 emissions and conversion rates decrease. The self-desulfurization process plays an important role in the reduction of SO_2 emissions during pressurized oxy-fuel co-combustion. The aim of this work is to advance the development and application of pressurized fluidized bed oxy-fuel co-combustion technology and promote a circular bioeconomy and carbon-free waste management for biomass derived from livestock manure.

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1. Introduction

Pressurized oxy-fuel combustion is a next-generation, energy-efficient CO_2 capture technology in combustion and also considered as one of the most promising carbon-capture solutions for coal-fired power plants with industrial prospects [1]. This technology improves system efficiency by increasing combustion pressure to reduce energy losses due to pressure fluctuations and recover the latent heat of steam from exhaust gases [2]. Additionally, the reaction kinetics advantages brought by pressurized combustion help reduce the size of the infrastructure, ultimately lowering the costs involved in carbon capture. Consequently, pressurized oxy-fuel combustion has become a key area of research on carbon capture for both academia and industry. Current research efforts on pressurized oxy-fuel combustion are still in their early stages [3], and researchers continue to explore future developments. One significant research direction is pressurized oxy-fuel co-combustion of coal with biomass, which can enable the transition from zero-

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carbon to negative-carbon emissions. For example, Symonds *et al.* [1] from CanmetENERGY evaluated the use of biomass and high-sulfur coal as feedstock for 100 MW (thermal) pressurized bubbling- and circulating-fluidized bed oxy-fuel combustion systems. Research indicated that the shift from coal to biomass had only a minimal impact on the total recoverable heat from power generation but significantly reduced power and oxygen requirements. Compared with pulverized coal-fired boilers, fluidized bed boilers offer greater fuel flexibility and improved heat transfer characteristics; thus, these are more suitable for pressurized oxy-fuel combustion systems, particularly for multi-fuel co-combustion, and are attracting wider attention.

Livestock and poultry manure is a highly valuable biomass resource and an unavoidable by-product of animal farming. Owing to the rapid development of intensive animal farming worldwide, the management and utilization of the increasing volumes of livestock manure result in both opportunities and challenges. According to FAOSTAT (2019), the European Union produced 1.389 billion tons of livestock manure in 2016, including 171241 tons of swine manure, 1090700 tons of cow manure, and 127317 tons of chicken manure [4]; more than 90% of this amount was directly applied as organic fertilizers in soil [5]. However, livestock manure releases greenhouse gases such as NH_3, N_2O, and CH_4, whereas nitrogen- and phosphorous-containing substances increase nutrient levels in soil and water bodies, disrupting ecosystem structures. Additionally, livestock manure contains pathogens, including *Escherichia coli*, *Streptococcus suis*, and *Salmonella* [6], which carry the risk of contagious diseases. Inappropriate handling of livestock manure can damage the environment and pose risks to human health; thus, traditional non-energy treatment methods, such as direct application to farmland, are becoming increasingly undesirable. However, from another perspective, livestock manure has potential as a carbon-neutral fuel and can be used for the sustainable generation of renewable energy, rather than merely being treated as waste for disposal.

In recent years, thermal conversion technologies have been recognized as one of the effective options for energy utilization of livestock manure [7], as they can produce high-value products such as heat, electricity, gaseous fuels, and biochar, thereby promoting a circular bioeconomy and carbon-free waste management. Research work on the techno-economic feasibility and environmental effects related to the energy utilization of livestock manure has been conducted successively. Billen *et al.* [8] compared the environmental impact of using poultry manure for fluidized bed combustion power generation with its direct application to soil. They concluded that poultry manure used for fluidized bed combustion power generation can replace fossil fuels, reduce greenhouse gas emissions and emissions of NH_3, NO_x, and N_2O, making it a more effective method. Choudhury *et al.* [9] conducted a quantitative analysis of the environmental impact of poultry manure combustion for power generation. Their research revealed that the ash from livestock manure combustion or co-combustion had the potential to replace conventional phosphorus-based fertilizers. Compared to untreated poultry manure, this ash had lower transportation costs and a reduced water-soluble inorganic phosphorus concentration. More importantly, the high-temperature operating conditions destroyed pathogens and microorganisms, eliminating the potential risk of disease transmission. In summary, from a technical and environmental standpoint, combustion of livestock manure—particularly co-combustion with coal—has been recognized as the most effective route for livestock manure-based power generation with the greatest industrial application potential [6,8,10], offering reasonable investment costs, controllable emissions, and high efficiency. As livestock manure has high water and ash concentrations, combusting it alone presents inherent challenges such as low stability and a low heating value. Therefore, co-combustion with fossil fuels or plant-based biomass is a logical and effective solution [6].

To date, compared to systematic theoretical analysis, practical studies on the use of livestock manure/coal mixtures in fluidized co-combustion—whether through mechanistic studies or process testing—remain limited [7]. A few small-scale or laboratory-scale co-combustion experiments have been conducted, primarily focusing on static-state or single-feed experimental setups, such as thermogravimetric analyses [4,11–15]. Research on coal and livestock manure co-combustion under oxy-fuel conditions remains scarce. The application and practical experience of pressurized fluidized bed oxy-fuel combustion are also still very limited globally. López-González *et al.* [11] and Zhu *et al.* [12] respectively conducted experimental studies on the oxy-fuel combustion of swine manure and the oxy-fuel co-combustion of pulverized coal and cow manure using thermogravimetric reactors, providing valuable references for a deeper understanding of the oxy-fuel combustion mechanism of livestock manure. Wzorek *et al.* [4], Turzyński *et al.* [13], Zhang *et al.* [14], and Atimtay *et al.* [15] have also studied the co-combustion of animal manure with fossil fuels or plant-based biomass in the air condition using either a thermogravimetric analyzer or a fixed-bed setup, focusing primarily on combustion kinetics and solid ash analysis. In terms of conventional fluidized bed air combustion, Jia and Anthony [16] tested the co-firing of poultry-derived fuel (PDF) with coal in a 0.8 MW (thermal) circulating-fluidized-bed combustor at CanmetENERGY. The test produced promising results, supporting that co-firing with coal in existing circulating fluidized bed boilers is a good way to utilize PDF. Varol and Gürel *et al.* [17,18] conducted combustion experiments with a mixture of chicken manure (CM) and Turkish lignite in a 350 kW·h circulating fluidized bed, and found that different fuel mixing ratios had no significant effect on HCl emissions, while polyaromatic hydrocarbons (PAHs) emissions increased with the increase of the CM proportion when the CM proportion was more than 50% by mass.

However, direct combustion or co-combustion of chicken manure may pose corresponding challenges in terms of ash behavior and pollutant emissions during boiler operation. For example, Lynch *et al.* [19] studied the ash agglomeration and deposition during combustion of chicken manure in a bubbling fluidized bed combustor, highlighting the role of K-silicates in initiating bed agglomeration, and the high KCl content in the fine baghouse ash. The ash from poultry manure combustion usually contains high levels of alkaline elements, especially K and Na, which can lead to the formation of low melting point eutectic compounds, resulting in slagging and fouling on the heating surface [20]. Katsaros *et al.* [10] pointed out that the higher concentrations of N, Cl, and S in chicken manure compared to wood chips and softwood pellets may cause emission problems during combustion. Moreover, the maximum aerosol emissions during chicken manure combustion were observed, which is closely related to ash deposition and corrosion in the heat exchanger, making it necessary to have proper dust cleaning equipment. But in their study, the ash melting temperatures of chicken manure and mixtures were estimated to be high. By analyzing the mineral composition of chicken manure, Ozdemir *et al.* [21] also concluded that chicken manure had the potential for slagging and fouling, and that co-combustion may be a more sustainable way of waste management and energy recovery.

To the best of our knowledge, no studies on the co-combustion of coal and livestock manure using pressurized fluidized bed oxy-fuel combustion technology have been conducted to date. This topic is an emerging and intriguing research direction. For such a complex, multicomponent, multiphase, pressurized, and high

temperature reaction system, a series of basic and key issues need to be systematically and deeply studied, including the continuous operation regulation and process optimization of fluidized bed pressurized oxy-fuel co-combustion (such as the stabilization of combustion state, optimization and matching of key operating parameters, and improvement of combustion performance and efficiency), emission patterns and transformation mechanisms of gas–solid pollutants in the process of pressurized co-combustion, and phenomena related to ash slagging, fouling, and agglomeration, as well as the synergistic effects between different parameters. Therefore, this study presents a new method for the energy utilization of livestock manure, which is to synergistically treat poultry manure in a coal-pressurized fluidized oxy-fuel combustion process, and achieve a stable co-combustion of coal and chicken manure in a laboratory-scale pressurized fluidized bed under typical oxy-fuel condition (30%O_2/70%CO_2, *i.e.*, Oxy-30). Key parameters including the combustion pressure (0.1–0.5 MPa) and chicken-manure proportion (0% to 100%) and their impacts on fundamental combustion efficiency, carbon conversion, nitrogen and sulfur pollutant emissions, and residue ash characteristics have been investigated. The aim of this work is to advance the application of pressurized fluidized bed oxy-fuel combustion for the treatment of a broad variety of fuel sources, such as biomass derived from livestock manure.

2. Experimental

2.1. Pressurized fluidized-bed oxy-fuel combustion system

The small-scale pressurized fluidized-bed oxy-fuel combustion system setup is shown in Fig. 1. The system consisted of five key units: small-scale fluidized bed reactor, feeding system, gas supply system, fly ash collector, and flue gas analyzer. The fluidized bed reactor was designed to operate at a maximum pressure of 1.0 MPa, and a maximum temperature of 1000 °C. The reactor had a length of 1300 mm, with internal and external diameters of 40 and 80 mm, respectively. High-pressure-resistant gas-tight flange connections were installed at the top and bottom of the reactor to ensure secure sealing. The fluidized bed was electrically heated by employing a three-segment silicon carbide rod, with external insulation provided by a nanoceramic material.

The gas supply system can provide various oxy-fuel atmospheres by using O_2 and CO_2 gas cylinders, with flow rates precisely controlled by mass flow controllers. The gases were mixed in a gas chamber and then passed through a gas preheater, where the temperature was raised to 600 °C before the gas mixture was introduced into the fluidized bed. Additionally, a dedicated CO_2 was used as the pressurizing gas for the feeder silo (regulated by a fine-tuning valve) to ensure the continuous and stable feeding of feedstock under pressurized conditions. The feeding system consisted of a feed tank, a frequency conversion motor, a screw feeder, and other components. The feed tank used a closed structure with a pressure-balance gas line. Fly ash particles in the flue gas were collected using a filter dust removal tank, which was composed of an external tank body, a metal mesh filter, and an internal hollow tube. The filtered flue gas was then passed through anhydrous calcium chloride to remove moisture and then purified and directed to the flue gas analyzer or vented if necessary. The internal pressure of the fluidized bed was regulated using a backpressure regulator.

2.2. Fuel properties

Bituminous coal, sourced from Datong, Shanxi, and chicken manure, obtained from a large breeding farm in Anhui, were used as fuel materials. One of the key challenges in using animal manure as fuel is the preparation and feeding. Fresh chicken manure typically has a high-water concentration and irregular shapes and sizes,

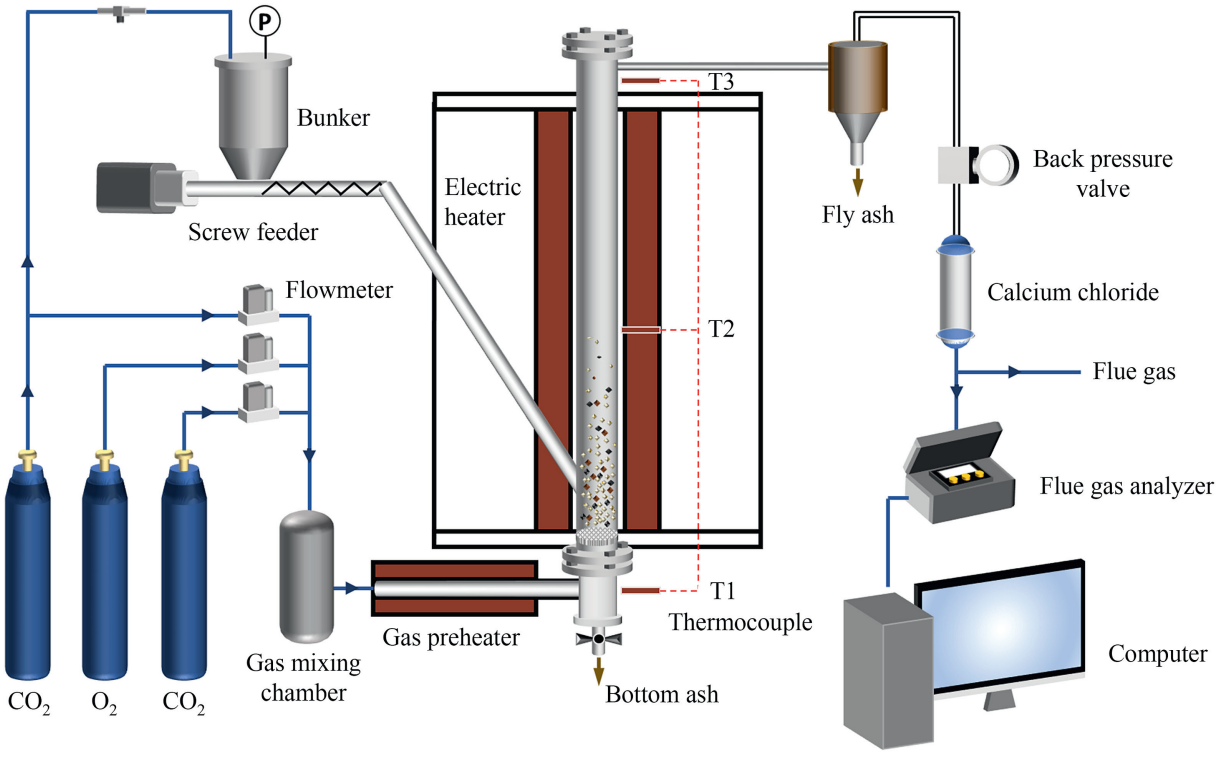

Fig. 1. Schematic diagram of the pressurized fluidized-bed oxy-fuel combustion system.

which is a mixture of excretions, bedding materials, animal feed, and feathers. Therefore, the chicken manure was first dried naturally (Fig. 2(a)) and then dried in an oven at 105 °C for 2 h. The dried samples were then crushed and sieved to obtain particles having a size range of 1.0–1.6 mm (Fig. 2(b)). Bituminous coal was treated using the same method and sieved to a particle size range of 0.6–2 mm (Fig. 2(c)). The dried chicken manure had a true density of 1651.84 $kg \cdot m^{-3}$, whereas the bituminous coal particles had a true density of 1794.39 $kg \cdot m^{-3}$; thus, both materials had similar densities. Quartz sand was used as the fluidizing medium with a particle size range of 0.3–0.4 mm, ensuring a fluidization number of 3–4 and a bubbling fluidization state when operating the fluidized bed.

Table 1 presents the results of the proximate analysis, ultimate analysis, and calorific analysis of coal and dried chicken manure. It can be seen that the moisture amount in the treated chicken manure decreases to below 10%; thus, the treated chicken manure is more suitable for the fluidized bed combustion process. Katsaros *et al.* [10] also recommended a moisture concentration of <10% (mass) to achieve effective poultry litter combustion. Compared with coal, chicken manure has a much higher volatile matter concentration but a lower fixed carbon content. This indicates that the combustion of chicken manure occurs predominantly in the gas phase, which may substantially affect the combustion performance of a pressurized fluidized bed. The ultimate analysis reveals that the carbon content of chicken manure is lower than that of coal, resulting in a much lower calorific value—approximately half that of coal. Additionally, chicken manure has a higher nitrogen concentration but a lower sulfur concentration than coal. However, compared to plant-based biomass such as rice husk and wood, both nitrogen and sulfur contents of chicken manure are higher [10,13].

2.3. Experimental process

A series of pressurized fluidized-bed oxy-fuel co-combustion experiments were conducted under a typical oxy-fuel atmosphere (Oxy-30), with chicken manure and coal mass mixture ratios of 0/100, 25/75, 50/50, 75/25, and 100/0 (determined on a mass basis). The combustion pressure ranged from 0.1 to 0.5 MPa, with the combustion temperature maintained at 800 °C and excess oxygen coefficient controlled between 1.20 and 1.30. Under atmospheric pressure condition, the feed rate of fuel was between 0.3 and 0.8 g min^{-1} (The feeding rate increased gradually as the blending ratio of chicken manure increased, because the calorific value of chicken manure was lower than that of coal). As the combustion pressure increased, the gas intake and fuel feed rate were adjusted proportionally to maintain a consistent combustion environment across different pressures, ensuring comparable gas superficial velocity, residence time, and excess oxygen levels. In the design and operation of the combustion system, attention should be paid to the adaptability of the feeder to the increase in feed rate under pressurized conditions and to the complexity and highly variable shape of the chicken manure particles. The combustion duration was maintained at approximately 1 h for each operating condition, with each condition repeated twice to ensure the accuracy of the experimental data. Online analysis of the combustion gaseous products was performed using two flue gas analyzers: Nova Plus and GA-21 Plus. These analyzers monitored O_2 (%), CO_2 (%), CO (0.0001%), NO (0.0001%), and SO_2 (0.0001%). Solid products, including fly ash and bottom ash, were collected, weighed, and analyzed to determine their chemical composition using elemental analysis (EA) and X-ray fluorescence spectrometry (XRF).

Fig. 3(a) presents the real-time operating curve of a typical operating condition, from the start to the end of feeding, with a fuel mixture of 50% coal and 50% chicken manure (by mass), and Fig. 3(b) shows the mean and standard deviation of flue gas data. The combustion environment was Oxy-30, with a combustion pressure of 0.1 MPa. As shown, fluidized oxy-fuel co-combustion of coal and chicken manure could operate stably, even with a high chicken-manure proportion of up to 50%. The O_2 concentration in the flue gas was maintained at approximately 5.5%, corresponding to an excess oxygen coefficient of ~1.22. The CO_2 concentration was maintained at approximately 90%, meeting the requirement for a high CO_2 concentration in oxy-fuel combustion for CO_2 capture. The concentrations of the main gaseous pollutants, NO and SO_2, were also stabilized at 0.043% and 0.032%, respectively, which were within the typical concentration ranges of pollutant emissions from fluidized bed combustion [22,23]. This demonstrates that the combustion field was well organized during the experiment, and also preliminarily proves the feasibility of oxy-fuel co-combustion of coal and chicken manure in a fluidized bed.

3. Results and Discussion

3.1. Combustion characteristics

First, the carbon conversion and combustion efficiency during the pressurized oxy-fuel co-combustion of coal and chicken manure were discussed. Fig. 4 illustrates the variation in the CO_2 concentration in the flue gas (V_{CO_2}) with changes in the combustion pressure (P = 0.1–0.5 MPa) and chicken-manure proportion (M_{pm} = 0% to 100%). Although chicken manure was used as fuel and its carbon content was considerably lower than that of coal, when the co-combustion mass ratios of chicken manure were 25% and 50%, the CO_2 concentration in the flue gas was higher than that



(a) (b) (c)

Fig. 2. Photographs of the fuel particles: (a) chicken manure dried naturally, (b) chicken manure after mechanical drying and crushing, (c) bituminous coal.

Table 1
Characteristics of bituminous coal and chicken manure (air dried basis).

Sample	Ultimate analysis/% (mass)					Proximate analysis/% (mass)				Lower calorific value/kJ·kg^{-1}
	C	H	N	S	O	M	A	V	FC	
Bituminous coal	70.59	2.45	1.07	1.28	5.68	1.71	17.22	14.80	66.27	30053.15
Dried chicken manure	31.92	4.77	3.21	0.70	22.77	7.39	29.24	56.40	6.97	15139.98

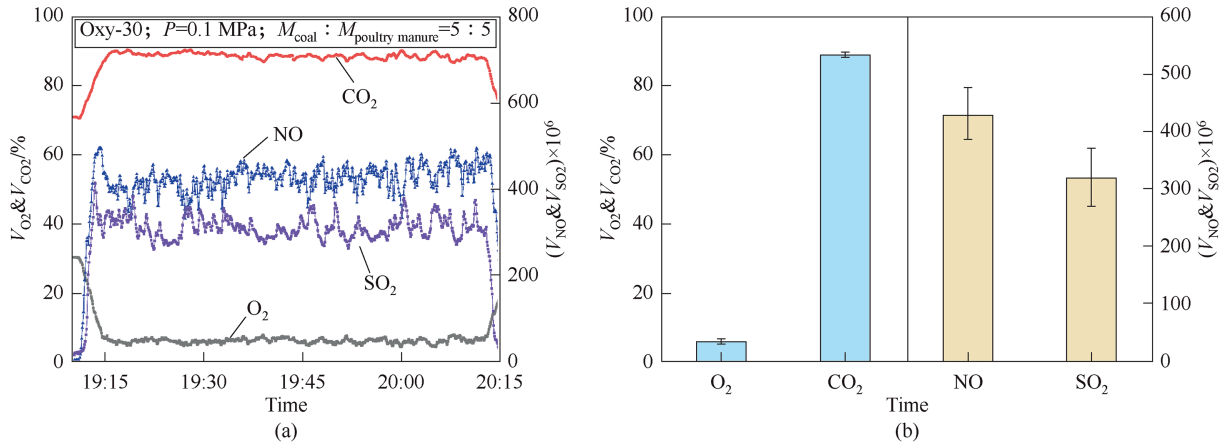

Fig. 3. (a) Real-time operation curve of Oxy-30 combustion with a fuel mixture of 50% coal and 50% chicken manure; (b) mean and standard deviation of the flue gas data.

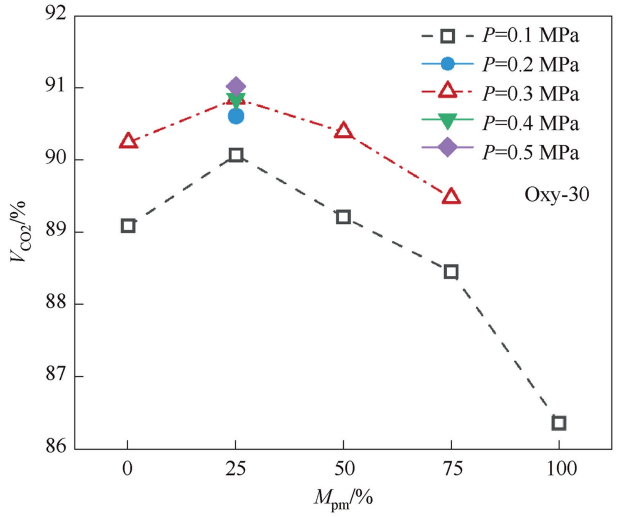

Fig. 4. Variation in the CO_2 concentration in the flue gas (V_{CO_2}) with changes in the combustion pressure (P) and chicken-manure proportion (M_{pm}).

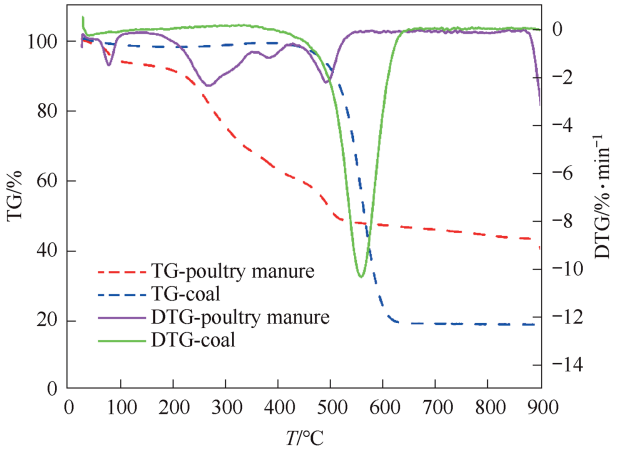

Fig. 5. Thermogravimetric and derivative thermogravimetric curves (TG and DTG, ramp rate: 10 °C·min^{-1}) for bituminous coal and chicken manure.

observed during coal-only combustion. This is because the high volatility and high reactivity of chicken manure enhance the combustion performance of coal and promote the conversion of carbon in the fuel to the final product CO_2. As shown in Table 1, the volatile-matter concentration of chicken manure is nearly 60%, approximately four times that of coal. Fig. 5 depicts that under oxy-fuel condition (Oxy-30), the thermogravimetric and derivative thermogravimetric curves (TG and DTG, ramp rate: 10 °C·min^{-1}) for bituminous coal and chicken manure, when combusted individually, exhibit distinct characteristics. It can be seen that in the Oxy-30, the coal displays a relatively broad mass-loss peak in the high-temperature range (400–700 °C), indicating the combustion of a small amount of volatile matter and a large amount of fixed carbon. In contrast, chicken manure exhibits multiple weight-loss peaks over a broader temperature range (150–600 °C).

The low-temperature region (150–430 °C) is the primary weight-loss range of chicken manure, which is mainly attributed to the decomposition of cellulose, hemicellulose, and lignin to release volatile substances [14,15,24]. Cellulose, hemicellulose, and lignin are the main structural components of lignocellulosic biomass. In general, the thermal decomposition process of animal manure is similar to that of lignocellulosic biomass [4]. In addition to hemicellulose, cellulose, and lignin, animal manure also contains proteins, starches, lipids, sugars, *etc.*, due to bacteria and chemical reactivity along the digestive tract. According to the available research data [25], in chicken manure samples, cellulose content (%, dry mass) was 14.49% ± 0.60%, hemicellulose content was 21.66% ± 0.22%, and lignin content was 7.83% ± 0.12%. The presence of these components makes chicken manure have the potential for

energy utilization. During the co-combustion of coal and biomass, the volatiles in the chicken manure are rapidly released and combusted, promoting the combustion of the blended fuel.

However, when the chicken-manure proportion was increased to 75% and 100%, the CO_2 concentration in the flue gas decreased significantly, especially when 100% chicken manure was combusted and the CO_2 concentration dropped to only 85%. This reflection may be attributed to the low carbon content of chicken manure itself, or partly related to the deterioration of the combustion field, which is unfavorable for carbon conversion (see analyses in Figs. 6 and 7). For fluidized bed combustion, the organization of a good combustion field relies on the stable gas-solid flow field, temperature field, composition field, and the cooperation between them. As stated above, due to the high alkali metal content of chicken manure, problems of ash or sand agglomeration may arise, thus affecting the fluidization quality, interfering with the gas-solid flow field as well as causing changes in the temperature field (*e.g.*, localized overheating and uneven temperature distribution). Additionally, as shown in Fig. 4, the CO_2 concentration increased with an increasing combustion pressure. When the pressure was raised from 0.1 to 0.2 MPa, the increase in the CO_2 concentration was particularly pronounced. The increase in the pressure also increases the concentration of the reactants, which, in turn, promotes fuel combustion and carbon conversion. During co-combustion at 0.1 and 0.3 MPa, the CO_2 concentration in the flue gas was the highest when the chicken-manure proportion is 25%.

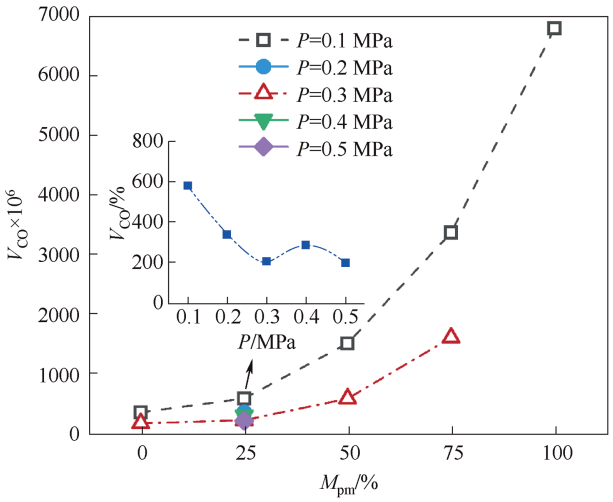

Fig. 6. Variation in the CO concentration in the flue gas (V_{CO}) with changes in the combustion pressure (P) and chicken-manure proportion (M_{pm}).

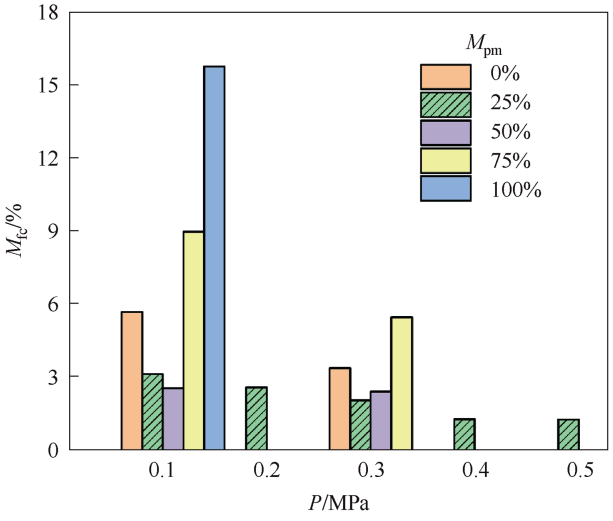

Fig. 7. Carbon concentration in the fly ash (M_{fc}) during oxy-fuel combustion at various pressures and chicken-manure proportions.

Subsequently, the carbon products of incomplete combustion were examined, including the flammable CO gas in the flue gas and the carbon residue in the ash. Fig. 6 shows the variation in the CO concentration in the flue gas (V_{CO}) with changes in the combustion pressure (P = 0.1–0.5 MPa) and chicken-manure proportion (M_{pm} = 0% to 100%). As observed, with the increase of chicken manure blending ratio, even the excess oxygen coefficient remained constant, the CO emission concentration notably increased, particularly when M_{pm} exceeded 50%. One possible reason for this phenomenon is the rapid release of a large amount of volatiles from the chicken manure, which may cause localized oxygen deficiency in the fluidized bed, thereby hindering the oxidation of CO. Moreover, the high alkali and alkaline earth metals content properties of chicken manure may lead to the deterioration of the fluidization state in the fluidized bed, thereby affecting the combustion field organization.

However, when the combustion pressure increased from 0.1 to 0.3 MPa at M_{pm} = 25%, the CO concentration decreased. When the pressure was further increased from 0.3 to 0.5 MPa, the CO concentration fluctuated. On one hand, an increase in the pressure reduces the release of volatiles but also increases the reaction rate, promoting the oxidation of CO to CO_2 [26]. This is the promoting effect brought about by the pressurization itself. On the other hand, as the combustion pressure increased, the fluidizing gas intake and fuel feed rate were increased proportionally, which may cause more carbon particles to accumulate in the fluidized bed, resulting in an increase in CO concentration. This effect is a result of the operational parameters associated with pressurization. Thus, when the pressure was increased, fluctuations in CO concentration were observed. Pang *et al.* [27] and Liu *et al.* [3] reported an opposite CO trend with varied pressure and attributed the trend to differences in the pressurization methods used in their fluidized bed systems. Although pressurized combustion can increase fuel processing capacity and heat load without the reactor size being changed, the observed increase in CO concentration suggests that the upper limit of the pressurization should be controlled appropriately to ensure sufficient combustion. This limitation, however, depends on the design and scale of the pressurized reactor.

Fig. 7 shows the carbon concentration in the fly ash (M_{fc}) during combustion at various pressures and chicken-manure proportions. The unburned carbon content in the bottom ash was found to be very low (less than 0.3%). The carbon concentration in the fly ash gradually decreased with an increasing pressure, suggesting that pressurization promotes the burnout of fuel carbon. However, when the pressure exceeded 0.4 MPa, the improvement associated with an increasing pressure diminished. At a combustion pressure of 0.1 MPa, increasing the chicken-manure proportion from 0% to 50% resulted in a decrease in M_{fc}, but M_{fc} significantly increased when the chicken-manure proportion was increased from 50% to 100%. The same phenomenon was observed at a combustion pressure of 0.3 MPa. The possible reason for the increase in M_{fc} is that compared with coal particles, chicken manure fuel has a higher volatile-matter concentration and a looser structure. Consequently, when a large amount of volatile substances is released from the chicken manure, the remaining unburned char particles are easily carried out of the fluidized bed with the flue gas before they can be completely combusted.

Furthermore, based on the concentrations of the combustible gas CO and the unburned carbon in the ash residue, the combustion

efficiency of the small-scale fluidized bed reactor was calculated using the heat loss method. During the experiment, the concentrations of H_2 and CH_4 in the flue gas were always lower than 0.01%, so most of the unburned gas-phase losses were attributed to CO in the flue gas. The unburned carbon concentration in the bottom ash was measured to be very low (less than 0.3%); therefore, the unburned solid-phase losses mainly originated from the unburned carbon in the fly ash. The calculation method of the combustion efficiency (η) is as follows. Where q_{CO} and q_{fc} denote the calorific value of CO and carbon in fly ash, respectively, and q_{in} is the heat input of the fuel.

$$\eta = 1 - \frac{q_{CO}}{q_{in}} - \frac{q_{fc}}{q_{in}}$$

As shown in Fig. 8, as combustion pressure increased, the fluidized bed combustion efficiency increased accordingly. Especially when the combustion efficiency was lower at atmospheric pressure, such as $P = 0.1$ MPa and $M_{pm} = 75\%$, the effect of pressurization on improving combustion efficiency was more significant. Although the CO concentration increased with the increase of the chicken-manure blending ratio from 0% to 50%, the unburned carbon in the ash decreased. Therefore, the combustion efficiency was the highest when $M_{pm} = 25\%$ under the pressure of 0.1 MPa and 0.3 MPa. The combustion efficiency of pure chicken manure was lower than 90%. So, this indicates that co-combustion is more suitable for the thermal treatment of chicken manure rather than single combustion.

3.2. Nitrogen pollutants

The current experimental research on chicken manure co-combustion is primarily focused on the combustion kinetic characteristics and solid ash residue analysis, with limited discussions on gaseous pollutants [11–15]. However, the emission characteristics of nitrogen and sulfur pollutants are critical considerations for the research and application of oxy-fuel combustion of coal and chicken manure in fluidized beds. As shown in Table 1, the nitrogen content of chicken manure is three times higher than that of coal. This is a key distinction between livestock manure-based biomass and traditional agroforestry-based biomass, because agricultural and forestry biomass typically has a lower nitrogen concentration. The elevated nitrogen content in livestock manure results from the presence of proteins, amino acids, uric acid, and other organic compounds. Fig. 9 shows the variation of NO volumetric concentration in the flue gas (V_{NO}) with the combustion pressure ($P = 0.1$–0.5 MPa) and chicken-manure proportion ($M_{pm} = 0\%$ to 100%). At 0.1 MPa, the NO concentration fluctuated when the chicken-manure proportion was relatively low (below 50%), and then gradually increased as M_{pm} increased from 50% to 100%. In contrast, at 0.3 MPa, the NO concentration increased when M_{pm} was increased from 0% to 25%, while the NO concentration decreased slightly or remained relatively stable when M_{pm} increased from 25% to 75%. Additionally, as the combustion pressure increased, NO emissions visibly decreased.

Further, the NO volumetric concentration emissions were converted to the mass of NO produced per unit heat input of the fuel and the NO yields (m_{NO}, mg·MJ^{-1}) were calculated, as shown in Fig. 10. At 0.1 MPa, when M_{pm} was increased from 0% to 50%, m_{NO} gradually decreased; however, when M_{pm} increased from 50% to 100%, m_{NO} increased significantly. Among them, the m_{NO} at $M_{pm} = 75\%$ was nearly identical to the yield of pure coal combustion. Under pressurized condition ($P = 0.3$ MPa), m_{NO} exhibited a

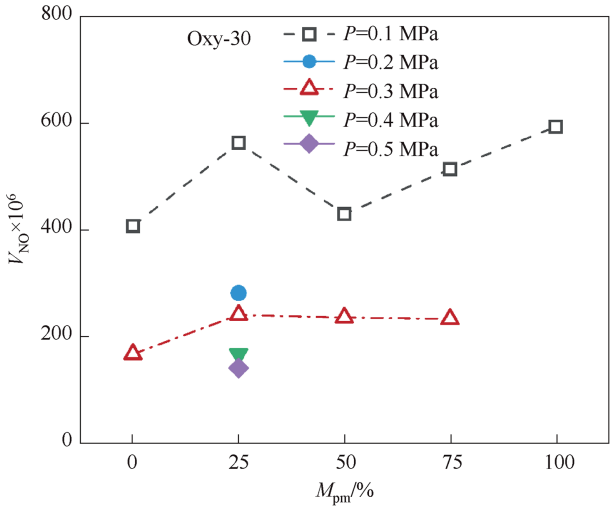

Fig. 9. Variation of NO volumetric concentration in the flue gas (V_{NO}) with the combustion pressure (P) and chicken-manure proportion (M_{pm}).

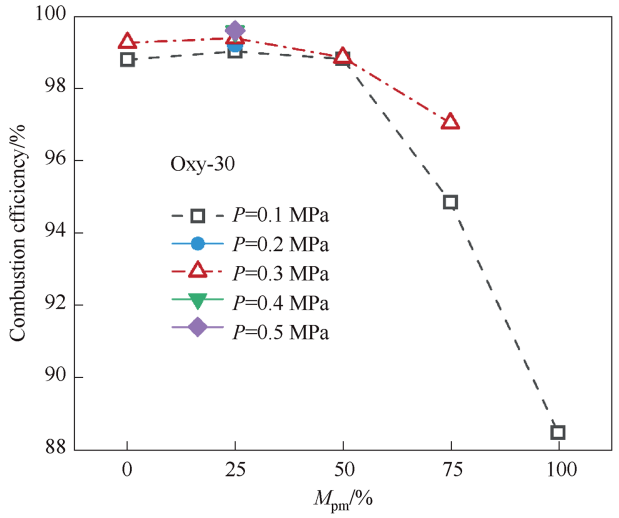

Fig. 8. Combustion efficiency of the small-scale fluidized bed reactor under different pressures and chicken-manure proportions.

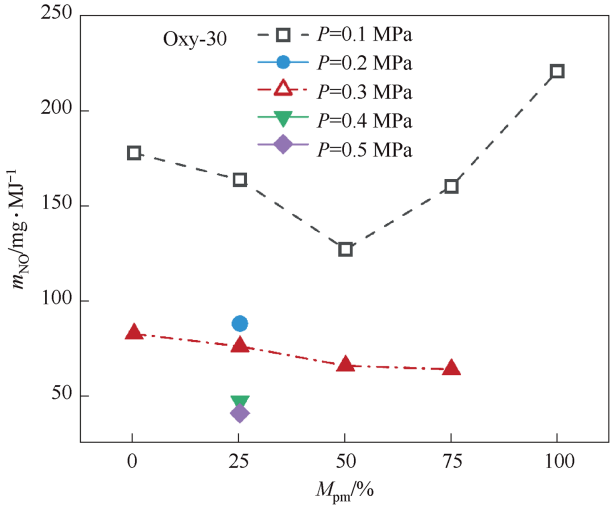

Fig. 10. Mass of NO produced per unit heat input of the fuel (NO yield, m_{NO}, mg·MJ^{-1}) at various pressures and chicken-manure proportions.

steady declining trend as M_{pm} increased from 0% to 75%. In addition, as the combustion pressure increased, the NO yield m_{NO} visibly decreased. The effect of pressurization on reducing NO emission is indisputable. The primary reason for this is that pressurization promotes heterogeneous reduction reactions of NO. The elevated pressure increases the resistance of the generated NO to diffuse out of the char particle, thereby slowing the rate at which NO leaves the carbon particle surface [28,29]. This extended residence time allows more NO to be reduced to N_2 *via* the heterogeneous reduction of NO by the char ($2NO + 2C \rightarrow N_2 + 2CO$). As discussed above, increasing the pressure tends to cause fluctuations in CO emissions, making CO-driven homogenous NO reduction unlikely to be the primary mechanism for the observed decrease in NO emission with increasing pressure.

Overall, under the combined influence of the pressure, fuel nitrogen content, and fuel combustion characteristics, the NO emissions from pressurized fluidized-bed oxy-fuel co-combustion did not follow a linear trend but instead exhibited a more complex behavior. The conversion rate from nitrogen in fuel to nitrogen in NO, denoted as X_{NO}, is presented in Fig. 11. As shown, the NO conversion rate under elevated pressure conditions was always lower than that under atmospheric conditions. For example, when $M_{pm} = 25\%$, X_{NO} decreased from approximately 13% to 3% as the pressure increased from 0.1 to 0.5 MPa. This also confirms the obvious effect of pressurization on NO emission reduction. However, it was also found that as the chicken-manure proportion increased, the difference in NO conversion rate between pressurized condition and atmospheric condition decreased. In other words, a high chicken-manure proportion reduced the effectiveness of increased pressure in suppressing the NO conversion rate, likely because the fixed carbon content in chicken manure is significantly lower than that of coal, which diminishes the extent of heterogeneous reduction of NO on char.

In addition, a surprising finding was that the NO conversion rate steadily decreased as M_{pm} increased, regardless of the operating pressure, although the difference in X_{NO} between $M_{pm} = 75\%$ and $M_{pm} = 100\%$ was very small at 0.1 MPa. Figs. 9 and 10 display that as M_{pm} increased, the NO concentration (V_{NO}) and emissions (m_{NO}) fluctuated and even increased, likely due to the nitrogen concentration in chicken manure being considerably higher than that in coal. However, in fact, the co-combustion of coal and chicken manure exhibited a synergistic effect in reducing NO conversion rate (X_{NO}), decreasing X_{NO} from nearly 25% when burning pure coal

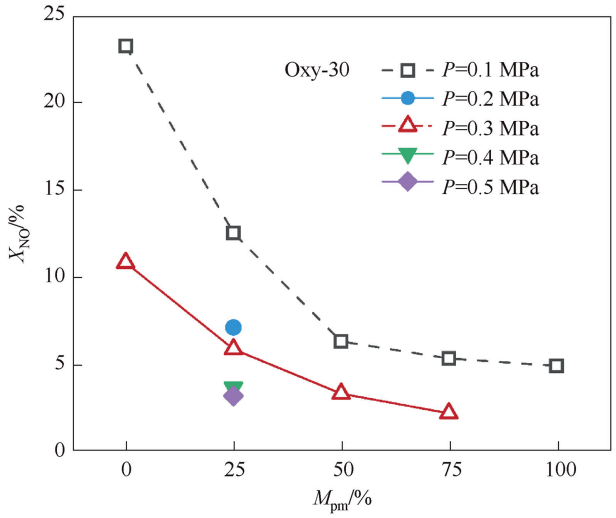

Fig. 11. Conversion rate from nitrogen in fuel to nitrogen in NO (X_{NO}) at various pressures and chicken-manure proportions.

to 5%. Possible reasons for this include the following: as the chicken-manure proportion increases, the CO concentration in the fluidized bed increases dramatically (as shown in Fig. 6), creating a more reducing environment that promotes the homogeneous reduction of NO by CO and the heterogeneous reduction of NO by char (as shown in R1 and R2). In addition, the nitrogen in biomass fuel is primarily released as NH_3, which can be oxidized to form NO (R3) but can also reduce NO to N_2 (R4) [30]. But HCN is the main product observed in the devolatilization of coal. Thus, as the chicken-manure proportion increased, the NO conversion rate decreased, which was mainly dependent on the enhanced NO reduction due to chicken manure properties and combustion characteristics. However, some studies [31,32] reported that the nitrogen in biomass, compared with coal, seemed to be released more readily. This may explain why the difference in X_{NO} between $M_{pm} = 75\%$ and $M_{pm} = 100\%$ was very small.

$$2NO + 2CO \rightarrow N_2 + 2CO_2 \quad (R1)$$

$$2NO + 2C \rightarrow N_2 + 2CO \quad (R2)$$

$$NH_3 + 1.25O_2 \rightarrow NO + 1.5H_2O \quad (R3)$$

$$NH_3 + 0.25O_2 + NO \rightarrow N_2 + 1.5H_2O \quad (R4)$$

3.3. Sulfur pollutants

Fig. 12(a, b) shows the SO_2 volumetric concentration in the flue gas (V_{SO_2}) and the mass of SO_2 produced per unit heat input of the fuel (SO_2 yield, m_{SO_2}, $mg \cdot MJ^{-1}$) under different combustion pressure ($P = 0.1$–0.5 MPa) and chicken-manure proportion ($M_{pm} = 0\%$ to 100%). As shown in the figure, with the chicken manure blending ratio increased from 0% to 75%, both the SO_2 concentration (V_{SO_2}) and emissions (m_{SO_2}) decreased. As indicated in Table 1, the sulfur content of chicken manure is lower than that of coal, which primarily explains the observed trend. Additionally, chicken manure contains a high concentration of alkali metals and alkaline earth metals (*e.g.*, Ca and K). During the fuel combustion process, especially in the char combustion stage, SO_2 released from fuel-sulfur diffuses outward through the pores of the char particles and undergoes sulfation reactions with Ca, K, and other alkaline compounds to produce sulfate (as per the reaction below), thus allowing sulfur to be effectively captured in the ash, also known as the self-desulfurization process [33,34]. Most of the self-desulfurization by ash occurs during char combustion rather than during devolatilization [35]. In fluidized bed combustion, the ash particles capture both SO_2 produced by parent fuel particles and SO_2 produced by other fuel particles [36], which has a significant impact on SO_2 emissions.

$$2CaO + 2SO_2 + O_2 \rightarrow 2CaSO_4$$

$$2CaCO_3 + 2SO_2 + O_2 \rightarrow 2CaSO_4 + 2CO_2$$

However, when the chicken-manure proportion was increased from 75% to 100%, SO_2 emissions increased slightly. A possible reason is that at a high chicken-manure proportion, the CO concentration in the fluidized bed increases significantly. The elevated CO concentration may cause the sulfation product $CaSO_4$ to undergo reductive decomposition, thereby compromising the self-desulfurization effect.

$$CaSO_4 + CO \rightarrow CaO + SO_2 + CO_2$$

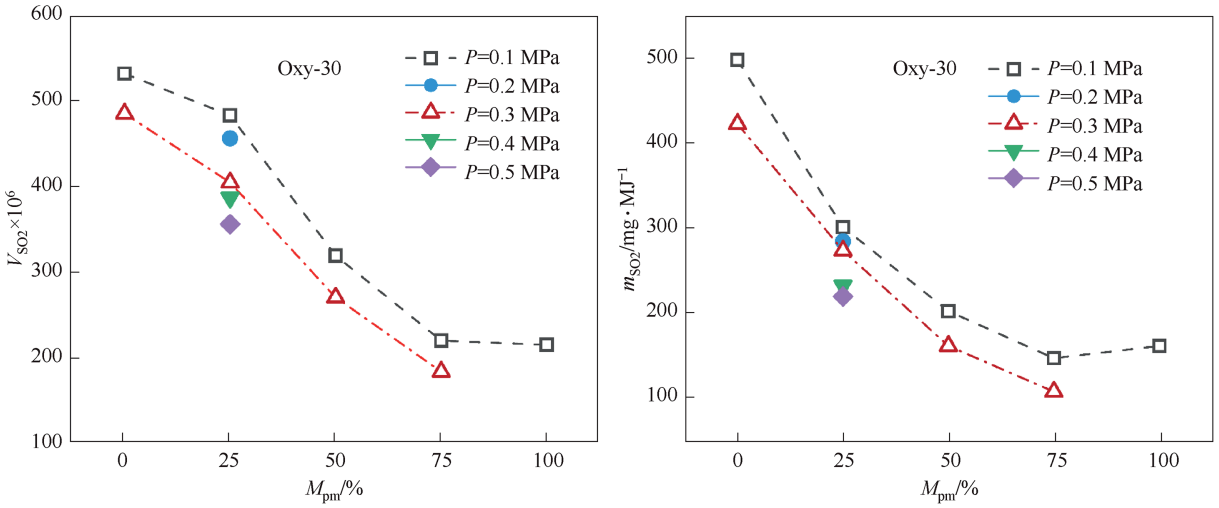

Fig. 12. (a) SO_2 volumetric concentration in the flue gas (V_{SO_2}) and (b) mass of SO_2 produced per unit heat input of the fuel (m_{SO2}, mg·MJ^{-1}) at various pressures and chicken-manure proportions.

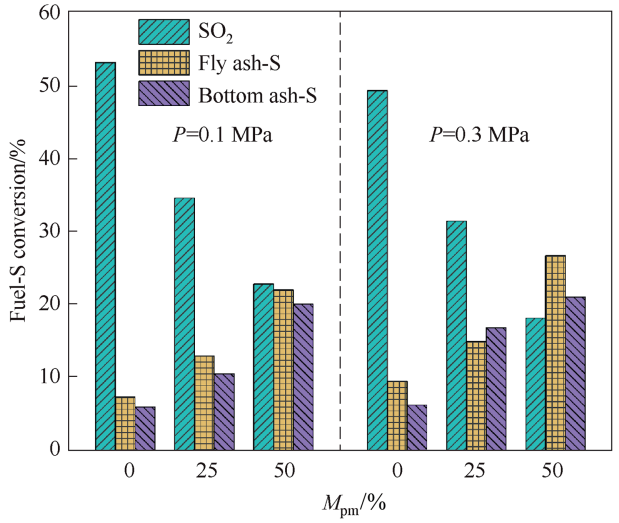

Fig. 13. Conversion rate of sulfur in the fuel to SO_2 (X_{SO_2}), sulfur in the fly ash (X_{FAS}), and sulfur in the bottom ash (X_{BAS}) at various pressures and chicken-manure proportions.

In addition, Fig. 12 reveal that increasing the combustion pressure decreased both the SO_2 volumetric concentration (V_{SO_2}) and the emissions (m_{SO_2}). The elevated pressure increases the resistance of the generated SO_2 to diffuse out of the char particle and prolongs the SO_2 residence time, thereby enhancing the self-desulfurization effect and ultimately reducing SO_2 emissions. Furthermore, the conversion rate of sulfur in the fuel to SO_2, to sulfur in the fly ash, and to sulfur in the bottom ash (denoted as X_{SO_2}, X_{FAS}, and X_{BAS}, respectively) were calculated and presented in Fig. 13. A comparison between 0.1 and 0.3 MPa reveals that even with different fuel mixtures, the conversion rate of SO_2 (X_{SO_2}) under elevated pressure condition was lower than that of atmospheric pressure condition, while the conversion rates to sulfur in the fly ash (X_{FAS}) and bottom ash (X_{BAS}) were higher than that of atmospheric pressure. Moreover, as the chicken-manure proportion increased from 0% to 50%, the conversion rate to SO_2 continued to decrease, whereas the conversion rate of sulfur to fly ash and bottom ash increased gradually. This demonstrates that the self-desulfurization process plays an important role in the reduction of SO_2 emissions during pressurized oxy-fuel co-combustion.

Self-desulfurization is affected by various factors that depend on fuel characteristics and combustion conditions [37]. From the

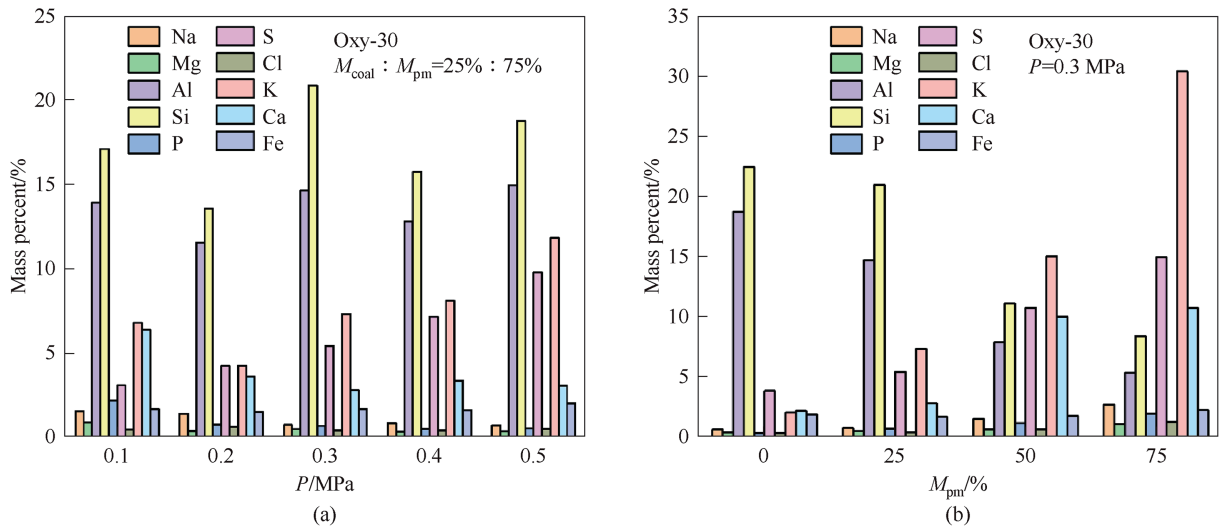

Fig. 14. Elemental composition of the fly ash during the pressurized oxy-fuel co-combustion process at (a) various pressures and (b) chicken-manure proportions.

perspective of fuel composition, the most important alkali oxide for self-desulfurization is CaO, which is formed by decomposition of $CaCO_3$ and combustion of Ca-containing organic matter [35]. From the perspective of operational parameters, the residence time of flue gas and ash particles, combustion pressure, and combustion temperature are important factors affecting the self-desulfurization effect. Previous studies have shown that self-desulfurization efficiency increases when the residence time is increased or when the combustion pressure is increased, either for coal combustion or coal-biomass co-combustion [36]. In addition, under pressurized oxy-fuel conditions, self-desulfurization may have two different pathways (*i.e.*, indirect sulfation and direct sulfation) [36]. Regarding the mechanism of self-desulfurization and the optimization of self-desulfurization efficiency need to be further explored in the future.

Besides, the elemental composition of the fly ash during the pressurized oxy-fuel co-combustion process was evaluated and the XRF results were depicted in Fig. 14. As shown in Fig. 14(a), with the increase of combustion pressure, the proportions of Na, Mg, and P in the ash decreased, and the S content increased, while the Al, Si, and Fe proportions exhibited no clear trend. The experimental results obtained by Duan *et al.* [38] on a pressurized coal combustion fluidized bed similarly showed that the pressure had no significant effect on the ash composition. It can be seen from Fig. 14(b) that when pure coal was combusted, the dominant elements in the fly ash were Si and Al. As the chicken-manure proportion increased, the concentrations of Na, P, S, Cl, K, and Ca increased. This confirms the enhanced self-desulfurization effect during co-combustion of coal and chicken manure. The most significant increase in the content of the element K with the increase of chicken manure proportion was observed. Firstly, chicken manure feedstock contains a large amount of potassium, which can typically be as high as 20% [10], directly contributing to the very high concentration of potassium in the fly ash. In addition, potassium is readily released into the gas phase during biomass combustion and forms low melting point compounds (*e.g.*, the K-salt particles), which sinter with the fly ash particles, thereby increasing the potassium concentration in the fly ash [39]. The ash produced from chicken manure combustion contained particularly high levels of P, Cl, K, and Ca. Although this composition makes the ash suitable for use as P and K fertilizers, it may also cause severe slagging, agglomeration, and corrosion during the combustion process.

Overall, for such a novel technology as pressurized fluidized bed oxy-fuel co-combustion of coal and chicken manure, the future development and scaling up are important issues worth exploring. Pure coal pressurized fluidized bed oxy-fuel combustion has been developed to pilot scale. For example, a 100 kW (thermal) pressurized circulating fluidized bed oxy-fuel combustion system was established at Southeast University in 2022 and operationally tested at a pressure of 0.6 MPa [40]. One advantage and feature of pressurized combustion is that it can increase the heat input and load of the reactor without changing the reactor size. However, this may lead to more serious ash problems such as slagging, corrosion, and agglomeration when it comes to scaling up pressurized fluidized bed oxy-fuel co-combustion of coal and chicken manure is scaled up. Experience with pilot-scale fluidized bed chicken manure combustion under conventional air conditions can be informative. For example, during the operation of a 37 MW poultry litter fired fluidized bed combustor located in the Netherlands [41], both in-bed agglomeration and severe agglomeration to the wall in the freeboard occurred occasionally, disturbing the fluidization and potentially causing complete de fluidization. It can be reasonably predicted that when pressurized oxy-fuel technology is coupled with chicken manure combustion, higher heat input may lead to more severe ash problems. Currently, some studies have proposed adding potassium feldspar or limestone into fluidized beds because potassium feldspar does not easily interact with the bed ash [42], and limestone provides calcium which reacts with phosphorus forming a high melting temperature calcium phosphate [43]. The ash handling and potential for slagging and corrosion under pressurized oxy-fuel combustion conditions need to be further explored in the future for better application of this technology.

4. Conclusions

In this work, experiments were carried out on the co-combustion of coal and chicken manure using pressurized fluidized bed oxy-fuel combustion technology. The key parameters including the combustion pressure (0.1–0.5 MPa) and chicken-manure proportion (0% to 100%) and their impacts on fundamental combustion efficiency, carbon conversion, nitrogen and sulfur pollutant emissions, and residue ash characteristics have been investigated. The main findings are as follows:

(1) A stable co-combustion of coal and chicken manure under typical oxy-fuel condition (Oxy-30) in a laboratory-scale pressurized fluidized bed was achieved. The pressurized conditions favored an increase in the CO_2 enrichment concentration and combustion efficiency. During co-combustion at 0.1 and 0.3 MPa, the CO_2 concentration in the flue gas was the highest when the chicken manure blending ratio was 25%.

(2) The CO concentration decreased when the combustion pressure (P) increased from 0.1 to 0.3 MPa, and then fluctuated as P further increased to 0.5 MPa. Increasing the chicken manure proportion (M_{pm}) from 0% to 50% resulted in a decrease in the carbon content in the ash (M_{fc}), but M_{fc} significantly increased when M_{pm} increased from 50% to 100%.

(3) As M_{pm} increased, the NO concentration (V_{NO}) and emission per unit heat input (m_{NO}) fluctuated and even increased. However, in fact, the co-combustion of coal and chicken manure exhibited a synergistic effect in reducing NO conversion rate. The effect of pressurization on reducing NO emission was significant, X_{NO} at $M_{pm} = 25\%$ decreasing from 15% to 5% as P increased from 0.1 to 0.5 MPa.

(4) As P increased from 0.1 to 0.5 MPa and M_{pm} increased from 0% to 50%, the conversion rate of fuel S to SO_2 decreased whereas the conversion rate to the sulfur in the fly ash and bottom ash increased gradually, which demonstrates that the self-desulfurization process plays an important role in the reduction of SO_2 emissions during pressurized oxy-fuel co-combustion.

CRediT Authorship Contribution Statement

Qinwen Liu: Writing – original draft. Yiwei Zhang: Investigation. Hengbing Ye: Investigation. Guanwen Zhou: Supervision. Yu Su: Supervision. Wenli Dong: Supervision. Chi-Hwa Wang: Supervision. Wenqi Zhong: Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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