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Pressurized oxy-fuel combustion of pulverized coal blended with wheat straw: Thermochemical characterization and synergetic effect



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ABSTRACT

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Keywords: Pressurized oxy-fuel combustion Coal/biomass co-combustion Thermogravimetric analysis Synergetic effect Pressurized oxy-fuel combustion is recognized as the second generation of oxy-fuel combustion technology that can facilitate carbon capture and reduce energy consumption, while coal and biomass combustion is also an effective way to decrease carbon emissions. This study innovatively integrates these two clean combustion technologies and investigates the effects of pressure and blending ratio on the combustion characteristics (ignition, combustion rate, volatile combustion, semi-coke combustion, burnout) of coal, biomass and their blends using an advanced magnetic levitation HPTGA. The results show that pressurization can improve the combustion rate of coal and biomass, but it will cause ignition delay when system pressure exceeds 0.5 MPa. The effect of pressure on combustion characteristics is both facilitative and inhibitive, in terms of reaction kinetics, pressurization can significantly promote combustion but it also plays a role in inhibiting oxygen diffusion, the competition between the two effect leads to an optimum value of pressure in the combustion performance of the blends, which is about 1 MPa. Biomass is more sensitive to pressure than coal, and pressurization changes the ignition mode of biomass. In addition, a new synergy index is defined to quantify the synergistic effect of the whole co-combustion process. Pressurization could suppress both catalytic and non-catalytic mechanisms in the co-combustion process, and different sensitivities of biomass and coal to pressurization lead to more significant differences in combustion time and temperature interval compared with normal pressure, all these factors contribute to negative synergistic effect under pressurization.

1. Introduction

In recent years, oxy-fuel combustion, which using pure oxygen separated from the air separation unit (ASU) and recycled flue gas (mainly CO₂) to replace the conventional air as the oxidizer, has been widely recognized as a competitive and advanced carbon capture technology [1,2]. However, in traditional oxy-fuel combustion systems, the ASU and CO₂ compression and purification unit (CPU) are carried out at high pressure, but the fuel combustion unit (pulverized coal boiler or fluidized bed boiler) is operated at atmospheric pressure, the unbalanced distribution of system pressure results in a large amount of energy loss [3,4]. To solve this problem, the pressurized oxy-fuel combustion (POFC) technology, also regarded as a new generation of oxy-fuel combustion, has gradually attracted the attention of both industry and academia. Boiler systems operating at high pressures can offer multiple advantages including limiting the boiler size, enhancing heat transfer and fuel mixing characteristics, maintaining of latent heat recovery potential of steam and reducing pollutant emissions [3,5,6]. Process simulations conducted by ASPEN Plus modeling show that compared with atmospheric oxygen fuel combustion, increasing the pressure can increase the net efficiency of the system by 3%–6% and significantly reduce and power cost [7–9].

Biomass is considered as one of the most promising green renewable energy sources in the 21st century due to its abundant resources, wide distribution, low emission of CO_2 and air pollutants [10,11]. However, the disadvantages of biomass, including low calorific value, high moisture content, and rich alkali metal and chlorine content, lead to the problem that direct combustion of pure biomass is prone to cause boiler burning instability and coking, corrosion of system. Therefore, co-firing with coal can improve the ignition and combustion performance of the fuel. Co-combustion provides the existing power plants with a rapid, low-risk, cost-effective biomass power generation technology. Extensive research has been conducted over the past few decades to determine the suitability of coal/biomass blends for diverse thermochemical conversion processes [12,13]. Tan et al. [14] initially proposed that utilize oxy-fuel combustion with coal/biomass co-firing technology as a

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Received 9 November 2022; Received in revised form 13 March 2023; Accepted 14 March 2023 Available online 22 March 2023 1743-9671/© 2023 Energy Institute. Published by Elsevier Ltd. All rights reserved. "negative emission method" to capture CO_2 , since then, the performance and synergistic effect mechanism of coal and biomass co-firing in oxy-fuel combustion has emerged as one of the most appealing topics in science investigation industrial applications all over the world [15]. During co-firing, the interactions between coal/biomass blends were primarily attributable to the catalytic and non-catalytic effect of biomass components on coal. Catalytic synergistic effect is normally dictated by Alkali and Alkali Earth Metals (AAEMs) in biomass, which increases the reactivity of chars derived from coal while non-catalytic synergistic effect is primarily related to the high volatile content in biomass [16–18].

Although the concept of pressurized oxy-fuel combustion technology was proposed by ThermoEnergy of the United States around 2000, prior to 2015, the majority of studies focused on the economic and thermal analysis of pressurized oxy-fuel combustion systems, aiming to demonstrate the benefits of pressurized oxy-fuel combustion technology in terms of enhancing the net system efficiency from macroscopical perspective [7-9]. Since 2015, more researchers have conducted basic experimental studies on various reactors, such as thermogravimetric analyzers, pressurized tube furnaces, pressurized entrained flow reactors and pressurized fluidized bed to explore the influence of coal combustion mechanism, such as reaction kinetics [5,19], ignition/combustion characteristics [6,20,21], pollutant emission [22–24], ash formation [25,26], heat and mass transfer [27,28]. So far, the pressurized oxy-fuel combustion technology has been at the stage of laboratory and pilot scale, and few kWth level pressurized oxy-fuel combustion systems have been successfully developed and operated globally. The largest publicly reported application is the construction of 1 MW pressurized fluidized bed oxygen-enriched combustion project conducted by the United States Department of Energy in 2016, and it was also discontinued due to the substantial damage of the burner in 2019 [29]. Up to now, the research object of pressurized oxy-fuel combustion was mainly coal or coal char, and the co-combustion of coal and biomass is rarely involved. Liu [1] and Tang [30] have studied the effect of pressure on sulfur and nitrogen pollutant emission and synergistic effect of coal and biomass co-combustion in a pressurized fixed bed (0.1–0.7 MPa) and a pressurized fluidized bed (0.1–0.3 MPa), found that pressure increase and biomass addition could contribute to better temperature distribution, greater combustion efficiency and lower NO_X and SO₂ emissions, but their operating pressure were relatively low. According to the simulation analysis of pressurized oxy-fuel system, the operating pressure with the optimal net efficiency is 1-1.6 MPa [9,31], so the range of 0.1–2 MPa is selected in this paper.

In general, the research on pressurized co-oxy-combustion of coal and biomass is very limited, and the performance and synergistic effects of high-pressure burning of biomass and mixed fuels are not clear yet, and the basic combustion data under high pressure are also scarce. Thermogravimetric analysis is a common experimental method to investigate combustion performance and reaction kinetics of solid materials due to its simplicity of operation, high data accuracy and repeatability. Although the TGA experiment has several limitations, such as a low heating rate and a small sample size, the greatest advantage of TGA is its accurate quantification, which can precisely measure the mass change and reaction rate of fuel during the whole combustion process. Many researchers have also conducted experimental studies on the combustion performance and interactions of co-combustion of different fuels based on TGA [32-37], indicating the reliability and feasibility of TGA research results, and the majority of these research concentrated on atmospheric pressure co-combustion. Therefore, we use magnetic levitation high-pressure thermogravimetry (HPTGA) to explore the characteristics of pressurized coal and biomass co-combustion. The advantage of magnetic levitation HPTGA is that the weight monitoring unit is completely separated from the reaction chamber, so as to avoid the interference of protective gas (N₂ or Ar) around the balance in the conventional HPTGA to the reaction atmosphere, thus the experimental results are more accurate and closer to industry. Moreover, magnetic levitation HPTGA is more convenient to explore the combustion performance in the wide pressure range. In this paper, the influence of pressure and blending ratio on co-oxy-fuel combustion is explored respectively, and the synergistic mechanism of coal and biomass in the process co-firing is discussed. A new synergistic index is defined to better quantify their interaction in pressurized combustion. This research can fill the gap in the field of pressurized coal/biomass co-combustion and provide basic data support for the industrial design and optimization of pressurized oxy-fuel combustion.

2. Experimental section

2.1. Sample preparation

A typical bituminous coal (from Pingliang, Gansu province, China) and wheat straw were selected as the experimental materials in this work.

Prior to the experiment, the raw Pingliang coal (PC) and wheat straw (WS) were crushed, ground and sieved to a particle size in the range of 96–150 μ m, then dried at 108 °C for 24 h and sealed in a desiccator for storage. WS was blended intensively with PC in different mass fractions (20%, 40%,60%,80%) before use.

The proximate and ultimate analyses of PC and WS were air-dried, based on Chinese national standards of GB/T212-2008, The PC and WS ash were prepared as per Chinese national standards of GB/T1574-2007. Then, X-ray fluorescence spectrometry (XRF, Thermo ARL 9900 USA) was used to analyze the material composition of PC and WS ash. The fuel properties analysis is presented in Table 1, and the mineral composition of ash samples is listed in Table 2.

2.2. Experimental apparatus and procedure

The pressurized oxy-fuel combustion experiments of individual fuels and their blends were implemented in a high-pressure magnetic levitation thermogravimetric analyzer{HPTGA, CHN-1807, Rubotherm Company, Germany). Its measurement principle is as follows: there is an electromagnet between the balance and the reaction chamber, which acts as a separation, therefore the reaction atmosphere environment is completely isolated from the balance, besides the measuring section does not need additional gas for protection, so there is no possibility of mixing the protective gas with the reaction gas, which can maximize to ensure the purity of the reaction atmosphere in the reaction chamber, reduce the experimental system error. The isolated magnetic levitation thermogravimetric allows experiments to be performed over a wide range of temperatures and pressures, and the real-time online fully automatic self-calibration function completely eliminates zero-point drift due to buoyancy and allows the system to calibrate the balance at any time during the whole experimental period, ensuring the longterm stability of the balance readings and the accuracy of the measurement data.

The HPTGA featured a theoretical maximum working temperature of 1200 °C, maximum pressure of 5 MPa, maximum heating rate of 50 °C/ min, balance sensitivity of 0.001 mg, and weighing range of ± 10 g. Fig. 1 shows that the HPTGA system is divided into four parts: gas regulating unit, weight monitoring unit, pressure regulating unit and circulating water cooling unit. The gas regulating unit was composed of two gas streams: pure argon was used as furnace gas to protect the outer wall of reaction chamber, and a mixture of O₂ and CO₂ was applied as the reaction gas to maintain a certain pressurized atmosphere during the experiment.

The non-isothermal method was adopted with a constant heating rate. Approximately 20 mg sample was spread evenly in the center of the quartz crucible to form a very thin layer, thereby reducing the influence of the diffusion effect between particles. After the pressure was set to the target value (0.1–2 MPa), the system would use Ar for fast stamping, when the pressure was stabilized, the gas supply was switched automatically to reaction gas $(30\%O_2+70\%CO_2)$ with the total reaction gas

Table 1

Proximate and ultimate analyses of PC and WS.

| Sample | Proximate | Proximate analysis (%) | | | | | Ultimate analysis (%) | | | |
|--------|-----------|------------------------|-----------------|-------|------------|-----------------|-----------------------|------|-----------------|-----------------|
| | Mad | A _{ad} | V _{ad} | FCad | HHV(MJ/kg) | C _{ad} | H _{ad} | Nad | S _{ad} | O _{ad} |
| PC | 5.40 | 24.53 | 25.44 | 44.63 | 20.97 | 55.32 | 3.47 | 0.64 | 0.53 | 10.11 |
| WS | 14.25 | 11.26 | 57.59 | 16.90 | 14.86 | 38.81 | 4.42 | 0.51 | 0.32 | 30.43 |

ad = air dry basis.

| Table 2 | 2 |
|---------|---|
|---------|---|

Mineral composition of PC and WS ash.

| | SiO ₂ | Al_2O_3 | Fe ₂ O ₃ | CaO | MgO | K ₂ O | Na ₂ O |
|----|------------------|-----------|--------------------------------|------|------|------------------|-------------------|
| PC | 55.01 | 23.03 | 7.35 | 6.93 | 2.62 | 1.55 | 0.83 |
| WS | 90.05 | / | 0.2 | 0.15 | 1.02 | 7.96 | 0.13 |

Magnetic levitation



Fig. 1. Schematic diagram of the HPTGA system.

flow rate of 400 mL/min). Waiting for the reaction chamber to be completely replaced by experimental gas (according to the preliminary test results, the displacement time under high pressure conditions is generally about 3 h), then the furnace was heated up from room temperature to 900 °C with 25 °C/min. During the course of heating up of the instrument, the sample weight and temperature were monitored online for analysis. It is worth noting that the increase of gas density under high pressure would cause the apparent weight gain obviously of the quartz crucible, therefore, a blank experiment had to be carried out to eliminate the influence of buoyancy. The operation with the blank sample was fully consistent with the experimental conditions, and the oxy-fuel combustion experiment of sample was subsequently carried out. Finally, using the same time span as the scale, the two sets of data were subtracted to obtain the experimental curve. Each experiment was implemented at least three times to ensure reproducibility and reliability.

3. Results and discussion

3.1. Effect of pressure on individual fuel and co-oxy-combustion

Combustion performance was evaluated using parameters directly obtained from TGA-DTG curves. Ignition temperature, peak temperature, burnout temperature are important characteristic parameters reflecting the combustion characteristics of pulverized solid fuel particles. The tangent method of TG-DTG curve was used to define ignition temperature (T_{ib} °C) [38]. Peak temperature (T_{maxs} °C) indicated the point at which the weight loss rate reached its maximum. Burnout temperature (T_{b} °C) was defined as the temperature when the combustion conversion rate reaches 98% and remains stable [39].

In order to evaluate the combustion performance more comprehensively, the ignition index (C_i , min³), burnout index (C_b , %/min⁴), and comprehensive combustion characteristic index (S, %/°C³ × min²) were introduced to measure the combustion performance of different fuel and their blending. Higher C_i and C_b corresponded to better ignition and burnout capabilities. *S* comprehensively reflected the flammability and burning performance of fuels, and a higher *S* value indicated better combustion reactivity. The above parameters were calculated using the following formula [40,41]:

$$C_i = \frac{(dw/dt)_{\max}}{t_i t_{\max}} \tag{1}$$

$$C_b = \frac{(dw/dt)_{\max}}{\Delta t_{1/2} t_{\max} t_b} \tag{2}$$

$$S = \frac{R}{E} \cdot \left[\frac{d(\frac{dw}{dt})}{dT} \right]_{T=T_i} \cdot \frac{(dw/dt)_{\max}}{(dw/dt)_{T=t_i}} \cdot \frac{(dw/dt)_{\max}}{t_b} = \frac{(dw/dt)_{\max}(dw/dt)_{\max}}{t_i^2 t_b}$$
(3)

In the above equations, $(dw/dt)_{max}$ represents the maximum reaction rate, %/min; $(dw/dt)_{mean}$ represents the average reaction rate, %/min; t_i represents the ignition time, min; t_{max} is the time corresponding to the maximum reaction rate, min; t_b represents the burnout time, min; $\Delta t_{0.5}$ denotes the time interval of $(dw/dt)/(dw/dt)_{max}=0.5$, min.

The whole oxy-combustion process of PC and WS at the heating rate of 25 °C/min in 30%O₂/70%CO₂ normbaric atmosphere is illustrated through typical TG-DTG curves. As shown in Fig. 2(a), with the increase of environmental temperature, the weight loss of PC can be divided into three representative stages. The first stage includes temperatures up to approximately 438 °C. The coal mass increases slightly with increasing temperature during the initial heating period because the gases in the surroundings become physically adsorbed on the coal particle surface, and the physical adsorption is non-selective. Meanwhile, the chemical adsorption on the coal surface is much weaker, causing the apparent weight of coal to rise in a short time [42]. As the temperature further increases, the thermal movement of gas molecules enhance, it is not sufficient to maintain molecular forces on the coal surface, thus the gas would be desorbed. And accompanied by the dehydration and devolatilization of coal, hence causes a constant decline in mass. The second stage exhibits a single board peak area (438-634 °C) in DTG curve,



Fig. 2. TG-DTG curves of normbaric oxy-combustion: (a) PC (b) WS.

corresponding to the weight loss of 63.61 wt% which can be ascribed to the concurrent combustion of volatile and fixed carbon components. Additionally, the third stage (634–900 $^{\circ}$ C) is the burnout of residual low reactive char into ash.

Compared with coal, the weightless properties of wheat straw are more complicated during the oxy-combustion process. As shown in Fig. 2(b), it can be divided into four stages. The first stage $(100-282 \degree C)$ results in a weight loss of around 7.49 wt% owing to moisture evaporation and the initial release of volatile compounds. The sharp peak in second stage $(282-377\degree C)$ in DTG curve is attributed to oxy-combustion of volatile fraction released from cellulose, hemicellulose, and trace amounts of lignin, the three main components of lignocellulosic



Fig. 3. TG-DTG curves of pressurized oxy-combustion of PC, WS and their blends.

materials. According to previous combustion results [43,44], hemicellulose decomposition by combustion occurs primarily below 300 °C, whereas it can be supposed that it was still decomposing both with lignin at 500 °C. Cellulose burns between 300 and 377 °C, so while lignin and hemicellulose show extended mass loss until 507 °C [40]. The third stage (417–535 °C) is the decomposition of lignin and the combustion of fixed carbon. Moreover, the last stage (535–900 °C) is the burnout of residual char.

The TG and DTG profiles of PC, WS, and their blends (80%PC+20% WS) combustion at the heating rate of 25 °C/min in 30%O₂/70%CO₂ environment as the pressure increased from 0.1 to 2 MPa are shown in Fig. 3. For coal oxy-combustion, when the pressure increases from 0.1 to 1 MPa, the DTG curves shift systematically to a lower temperature zone, while the peaks expand larger, indicating that elevating the pressure could accelerate the pyrolysis and combustion of coal, shorten the time from ignition to burnout. As a result, the combustion properties of coal are significantly improved by increasing the pressure within between 0.1 and 1 MPa. The DTG curves shift to higher temperature zone when the pressure further increased to 2 MPa, and the DTG peaks develop substantially sharper and narrower, indicating although the rising pressure delay the ignition and burnout time, it doesn't prevent the coal from burning at a faster rate. Meanwhile, the biomass combustion exhibits the same trend with increasing pressure, but the turning point is advanced to 0.5 MPa. Additionally, Zhou [45] has proposed a method to visually determine the ignition mode by the number of weightlessness peaks in DTG curves, with double peaks representing homogeneous ignition and single peaks representing heterogeneous ignition. As can be seen from Fig. 3(d), As the system pressure exceeds 1 MPa, the DTG curve of WS changes from two distinct peaks to a single peak, and the boundary between volatile combustion and fixed carbon combustion is no longer obvious, indicating that the ignition mode of WS transforms from homogeneous to heterogeneous ignition. For the heating rate as low as 25 °C/min, the main parameter that affects the ignition mode most is the relative magnitude of the heating rate on the surface of fuel particle and the volatile release rate. When the heating rate on particle surface is higher than the pyrolysis rate, ignition occurs on particle surface, called heterogeneous ignition. On the contrary, ignition occurs in the gaseous boundary layer around particles, called homogeneous ignition. The volatiles and char ignite concurrently on the particle surface when the two rates are similar, which is known as the combined ignition mode [46,47]. The heating rates of particles in HPTGA remain consistent (25 °C/min), as a result, the rate of volatile release has the greatest influence on the ignition pattern. Increasing pressure reduces the release rate and the overall amount of volatile [48,49] thus changing the ignition mode of biomass particles.

The maximum and average combustion rate of different samples were calculated according to TG and DTG curves in Table 3, With the pressure increases, the maximum and average combustion rates of coal, biomass and its blends show a rising trend, but the magnitude of the growth diminishes, exhibiting a multiplicative increase below 1 MPa and a sluggish growth above 1 MPa. At normbaric condition, the internal diffusion mass transfer process of particles is caused by concentration gradient. Both coal and biomass are degraded by heat throughout rising system temperature, producing a substantial quantity of volatile compounds. Owing to the rapid slip velocity between volatiles and surrounding environment, the released volatile compounds could rarely concentrate around the sample particles. The majority of them escaped from the coal or biomass particles, making the remaining char difficult to burn and reducing the reaction rate. As external environmental pressure increases, it becomes more challenging for internal volatile content to disperse from coal or biomass particles. This inhibition will result in higher concentration of volatiles in the stomata of solid particles and strengthening of the secondary reaction between volatiles, then cause an increase in gaseous products and the internal pressure of sample particles increases. Forced convection would be formed when internal pressure exceeds external pressure, which allowing for faster

Table 3

Combustion rate for samples under different pressure in $30\% O_2/70\% CO_2$ atmosphere.

| Sample | T _{max1} (°C) | T _{max2} (°C) | dW/dt _{max1} (%/min) | dW/dt _{max2} (%/min) | dW/dt mean (%/min) |
|---------------|---------------------------|---------------------------|-----------------------------------|-----------------------------------|-----------------------|
| PC | 534 | - | 11.28 | _ | 8.11 |
| PC | 423 | - | 22.39 | - | 10.59 |
| PC | 416 | - | 51.21 | - | 15.67 |
| PC | 442 | - | 63.35 | - | 33.23 |
| PC | 473 | - | 73.88 | - | 47.73 |
| | | | | | |
| WS | 316 | 417 | 20.66 | 29.48 | 7.96 |
| WS | 296 | 430 | 62.26 | 2.42 | 11.32 |
| WS | 306 | - | 94.39 | - | 14.36 |
| WS | 318 | - | 116.69 | - | 19.97 |
| WS | 334 | - | 124.82 | - | 35.80 |
| | | | | | |
| 80PC/ 20WS | 323 | 518 | 9.47 | 3.67 | 5.50 |
| 80PC/ 20WS | 300 | 436 | 14.05 | 5.37 | 7.02 |
| 80PC/ 20WS | 349 | 430 | 49.99 | 5.83 | 14.53 |
| 80PC/ 20WS | 370 | 454 | 52.50 | 5.08 | 12.32 |
| 80PC/ 20WS | 394 | 475 | 62.40 | 2.83 | 13.14 |

and more efficient mass transport compared with normally diffusion in atmospheric environment [50]. Besides, as pressure increases, the interior of fuel particles tends to fragment, and the degree of carbon deposition intensifies [21]. Volatile compounds can concentrate around the fuel particles because the slip velocity between them and surrounding gas is slower under pressure than under normal condition. So, pressurization helps to increase the combustion rate and shorten the reaction time.

Furthermore, for the low heating rate of HPTGA, the combustion reaction rate is dominated by chemical reaction kinetics, as the model-free method proposed by Liu [51], the combustion rate can be described by Eq. (1),

$$\frac{d\alpha}{dt} = k(T)f(\alpha)P_{O_2}^m \tag{4}$$

where α represents the conversion rate of sample, $f(\alpha)$ is a structural factor, which is a function of α and depends on the structural characteristics of sample particles, $P_{O_2}^m$ represents the partial pressure of oxygen, and *m* is the reaction order. k(T) represents the reaction rate parameter, which is determined by Arrhenius's equation. Under constant experimental conditions, with the increase of total pressure, the partial pressure of oxygen also increases, thus enhance the combustion rate.

During the system pressure further increased from 1 MPa to 2 MPa, the effect of combustion promotion weakens progressively. The whole process of reaction gas can be regarded as steady-state diffusion, and according to Fick's first law, the oxygen diffusion flux (molar flow rate) at the interface of sample particles can be defined by Eq. (3) [52]:

$$N_{O_2} = \frac{D_{O_2}}{h_0} A_N c(O_2)$$
(5)

where D_{O_2} represents the diffusion coefficient of oxygen, h_D represents the vertical distance between the top of crucible and the sample surface, A_N represents the horizontal cross-sectional area of the crucible, and $c(O_2)$ represents the concentration of oxygen.

In the thermogravimetric experiment, the flow rate of the reaction gas is high enough, thus it can be assumed that the oxygen concentration does not change with time, and all experiments are conducted with the same crucible, the value of h_D and A_N are consistent. Accordingly, we

can infer that the diffusion flux of oxygen is only related to its diffusion coefficient. The diffusion coefficient of the binary mixture is generally calculated using the formula proposed by Fuller [26]:

$$D_{12} = \frac{0.1013T^{1.75} \left(\frac{1}{M_1} + \frac{1}{M_2}\right)^{0.5}}{p \left[\left(\sum v_1\right)^{1/3} + \left(\sum v_2\right)^{1/3} \right]^2}$$
(6)

where *p* represents the system pressure, *T* represents the system temperature, M_i represents the molar mass of gas molecules, and v_i represents the diffusion volume of gas molecules.

As shown in Eq. (4), when the mixed reaction gas fraction and the heating rate are constant, $D_{O_2} \propto_p^1$. Total pressure increases limit oxygen diffusion coefficient, slows oxygen supply rate in burning region, and weakens pressure acceleration on sample combustion process. Therefore, when pressure further increased from 1 MPa to 2.0 MPa, the upward trend of sample combustion rate reduces, ignition and burnout are both delayed.

As shown in Fig. 4, initially, the pressurization led to an early ignition, but when the pressure exceeds 0.5 MPa, PC, WS and their blends show a delay in ignition. with further increase of pressure, the C_i and C_h coal first increase and remain stable after reaching 1 MPa. As mentioned above, the effect of pressure on combustion characteristics is both facilitative and inhibitive, in terms of reaction kinetics, pressurization can significantly promote combustion but it also plays a role in inhibiting oxygen diffusion, the competition between the two leads to an optimum value of pressure in the combustion performance of the blends, which is about 1 MPa.

It's worth noting that compared with coal, biomass has a much larger exponential increase in C_i and C_h after pressure elevated,WS shows a higher sensitivity to pressure, As the system pressure surpasses 1 MPa, the line between volatile and fixed carbon combustion has blurred. the ignition of mechanism transforms from heterogeneous to homogeneous ignition. The high O/C mole ratio of WS contributes to the formation of char particles with high reactivity. Simultaneously, the high ratio of volatile matter to fixed carbon in WS results in the creation of a porous

structure, which improves the overall reactivity. Furthermore, thermal stability of cellulose and hemicellulose derived from biomass is lower than that of organic macromolecular grids in coal. Under elevated pressure, the former decomposes more readily into volatile gases. Additionally, the diffusion rate of volatiles reduces after pressurization, making it difficult for them to escape to the surface of biomass char particles. Consequently, the interior of volatile particles is simpler to ignite than the exterior, the ignition mechanism gradually shifts to heterogeneous ignition. In general, the comprehensive combustion characteristics of WS continue to rise with the increase of pressure.

As seen in Figs. 3(e) and Fig. 4, under atmospheric pressure, the DTG curves of PC/WS blends can be separated into two distinct peaks, and the combustion process can be divided into four distinct stages. The first stage (100-288 °C) consists of the evaporation of water and the initial emission of volatiles. In the second stage (288-366 °C), volatiles are precipitated and burned. The third stage (366-623 °C) is the burning of fixed carbon in biomass and coal, and the fourth stage (after 623 °C) is the emission of residual carbon and the loss of fly ash. When WS is blended with PC, the ignition mechanism transforms from heterogeneous to homogeneous. The combustion of volatile matter in coal, which serves as a preheating agent for the combustion of fixed carbon in coal, is facilitated by the ignition of WS. As a result, the ignition temperature is greatly lowered, the maximum combustion weight loss rate advances, and the overall combustion properties improve. When the pressure increases from 0.1 MPa to 2 MPa, the ignition temperature of PC/WS blends rises after initially decreasing. After the pressure exceeds 1 MPa, the volatile combustion region primarily shifts to the low temperature zone before moving to the high temperature area. Similar DTG peak values at various pressures and little variation in interval length suggest that the pressure has a negligibly slight boosting influence on the mixture's volatile combustion stage. When the pressure reaches 2 MPa, the first peak value of the DTG of the PC/WS blends decreases dramatically and the combustion zone lengthens, indicating that the combustion of volatile is significantly inhibited, which is due to the precipitation of volatile is restrained under higher pressure, hydrogen molecules in solids retention time extend gradually, then the secondary pyrolysis



Fig. 4. Combustion characteristic parameters for samples under different pressure: (a) T_i/T_b;(b) C_i;(c) C_h; (d) S.

reaction with semi-coke would be carried out by deposition and repolymerization, generating small molecular compounds attached to the surface of the semi-coke [49], thereby increasing the yield of semi-coke and lengthening the burning time of the volatile. In addition, high pressure reduces the speed of diffusion, causing the initial volatile in the blending particles' residence duration to increase and ignition delay. The second DTG peak of blends combustion grows rapidly with the rising pressure, and the combustion duration shorten sharply, showing that the pressure has a clear promotional effect on the fixed carbon combustion stage. The comprehensive combustion characteristic index of PC/WS blends raise first and subsequently reduce with increasing pressure, reaching its maximum value at 1 MPa, indicating that the combustion performance is optimal at this pressure.

3.2. Effect of blending ratio on co-oxy-combustion

In order to determine the ideal blending ratio of coal and biomass under pressurized oxy-fuel conditions, in the atmosphere of 30% $O_2+70\%$ CO_2 at 0.5 MPa, experiments were carried out with different mass mixing ratios (WS addition ratios were 0%, 20%, 40%, 60%, 80%, 100%). The results are shown in Fig. 5, and the combustion rate are calculated in Table 4. The combustion characteristic parameters are shown in Fig. 6.

As can be seen in Fig. 5 (b), the combustion of PC/WS blends can be roughly divided into four stages. The temperature from 100 °C to 260 °C of stage 1 is the process of water evaporation and initial analysis of volatiles, and the temperature from 260 °C to 340 °C of stage 2 provides the necessary thermodynamic conditions for the devolatilization and combustion of volatiles in biomass. 340–530 °C of stage 3 is fixed carbon combustion of coal and biomass. 530°C-900 °C of stage 4 is the burnout of residual carbon and the formation of ash. In stage 2, as the proportion of biomass increases, the TG curve shifts toward the lower temperature zone, the DTG peak value rises, and Ci and Ch exhibit an upward trend. It demonstrates that adding biomass can improve coal combustion conversion at the same temperature. This is because the volatiles of WS are more than twice that of PC, the higher the proportion of WS in the mixture, the more intense the combustion of volatiles is. The volatile in biomass can be released and burn at lower temperatures, accompanied by the generation of hydrogen rich gas product and heat, which can improve the ignition and combustion of coal [53]. Furthermore, biomass is abundant in alkali metals and mineral components with a catalytic effect on coal, as shown in Table 2, the potassium content of WS (7.96%) is much higher than that of PC (1.55%), hence enhancing coal combustion performance [54]. In Stage 3, when the biomass ratio rises, the peak DTG value first rises and then falls, and the burnout temperature initially decreases and then gradually increases. The co-combustion properties are at their best when the WS ratio reaches 60%.

Table 4

Combustion characteristic parameters for different blending ratio at 0.5 MPa.

| Sample | T _{max1} (°C) | T _{max2} (°C) | dW/dt _{max1} (%/min) | dW/dt _{max2} (%/min) | dW/dt mean (%/min) |
|---------------------|---------------------------|---------------------------|-----------------------------------|-----------------------------------|-----------------------|
| PC 80PC/ 20WS | 423 300 | - 436 | 22.39 5.37 | _ 14.05 | 10.59 7.02 |
| 60PC/ 40WS | 310 | 438 | 34.17 | 18.19 | 8.38 |
| 40PC/ 60WS | 304 | 355 | 45.09 | 9.40 | 12.47 |
| 20PC/ 80WS | 300 | 430 | 53.28 | 6.23 | 8.92 |
| WS | 296 | 430 | 62.26 | 2.42 | 11.32 |

3.3. Interactions during co-oxy-combustion process

3.3.1. Analysis method of interactions

The theoretical TG and DTG curves, which indicate the sum of the individual mass loss from different fuels as a weighted average, are calculated by Eq. (5)(6) to further investigate the interactions between the burning of coal and biomass blends, in addition, their deviation from the experimental curve by Eq (7)(8) [55].

$$TG_{cal} = (1 - x)TG_{(PC)exp} + xTG_{(WS)_{exp}}$$
⁽⁷⁾

$$DTG_{cal} = (1 - x)DTG_{(PC)exp} + xDTG_{(WS)_{exp}}$$
(8)

$$\Delta w = TG_{\rm exp} - TG_{\rm cal} \tag{9}$$

$$\Delta D = DTG_{\rm exp} - DTG_{\rm cal} \tag{10}$$

Where the *x* represents the mass fractions of WS in blends; $TG_{(PC)exp}$, $TG_{(WS)exp}$, $DTG_{(WS)exp}$ and $DTG_{(PC)exp}$ refer to TG or DTG experimental curve of PC or WS; Δw and ΔD represent the deviation between theoretical value and experimental curve.

The strength of the interaction increases with the value of Δw or ΔD . Positive synergy is indicated by a value less than 0 and negative synergy by a value greater than 0. Δw can better represent the sensitivity of conversion rate to temperature, whereas ΔD represents the sensitivity of combustion rate to temperature.

3.3.2. The effect of pressure on the interaction

It can be seen from Fig. 7 that in comparison to the theoretical curve, the experimental TG curve moves to the lower temperature region at atmospheric pressure. In 100–400 °C, the value of Δw is about 0, indicating that there is no obvious synergistic effect between dehydration and volatile combustion stages. In 400–650 °C, Δw <0, the peak of experimental DTG curve moves to the lower temperature region and the value is larger than the theoretical curve, indicating that co-combustion



Fig. 5. TG and DTG curves of co-combustion with different WS addition ratio at 0.5 MPa.



Fig. 6. Combustion characteristic parameters for samples with different WS addition ratio at 0.5 MPa: (a) Ti/Tb; (b) C_i/C_h; (c) S.

have an obvious positive synergistic effect in the section of PC/WS blends fixed carbon combustion. The rapid combustion of WS volatiles can increase the reaction temperature and provide additional heat, which is conducive to accelerate the subsequent combustion of semicoke in high temperature region. In the process of biomass pyrolysis, high proportion of oxygen-rich substances can produce pyrolysis gas with high activity, then promote the following gas-solid heterogeneous combustion reaction [56]. Intermediates released from lignin in biomass, such as phenoxy radicals, attack coal molecules, leading to the fracture of aliphatic C–C bonds, and facilitate the thermal pyrolyzation and combustion of coal at high temperature [57].

The experiments under pressure are markedly different from that under atmospheric pressure, when the environmental pressure exceeds 0.5 MPa, the value of Δw changes from negative to positive, indicating that the interaction between PC and WS changes from positive synergy to inhibitory effect, and the Δw curves show two obvious peaks corresponding to volatile combustion stage and fixed carbon combustion stage respectively. Δw curves shift to the higher temperature region, and the peak values gradually increase as the pressure elevated from 0.5 MPa to 2 MPa, indicating that the negative synergistic effect of PC/WS cocombustion gradually enhance, which can also be seen in Fig. 7 (d) (e) (f) (g), compared with the theoretical curve, the DTG experimental curves show that the first peaks move backward and grow larger, demonstrating that in the stage of volatile combustion, the actual burning rate is lower than the theoretical rate while the actual ignition temperature is higher than the theoretical value. Volatiles combustion of biomass and pyrolysis of coal are mainly carried out in this temperature range. With the increase of pressure, volatiles are more prone to secondary reactions, resulting in carbonaceous sediments that will cover the surface of the PC/WS blends at low temperatures, hindering mass transfer between them, resulting in the intensification of negative synergy and ignition delay [57]. In the stage of fixed carbon combustion, with a rise in pressure, the peak of Δw exhibits a tendency of initially increasing and then falling. The second peaks of experimental DTG exceed the theoretical value when the pressure is more than 1 MPa, Combined with Fig. 7(e), it can be seen that the interactions between PC and WS under pressurized conditions put off the ignition and combustion process, but increase the actual combustion rate of the blends. The ash composition of WS contains a high content of K, as shown in Table 2, and the volatiles (such as K⁺, KCL, and KOH) released by alkali metal compounds at 400–550 °C have a catalytic effect on the decomposition of PC, which can suppress the formation of stable chemical structures and weaken the strength of C-C bond in PC, thus promoting the combustion rate of the blended fuel [58].

3.3.3. The effect of blending ratio on the interaction

As can be seen from Fig. 8, under the pressure of 0.5 MPa, when WS addition ratio is between 40% and 60%, the interaction between PC and WS is beneficial to combustion in the fixed carbon combustion region (300 °C–500 °C), since the hydrogen-carbon molar ratio of WS (H/C =

1.37) is much higher than that of PC (H/C = 0.75), a large number of hydrogen donors (H and OH radicals) produced by WS participate in thermal decomposition of PC and inhibit the re-polymerization and cross-linking reactions of free radicals during PC/WS co-combustion [59,60]. In addition, large amounts of intermediates (such as phenoxy radicals) generated by thermal decomposition of lignin in biomass at low temperatures will induce the decomposition of dense, heat-resistant coal structures [33]. However, when the blending ratio exceeds 60%, the ash content of WS is as high as 11.26%, so the ash produced by rapid combustion of biomass at low temperatures will coat the surface of PC, which will further inhibit the diffusion of oxygen and contact with semi-coke particles, resulting in a negative synergistic effect between them. This indicates that at 0.5 MPa, there is an optimal biomass addition ratio, and the co-combustion characteristics may be stable or even decrease after reaching a certain blending ratio. In this study, the optimal ratio is 60%.

3.4. A new novel index for describing interaction of co-oxy-combustion

From the above analysis, using the deviation of theoretical value and experimental value of TG and DTG curves the to evaluate co-combustion synergistic effect has certain limitations, and the performance of the whole combustion process also includes a series of parameters such as ignition temperature and peak temperature and so on. Therefore, a new synergy index is proposed in our study, comprehensively involved the change of deviation and combustion characteristic parameters, and can better evaluate the co-combustion interaction between coal and biomass.

$$SI = \exp\left(-\frac{\overline{\Delta w} \times 10^3}{T_i T_p}\right)^{\frac{1}{3}}$$
(11)

Where $\overline{\Delta w}$ Represents the average deviation of the TG curve, T_i is the ignition temperature, T_p is the peak temperature. *SI* can be used to establish a comparison benchmark. When SI > 1, there is positive synergy, SI < 1, there is negative synergy, and SI = 0, there is no synergy, only additive behavior. The calculation results are shown in Table 5.

Under atmospheric pressure, when the adding proportion of WS is 20%, there is an obvious positive synergistic effect between PC/WS blends (SI = 1.0543), however, under the condition of pressure, adding the same proportion of WS shows negative synergistic effect, and the higher the pressure, the smaller the SI, indicating that the negative synergistic effect increases with the increase of pressure. In general, the co-combustion synergistic mechanism of biomass and coal is related to the non-catalytic mechanism and the catalytic mechanism. The former refers to the release of biomass volatiles and the formation of free radicals and the transfer of hydrogen from biomass to coal, while the latter is based on the catalytic action of AAEMs in biomass [32,57,59]. At a certain blending ratio, the increase of pressure will reduce the total amount of volatiles released from biomass. The mechanism of volatile







Fig. 8. Comparison of experimental/calculated TG/DTG curves with different WS addition ratios at 0.5 MPa.

Table 5Synergy indices of PC/WS blends.

| Sample | Pressure (MPa) | $\overline{\Delta w}$ | T _i (°C) | T _p (°C) | SI |
|-----------|----------------|-----------------------|---------------------|---------------------|--------|
| 80PC/20WS | 0.1 | -0.0221 | 287 | 518 | 1.0544 |
| 80PC/20WS | 0.5 | 2.4192 | 280 | 436 | 0.7629 |
| 80PC/20WS | 1 | 3.5758 | 320 | 430 | 0.7436 |
| 80PC/20WS | 1.5 | 3.9772 | 335 | 454 | 0.7432 |
| 80PC/20WS | 2 | 4.3292 | 347 | 475 | 0.7429 |
| 80PC/20WS | 0.5 | 2.4192 | 280 | 436 | 0.7629 |
| 60PC/40WS | 0.5 | -0.3388 | 295 | 310 | 1.1673 |
| 40PC/60WS | 0.5 | -2.4644 | 290 | 304 | 1.3546 |
| 20PC/80WS | 0.5 | 1.3505 | 288 | 300 | 0.7788 |

precursor transport inside the particle is attributed to either volume diffusion due to concentration difference or forced convection due to pressure gradient. Rising external pressure reduces the pressure gradient between the inside and outside of biomass particles, slows volatile diffusion through the semi-coke pores to the surrounding, and raises the residence time of volatile precursor in the particles. Therefore, compared with atmospheric pyrolysis, the secondary reaction of pressurized pyrolysis is strengthened, the generation rate of semi-coke is increased, and the total release of volatile matter is reduced, and the hydrogen donor (H and OH radicals) produced by WS is also reduced [49,61,62], hence higher pressure reduces non-catalytic co-combustion promotion effect. In addition, raising pressure will significantly reduce the volatilization yields of ion exchangeable Mg and Ca K in AAEMs at the same temperature of biomass [59,62]. Therefore, the catalytic

promotion effect of AAEMs on coal is also weakened by pressurization. In addition, the more crucial reason is that elevating pressure promotes biomass combustion more deeply than coal combustion. Varying degrees of same pressure influence on coal and biomass result in distinct ignition and combustion temperature ranges for blended fuels. according to Fig. 3(c), elevating system pressure will cause main combustion process of WS move towards to lower temperature zone, consumedly shorten reaction duration, and the combustion of WS have basic finished burning before 400 °C, while combustion of PC still continues, thus the formation ash will coat on the surface of coal particles, hindering heat and mass transfer processes, finally, the negative synergistic effect of PC/WS hybrid combustion is enhanced. Similarly, at atmospheric pressure, the addition of 20% WS has a significant promotion effect on combustion, but at 0.5 MPa, the promotion effect only occurs when the addition ratio exceeds 40%. As mentioned above, pressurization inhibits the effect of both non-catalytic and catalytic mechanisms, so the biomass ratio needs to be raised to achieve the same enhancement effect, and the co-combustion characteristics may be stable or even decrease after reaching a certain blending ratio. In this study, the optimal ratio is 60% (SI = 1.3545).

4. Conclusion

In this paper, the effects of pressure and blending ratio on the combustion of coal/biomass separately and co-oxy-combustion are investigated with HPTGA, and it is verified that the pressure increase from 0.1 to 2 MPa helps to improve the reaction rate and comprehensive combustion characteristics of PC and WS, but it causes ignition delay after system pressure exceeding 0.5 MPa. The effect of pressure on combustion characteristics is both facilitative and inhibitive, in terms of reaction kinetics, pressurization can significantly promote combustion but it also plays a role in inhibiting oxygen diffusion, the competition between the two leads to an optimum value of pressure in the combustion performance of the blends, which is about 1 MPa.

The effect of pressurization on enhancing the combustion of biomass is substantially greater than that of coal. At 0.1–2 MPa, PC consistently exhibits heterogeneous ignition, whereas the ignition mechanism of WS shifts from homogeneous to heterogeneous at 1 MPa and above.

A new synergy index is proposed in our study, comprehensively involved the change of deviation and combustion characteristic parameters, and can better evaluate the co-combustion interaction between coal and biomass. Under atmospheric pressure, the cocombustion of PC and WS presents a positive synergistic effect, while it appears a negative synergistic effect in pressurized situation, and the negative effect tends to increase with the rising pressure, which is due to the different sensitivities of PC and WS to pressurization lead to more significant differences in combustion time and temperature interval compared with normal pressure, and pressurization could suppress both catalytic and non-catalytic mechanisms in the co-combustion process, all these factors contribute to negative interaction of co-oxycombustion. The WS blending ratio needs to be increased after pressurization to produce a positive synergistic effect, and the optimal blending ratio is about 60%.

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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