

## **Effect of oxy-fuel combustion with steam addition on coal ignition and burnout in an entrained flow reactor**

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### **Abstract**

The ignition temperature and burnout of a semi-anthracite and a high-volatile bituminous coal were studied under oxy-fuel combustion conditions in an entrained flow reactor (EFR). The results obtained under oxy-fuel atmospheres (21%O<sub>2</sub>/79%CO<sub>2</sub>, 30%O<sub>2</sub>/70% O<sub>2</sub> and 35%O<sub>2</sub>/65%CO<sub>2</sub>) were compared with those attained in air. The replacement of CO<sub>2</sub> by 5, 10 and 20% of steam in the oxy-fuel combustion atmospheres was also evaluated in order to study the wet recirculation of flue gas. For the 21%O<sub>2</sub>/79%CO<sub>2</sub> atmosphere, the results indicated that the ignition temperature was higher and the coal burnout was lower than in air. However, when the O<sub>2</sub> concentration was increased to 30 and 35% in the oxy-fuel combustion atmosphere, the ignition temperature was lower and coal burnout was improved in comparison with air conditions. On the other hand, an increase in ignition temperature and a worsening of the coal burnout was observed when steam was added to the oxy-fuel combustion atmospheres though no relevant differences between the different steam concentrations were detected.

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## **1. Introduction**

The use of coal in power plants generates a large amount of CO<sub>2</sub>, which is the chief contributor to global climate change. However, coal is the most abundant and geographically the most widely distributed fossil fuel. The stability of its supply and its relatively low cost ensure its inclusion in the energy mix in the foreseeable future [1].

Several strategies for the reduction and capture of CO<sub>2</sub> from large-scale stationary power plants are being considered. The main problem is that the concentration of CO<sub>2</sub> in conventional coal-air combustion flue gas is low (typically 14-16%) and its direct storage is not economically feasible [2]. As an alternative, the oxy-fuel combustion process has been proposed as a new promising technology for CO<sub>2</sub> capture from fossil fuel power plants. This technology entails the combustion of coal in a mixture of oxygen (instead of air) and recycled flue gas (RFG), which mainly contains CO<sub>2</sub> and H<sub>2</sub>O. The net volume of flue gas is reduced and a highly concentrated CO<sub>2</sub> (95%) flue gas is produced, which can be directly stored in a supercritical state by means of compression.

The combustion of coal in the O<sub>2</sub>/CO<sub>2</sub> atmosphere of an oxy-coal combustion boiler can be expected to be different from that of an O<sub>2</sub>/N<sub>2</sub> atmosphere of a conventional coal-air combustion boiler, because CO<sub>2</sub> has a larger specific molar heat than N<sub>2</sub>, and the coal can be gasified by the CO<sub>2</sub> [3]. The replacement of N<sub>2</sub> by CO<sub>2</sub> may affect the operation parameters of the combustion furnace. The propagation speed and stability of the flame and the gas temperature profile may decrease or the unburned carbon content may

increase. During the oxy-fuel combustion process, this problem can be overcome by increasing the oxygen concentration in the oxy-fuel combustion atmosphere (up to approximately 30%) in order to match the combustion performance achieved in air, in relation to flame temperature, ignition time, heat transfer, gas temperature profile and char burnout. This would also contribute to the overall efficiency of the oxy-fuel process [4]. The flexibility acquired by adjusting the O<sub>2</sub> concentration in the feed gas to control the heat flux and flame temperature would have an additional advantage in that it would allow a more flexible selection of fuels, especially for coal-fired power plants [5].

One of the advantages of this technology is that it would prevent the formation of thermal NO<sub>x</sub> due to the absence of nitrogen. It has been shown that a reduction in NO<sub>x</sub> emissions can be achieved with oxy-fuel combustion compared to air-firing [6-8].

In addition, oxy-coal combustion allows the capture and storage of carbon as a retrofit technology in conventional pulverized coal boilers.

Although oxy-fuel recycle combustion requires that some modifications are carried out in the existing pulverized coal combustion technology that has well-proven reliability and industrial acceptance, it is a promising approach for continued use of coal for electric power production [9]. However, the successful implementation of O<sub>2</sub>/CO<sub>2</sub> technology in conventional pulverized coal boilers requires a full understanding of the changes that occur when N<sub>2</sub> is replaced by CO<sub>2</sub> in the combustion atmosphere [10]. The ignition of coal particles is an important preliminary step in the coal combustion process due to its influence on flame stability, the formation and emission of pollutants and flame extinction [11]. Thus, the study of the reactivity and the minimum gas

temperature for the ignition of coal particles will be very important when designing the boiler and for controlling the combustion process [12].

Although previous work has already dealt with the oxy-fuel combustion process in entrained flow reactors (EFR) [9] or drop tube furnaces (DTF) [13,14], and simulation studies have been published [15-17], the presence of steam in the oxidizer stream has not received significant consideration [18,19]. Recently, Smart et al. [20] showed that the combustion of coal in an O<sub>2</sub>/CO<sub>2</sub>/H<sub>2</sub>O atmosphere, in relation to ignition, burnout, flame stability and emissions, requires further clarification.

In this work, an entrained flow reactor was used in order to study the effect of replacing N<sub>2</sub> with CO<sub>2</sub> and of enhanced levels of O<sub>2</sub> in an oxy-fuel combustion atmosphere upon coal ignition temperature and burnout. The addition of steam has also been evaluated in order to study the effect of the wet recirculation of flue gas.

## **2. Experimental**

### *2.1. Coals*

Two coals of different rank were used: a semi-anthracite from the Hullera Vasco-Leonesa in León, Spain (HVN), and a high-volatile bituminous coal from the Batán coal washing plant in Asturias, Spain (BA). The samples were ground and sieved to obtain a particle size fraction of 75-150 µm. The results of the proximate and ultimate analyses and higher heating values of the samples are shown in Table 1.

### *2.2. Experimental device and procedure*

An entrained flow reactor was used to study the ignition and the oxy-fuel combustion characteristics of the coal at high heating rates and short residence times. A diagram of

the experimental device used is shown in Fig. 1. The reactor has a reaction zone with a maximum length of 1700 mm and an internal diameter of 40 mm. The EFR is electrically heated and is able to work at a maximum temperature of 1100 °C. The coal samples were fed in from a hopper and the mass flow was controlled using a mechanical feeding system. The samples were introduced through an air-cooled injector to ensure that their temperature did not exceed 100 °C before entering the reaction zone. The gases were preheated to the oven temperature before being introduced into the reactor through flow straighteners. The flow rates of N<sub>2</sub>, CO<sub>2</sub> and O<sub>2</sub> from the gas cylinders were controlled by mass flow controllers. The steam was generated by a vaporizer, in which water was heated up to 250 °C. A high performance liquid chromatography (HPLC) pump was used to control the flow rate of water towards the vaporizer. The steam was mixed with the inlet gases before they were introduced into the pre-heater. A water-cooled collecting probe was inserted into the reaction chamber from below. Nitrogen was introduced at the top of the probe to quench the reaction products. Particles were removed by means of a cyclone and a filter, and the exhaust gases were monitored using a battery of analysers (O<sub>2</sub>, CO<sub>2</sub>, CO, NO and SO<sub>2</sub>).

During the ignition tests, the reactor was heated at 15 °C min<sup>-1</sup> from 400 to 800 °C. The gas flow used in these tests ensured a particle residence time of 2.5 s at 500 °C, and the excess oxygen (defined as the O<sub>2</sub> supplied in excess of that required for the stoichiometric combustion of coal) was fixed at a value of 25%. Ignition is the initiation stage of combustion and it is defined as the temperature at which the heat generated by the reactions exceeds the heat losses by various mechanisms of heat transfer. The criterion for determining the ignition temperature was based on the first derivative temperature curves of the gases produced. The ignition temperature was taken as the

temperature at which the first derivative temperature curve, normalized by the maximum derivative value, reached a value of 10% [12].

On the other hand, the combustion experiments were carried out at a reactor temperature of 1000°C and a particle residence time of 2.5 s. The burnout is defined as the loss of mass of a fuel during its combustion and it is expressed as the ratio of mass loss during combustion to total mass in the input coal.

Four binary mixtures of O<sub>2</sub>/N<sub>2</sub> and O<sub>2</sub>/CO<sub>2</sub> and several ternary mixtures of O<sub>2</sub>/N<sub>2</sub>/H<sub>2</sub>O(v) and O<sub>2</sub>/CO<sub>2</sub>/H<sub>2</sub>O(v) were employed to study the behaviour of the coals. Thus, for the ignition and combustion tests, air (21%O<sub>2</sub>/79%N<sub>2</sub>) was taken as reference and three binary mixtures of O<sub>2</sub> and CO<sub>2</sub> were compared: 21%O<sub>2</sub>/79%CO<sub>2</sub>, 30%O<sub>2</sub>/70% CO<sub>2</sub> and 35%O<sub>2</sub>/65%CO<sub>2</sub>. The addition of 5, 10 and 20% of steam was also evaluated for all the air and the oxy-fuel combustion atmospheres, as a substitute for N<sub>2</sub> or CO<sub>2</sub> respectively, in order to study the effect of the wet recirculation of flue gas upon coal ignition and oxy-fuel combustion.

### **3. Results and discussion**

#### *3.1. Ignition tests*

The ignition temperatures of coal BA and semianthracite HVN under the different atmospheres studied are shown in Fig. 2. The ignition of HVN took place at higher temperatures than that of BA in all the atmospheres studied, in accordance with coal rank (Table 1). The reactivity of the high-volatile bituminous coal, BA, was higher than that of the semi-anthracite, HVN, which caused the heat to be released earlier and led to a reduction in the ignition temperatures.

In the case of HVN, when  $N_2$  (21% $O_2$ -79% $N_2$ ) was replaced by  $CO_2$  (21% $O_2$ -79% $CO_2$ ), the ignition temperature increased (Fig. 2a). This can be attributed to the higher specific molar heat of  $CO_2$  compared to that of  $N_2$ . Before ignition, the coal particles were heated up by the ambient environment, after which the release of coal volatiles occurred, the radiant heat from the particles causing an increase in the gas temperature. The fact that the specific molar heat of  $CO_2$  is higher than that of  $N_2$  would produce a comparatively lower gas temperature and, therefore, a reduction in coal particle temperature during oxy-fuel combustion in comparison to combustion in air at the same oxygen concentration (21%). The result of this would be a decrease in the burning rate and a delay in the ignition of volatiles and char under a 21% $O_2$ -79% $CO_2$  atmosphere, as pointed out by Liu et al. [21] and Molina and Shaddix [10].

However, when the oxygen concentration was increased (30% $O_2$ -70% $CO_2$  and 35% $O_2$ -65% $CO_2$ ), the ignition temperature decreased to below that of the air conditions (Fig. 2a). If the  $O_2$  concentration increases, the mass flux of  $O_2$  to the coal surface particles, the rate of devolatilization and the oxidation rate of volatiles will increase. This shortens the coal particle auto-ignition time considerably [14]. In contrast a reduction in oxygen concentration retards the ignition of volatile matter and char [22].

Fig. 2b shows the ignition temperatures for coal BA under the different atmospheres studied. It can be observed that, when  $N_2$  was replaced by  $CO_2$  in the atmosphere with 21%  $O_2$ , the ignition temperature also increased, indicating a delay in ignition under this oxy-fuel combustion atmosphere due to the lower gas temperatures caused by the higher specific molar heat of  $CO_2$ . It should be noted that a marked difference was observed in the ignition temperatures under both atmospheres, possibly due to the high volatile

content of coal BA (Table 1), since CO<sub>2</sub> decreases the rate of devolatilization because of the lower mass diffusivity of the volatiles in CO<sub>2</sub> mixtures [9].

On the other hand, when the oxygen concentration was increased, the ignition temperature decreased to lower values than that in air (Fig. 2b). Nevertheless, it can be seen that the ignition temperatures of coal BA (without steam addition) under the 30%O<sub>2</sub>/70%CO<sub>2</sub> and 35% O<sub>2</sub>/70%CO<sub>2</sub> atmospheres were very close to that reached under air conditions. The high volatile content of coal BA may have caused a substantial accumulation of volatiles in the vicinity of the particles prior to their ignition, which in turn may have affected the availability of oxygen [14]. If the cloud of volatiles persisted on the char surface for a long time, this would cause O<sub>2</sub> depletion on the char surface, decreasing the rate of devolatilization and delaying the ignition process. Stanmore et al. [23] also observed serious oxygen depletion in the volatile cloud during coal combustion in a drop tube furnace. This behaviour was observed in this work after N<sub>2</sub> was replaced with CO<sub>2</sub> at different O<sub>2</sub> concentrations for the HVN and BA samples both with and without steam addition (Fig. 2).

On the other hand, Fig. 2 shows that the replacement of N<sub>2</sub> or CO<sub>2</sub> by steam caused a slight increase in the ignition temperatures in all the atmospheres for HVN, but no significant differences were observed between the atmospheres with 10 and 20% of steam. In the case of the BA coal, the ignition temperatures increased after the addition of steam to the atmospheres with 21% of O<sub>2</sub>, whereas no differences were observed when steam was added to the oxy-fuel atmospheres with 30 and 35% of O<sub>2</sub>. Nozaki et al. [18] found that the flame temperatures were lower when a wet recycle flue gas (16% of H<sub>2</sub>O) was used compared to a dry recycle flue gas, and they concluded that drying the gas improved ignition stability. In our work, it was the addition of steam that



probably led to lower temperatures, causing an increase in the ignition temperatures under atmospheres containing significant water vapour content.

### 3.2. Combustion tests: Burnout

Coals HVN and BA were burned under different levels of excess oxygen for each atmosphere studied. The fuel ratio, defined as the ratio between the coal mass flow rate and the stoichiometric value, was used to determine the excess oxygen during combustion.

The HVN and BA burnouts, with and without steam addition, are shown in Fig. 3 and 4, respectively. The coal burnout decreased as the fuel ratio increased due to the lower availability of oxygen at higher fuel ratio values. At low fuel ratio values (high oxygen excess), the BA burnout curves showed an asymptotic approach towards a value of 100% (Fig. 4). However, the HVN burnout showed an almost linear dependence on the fuel ratio in both air and oxy-fuel conditions (Fig. 3). Even at low values of fuel ratio (high excess oxygen), the HVN coal showed low burnout values, reflecting the lower reactivity of high rank coals.

For the HVN coal (Fig. 3), both with and without steam addition, the burnout obtained under the 21%O<sub>2</sub>-79%CO<sub>2</sub> atmosphere was lower than that reached under 21%O<sub>2</sub>-79%N<sub>2</sub> conditions. Liu et al. [21] observed that, when air was replaced by 21%O<sub>2</sub>/79%CO<sub>2</sub>, gas temperatures dropped significantly. As mentioned above, CO<sub>2</sub> has a higher specific molar heat than N<sub>2</sub>, which implies that when N<sub>2</sub> is replaced by CO<sub>2</sub> the heat capacity of the gases increases, leading to lower flame and gas temperatures. According to Zhang et al. [14], the specific heat capacity of diluent gas is one of the principal factors affecting char surface temperature for any given O<sub>2</sub> fraction. Therefore,

the particle temperature during the 21%O<sub>2</sub>-79%CO<sub>2</sub> atmosphere can be expected to be lower, causing the combustion rate of the char and the coal burnout value to fall [24]. Bejarano and Levendis [4], when performing combustion experiments in a drop tube furnace, found that coal particles burned at higher temperatures and shorter combustion times in O<sub>2</sub>/N<sub>2</sub> than in O<sub>2</sub>/CO<sub>2</sub> environments for equivalent oxygen fractions. In this work, the ignition temperature under the 21%O<sub>2</sub>-79%CO<sub>2</sub> atmosphere was lower than in air, resulting in delayed ignition and a longer combustion time.

Li et al. [13] studied coal combustion characteristics under oxy-fuel and air conditions using a drop tube furnace. These authors attributed the different behaviours under both atmospheres to the fact that the lower diffusivity of O<sub>2</sub> in CO<sub>2</sub> than in N<sub>2</sub> affects the transport of O<sub>2</sub> to the surface of the particles, leading to a reduced combustion of the volatile matter released from the coal particle and reduced char combustion rates under oxy-fuel conditions. Furthermore, once a coal is ignited, the flame formed provides a vast heat release that consumes the extra volatiles evolved and the char particles. However, the diffusivity of the fuel vapour or small hydrocarbons throughout the volatile cloud and CO<sub>2</sub> boundary layer would be retarded when the N<sub>2</sub> is replaced by CO<sub>2</sub>, thereby reducing the rate at which the volatiles are consumed [14].

Under the 30%O<sub>2</sub>-70%CO<sub>2</sub> and 35%O<sub>2</sub>-65%CO<sub>2</sub> atmospheres, the burnout of HVN was higher than in air (Fig. 3), since the higher oxygen concentration produced an increase in the char combustion rate. Though the gas temperature increases only slightly when the O<sub>2</sub> fraction in bulk gas is increased, it is likely that the increase in the mass flux rate of O<sub>2</sub> from the bulk gas to the coal surface at higher O<sub>2</sub> concentrations promotes the consumption rate of the volatiles [9], providing extra heat feedback to the coal particle to enhance its devolatilization, ignition and combustion [24]. Liu et al. [21] found that

coal combustion in 30%O<sub>2</sub>/70%CO<sub>2</sub> matched the gas temperature profile of coal combustion in air but that it showed a better char burnout.

Increasing the O<sub>2</sub> fraction in CO<sub>2</sub> up to 30% is still insufficient to match the specific heat capacity of air. However, coal burnout in the 30%O<sub>2</sub>-70%CO<sub>2</sub> atmosphere reached a higher value than in air, which means that another parameter must have changed to offset the negative effect of the specific heat capacity of the gas. According to Zhang et al. [14], in atmospheres containing CO<sub>2</sub>, the extra CO derived from char-CO<sub>2</sub> gasification increases the reactivity of the local mixture of fuel and oxidizer gas, which may account for the explanation of the increase in the coal burnout. Homogeneous oxidation of the gasification-derived CO would generate extra heat, which would in turn increase the adiabatic flame temperature and the oxidation of both the volatiles and the char. Zhang et al. [14] underlined the importance of the extra CO and its homogeneous oxidation in the vicinity of the char particle, as it would enhance thermal feedback to the char particles, in the same way as volatile oxidation does. The char gasification reaction would also be beneficial for coal conversion.

For coal BA (Fig. 4), both with and without the addition of steam, no significant differences can be observed between the different atmospheres studied because BA reached a very high burnout under all conditions due to its high reactivity.

In order to facilitate the comparison of the behaviours of these coals under air and oxy-fuel atmospheres both with and without steam addition, the burnouts were interpolated at a fuel ratio of 0.8 using the curves shown in Fig. 3 and 4. The results for both coals, HVN and BA, are shown in Fig. 5a and Fig. 5b, respectively. It can be seen that the burnout of HVN in air and under an oxy-fuel atmosphere with 21% of oxygen, decreased slightly after the addition of steam, but no relevant differences were found

between the three steam concentrations (Fig. 5a). The lower burnout observed after the addition of steam seems to indicate the combustion temperature under these atmospheres was lower. The higher specific molar heat of  $\text{H}_2\text{O}(\text{v})$  compared to  $\text{N}_2$  would explain the decrease in burnout after the addition of steam to the air atmosphere. However, under oxy-fuel atmospheres, although the specific molar heat of water vapour is lower than that of  $\text{CO}_2$ , no increase in burnout was observed when steam was added to the 21% $\text{O}_2$ -79% $\text{CO}_2$  atmosphere. This suggests that other factors must have influenced oxy-fuel combustion with the addition of steam. Thus, enhancements in thermal radiation or endothermic radical formation (O, OH, H, etc.) might explain the reduction in combustion temperature [25]. High proportions of  $\text{H}_2\text{O}$  in the furnace gases may have resulted in higher gas emissivities due to the high emission capacity of water vapour via radiative heat transfer. Wall et al. [26] estimated that total emissivity due to the joint emission of the gas and the particulate was higher under wet oxy-fuel conditions than under a dry oxy-fuel atmosphere, and it presented the lowest value in air. Nozaki et al. [18] found that drying the flue gas increased the gas temperature near the burner by about 150 °C for the same volumetric gas flow rate and thus helped to stabilize the flame by improving ignition stability. Zhou and Moyeda [19] by means of a computational fluid dynamics (CFD) simulation showed that the wet flue gas recycle led to a lower flame temperature than the dry flue gas recycle.

Under the oxy-fuel combustion atmospheres with 30 and 35% of oxygen, the addition of steam did not significantly affect coal burnout (Fig. 5a), which was due to the fact that more oxygen was available. As a result, the burnout of HVN was less affected by the presence of low concentrations of steam.

For the BA coal, no different burnout values can be observed between the atmosphere with and without steam (Fig. 5b), because of the high reactivity of this coal.

#### **4. Conclusions**

The ignition temperature and burnout of a semi-anthracite and a high-volatile bituminous coal were studied under oxy-fuel combustion conditions in an entrained flow reactor (EFR). The results showed that the ignition temperature was higher under a 21%O<sub>2</sub>/79%CO<sub>2</sub> atmosphere than in air conditions, whereas the burnout was lower. This was due to the higher specific molar heat of CO<sub>2</sub> compared to N<sub>2</sub> and the lower diffusivity of O<sub>2</sub> in CO<sub>2</sub> than in N<sub>2</sub>. However, when the O<sub>2</sub> concentration was increased to 30 and 35% in the oxy-fuel combustion atmosphere, the ignition temperature was lower than in air, whereas the burnout value was increased above that reached under air-firing. It was due to an increase in the mass flux of O<sub>2</sub> to the coal particles. Finally, higher ignition temperatures and lower coal burnout values were observed after the addition of steam in oxy-fuel combustion atmospheres under the experimental conditions of this study, but no relevant differences were observed between the different steam concentrations. This could have been due to higher losses of heat by thermal radiation.

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### **Figure captions**

Fig. 1. Schematic diagram of the entrained flow reactor (EFR) used in the experiments

Fig. 2. Ignition temperature of HVN (a) and BA (b) coals under different atmospheres (the  $\text{H}_2\text{O}(\text{v})$  is added as a substitute for  $\text{N}_2$  or  $\text{CO}_2$ )

Fig. 3. Burnout of HVN coal under different atmospheres at different fuel ratios

Fig. 4. Burnout of BA coal under different atmospheres at different fuel ratios

Fig. 5. Burnout of HVN (a) and BA (b) coals under different atmospheres at a fuel ratio of 0.8 (the  $\text{H}_2\text{O}(\text{v})$  is added as a substitute for  $\text{N}_2$  or  $\text{CO}_2$ )



Table 1. Proximate and ultimate analyses and high heating value of the coals

Sample	HVN	BA
Origin	Spain	Spain
Rank	sa	hvb
<b>Proximate Analysis<sup>a</sup></b>		
Moisture content (wt.%)	1.1	1.2
Ash (wt.%, db)	10.7	6.9
V.M. (wt.%, db)	9.2	33.9
F.C. (wt.%, db) <sup>b</sup>	80.1	59.2
<b>Ultimate Analysis (wt.%, daf)<sup>a</sup></b>		
C	91.7	88.5
H	3.5	5.5
N	1.9	1.9
S	1.6	1.1
O <sup>b</sup>	1.3	3.0
High heating value (MJ/kg, db)	31.8	33.1

sa: semi-anthracite; hvb: high-volatile bituminous coal.

db: dry basis; daf: dry and ash free bases.

<sup>a</sup> The proximate analysis was conducted in a LECO TGA-601, and the ultimate analysis in a LECO CHNS-932.

<sup>b</sup> Calculated by difference.

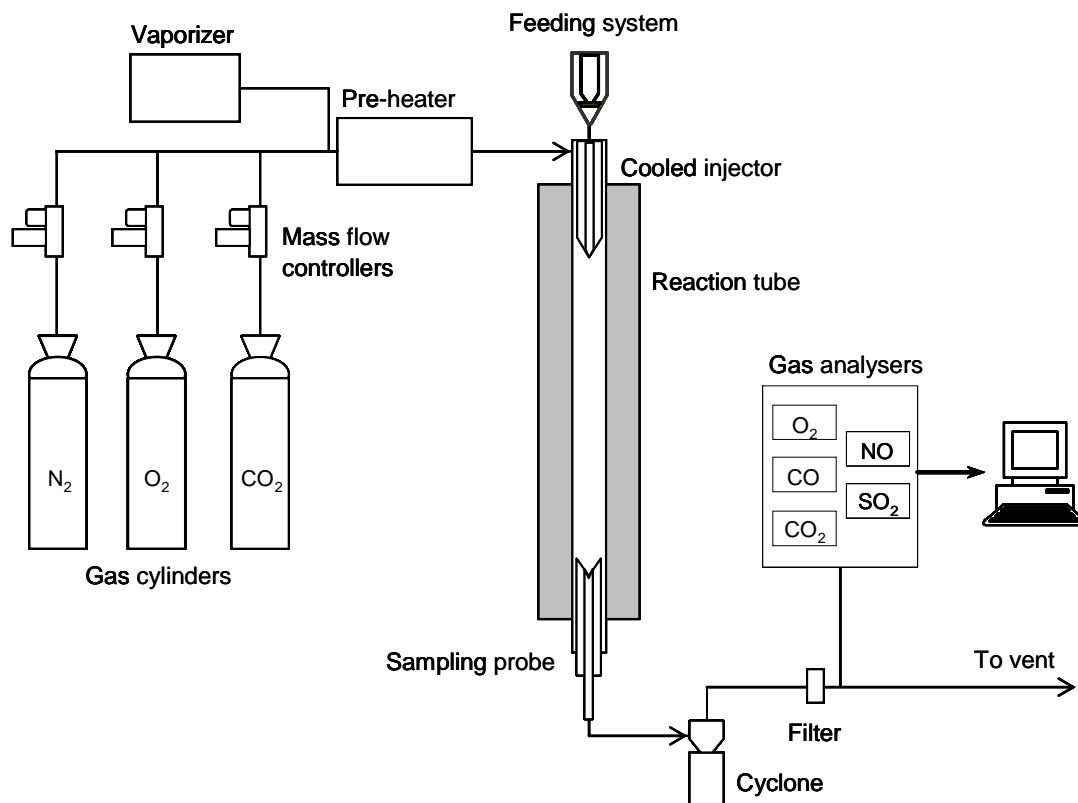


Fig. 1. Schematic diagram of the entrained flow reactor (EFR) used in the experiments

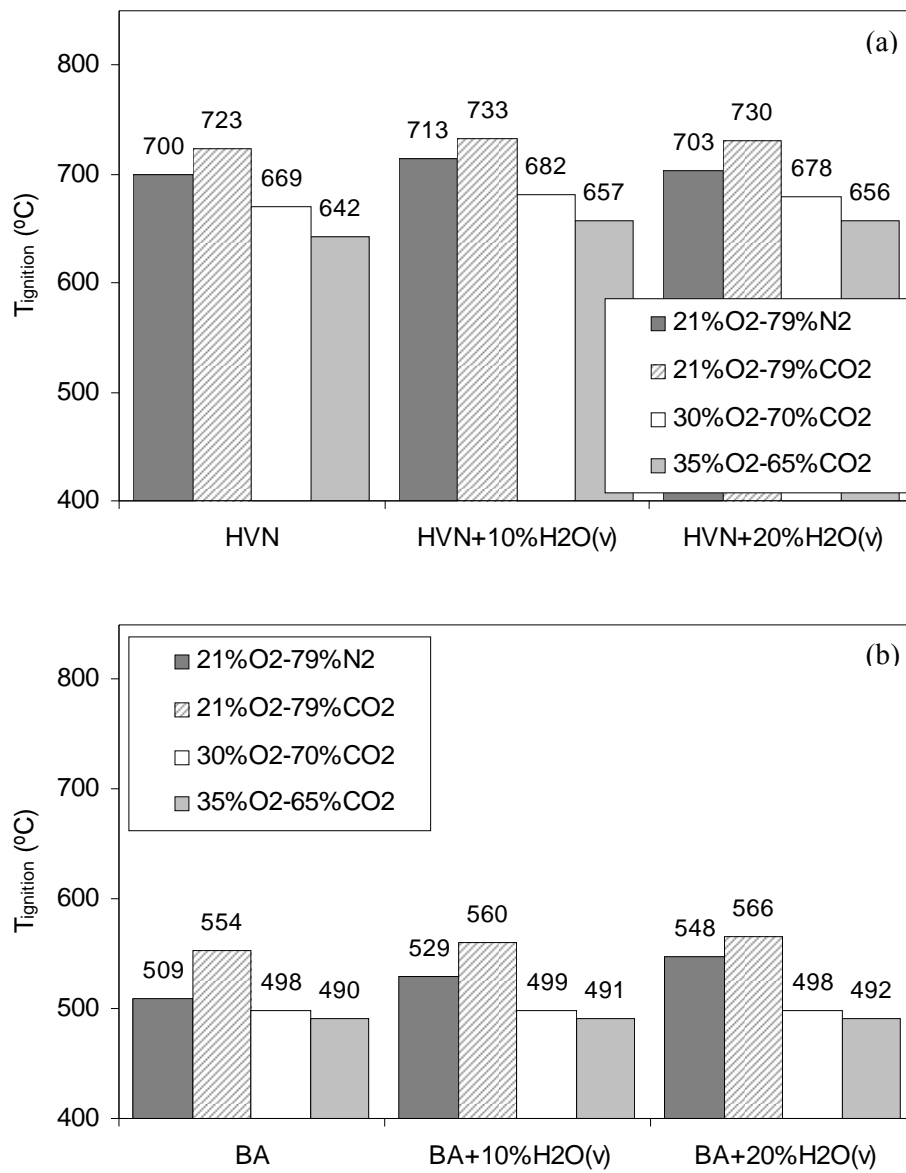


Fig. 2. Ignition temperature of HVN (a) and BA (b) coals under different atmospheres (the  $H_2O(v)$  is added as a substitute for  $N_2$  or  $CO_2$ )

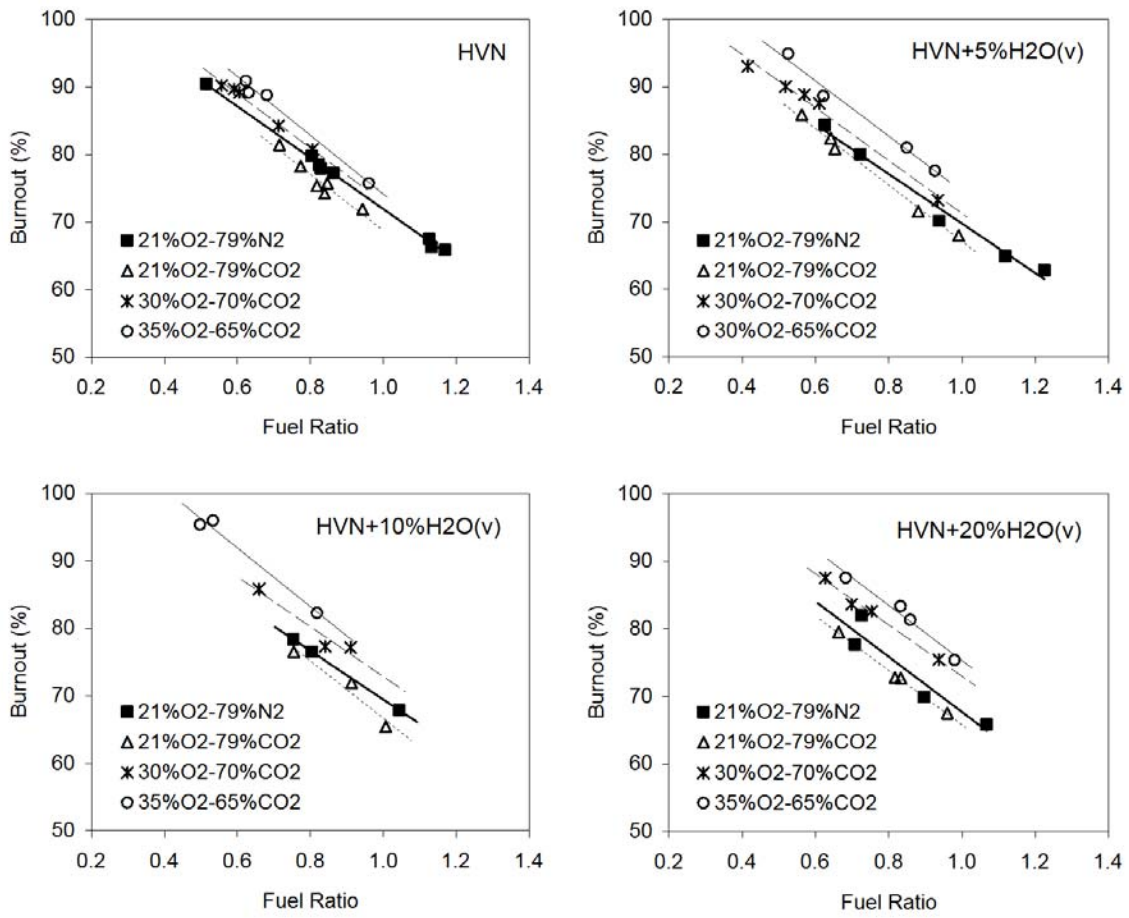


Fig. 3. Burnout of HVN coal under different atmospheres at different fuel ratios

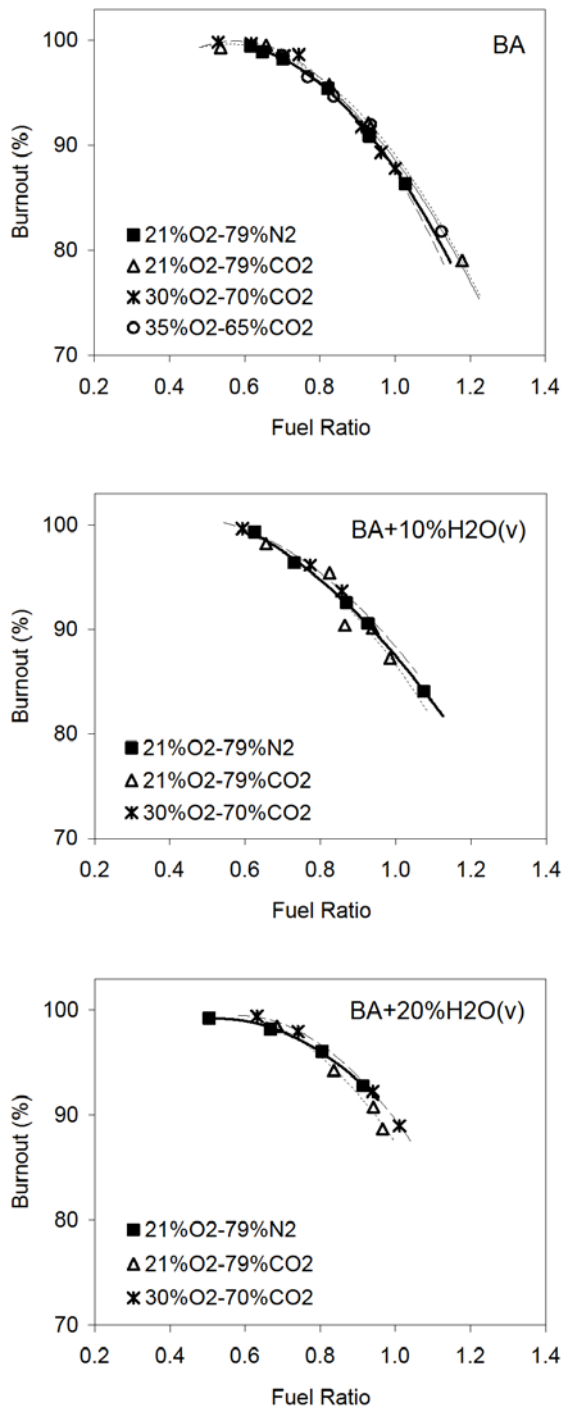


Fig. 4. Burnout of BA coal under different atmospheres at different fuel ratios

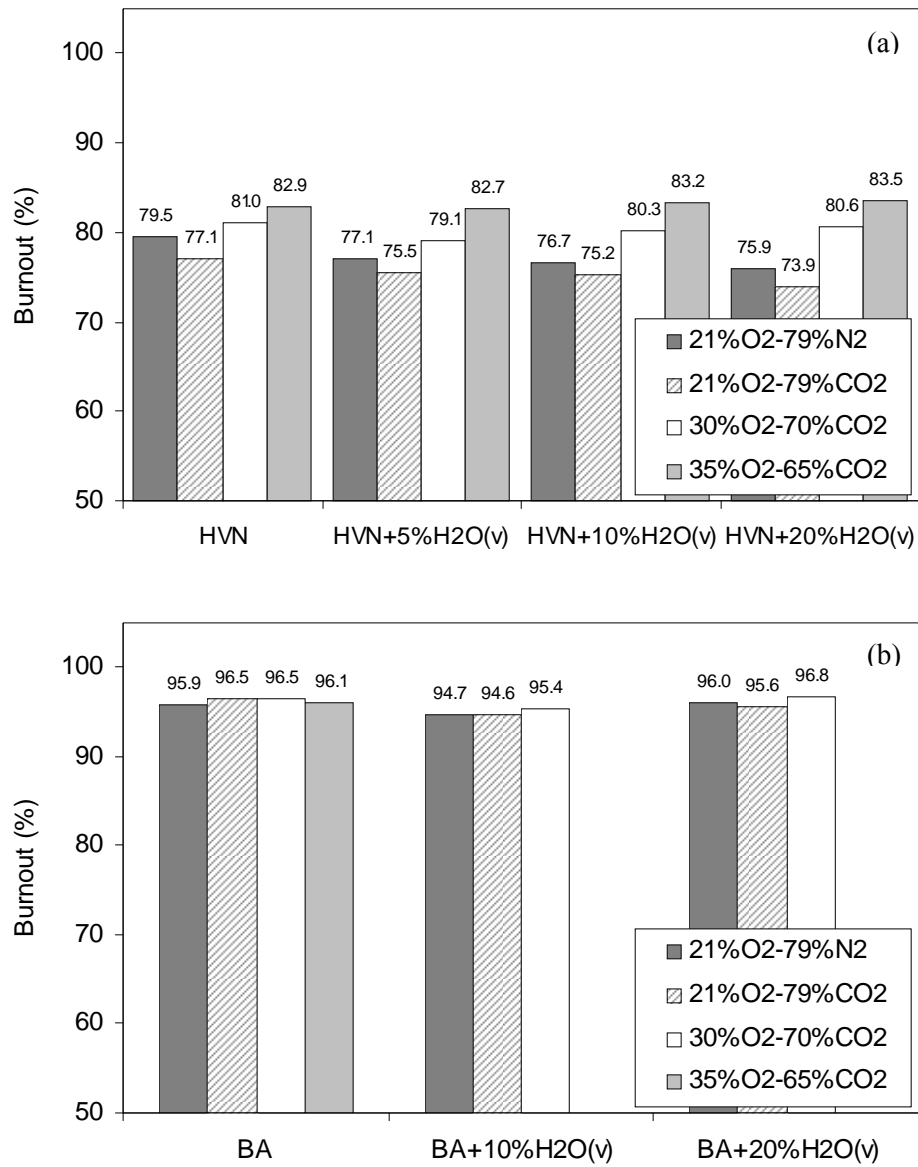


Fig. 5. Burnout of HVN (a) and BA (b) coals under different atmospheres at a fuel ratio of 0.8 (the  $H_2O(v)$  is added as a substitute for  $N_2$  or  $CO_2$ )