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A study of oxy-coal combustion with steam addition and biomass blending by thermogravimetric analysis

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Abstract The thermal characteristics of pulverized coal have been studied under oxy-fuel combustion conditions using non-isothermal thermogravimetric analysis (TG). The atmospheres used were $21\%O_2/79\%N_2$, $21\%O_2/79\%CO_2$, $30\%O_2/70\%O_2$, and $35\%O_2/65\%CO_2$. Coal blends of coal with 10 and 20% of biomass were also studied under these atmospheres. The addition of 10 and 20% of steam was evaluated for the oxy-fuel combustion atmospheres with 21 and 30% of O_2 in order to study the effect of the wet recirculation of flue gas. The results obtained were similar for all the different rank coals and indicated that replacing N_2 by CO_2 in the combustion atmosphere with 21% of O_2 caused a slight decrease in the rate of mass loss and delayed the burning process of the coal, biomass and coal/biomass blend samples. When the O_2 concentration was increased to 30 and 35% in the oxy-fuel combustion atmosphere, the rate of mass loss increased, the burning process occurred at lower temperatures and it was shorter in duration. An increase in the rate of mass loss and a reduction in burning time and temperature were observed after the addition of steam to the oxy-fuel combustion atmosphere. No relevant differences between the 10 and 20% steam concentrations were observed.

Keywords Oxy-fuel combustion · Pulverized coal · Biomass · Steam · TG

Introduction

Interest in coal as an energy source for the future has recently been revived due to the stability of its supply and it is relatively low cost. For these reasons, it is likely that coal will occupy an important position in the energy mix in the foreseeable future [1, 2]. However, the emissions of greenhouse gas carbon dioxide from coal-fired power plants are very high and their effect on global climate change has been acknowledged worldwide. Various strategies for their reduction and sequestration are under study, including improvements in the efficiency of existing power plants, the introduction of advanced combined cycle power plants and the capture and storage of CO_2 [3].

Carbon capture and storage (CCS) is considered one of the most realistic options for reducing CO_2 emissions from fossil fuel use, particularly from large-scale stationary power plants. The main problem, however, is that the combustion of fossil fuel in air leads to a diluted concentration of CO_2 in the flue gas stream (typically 15% by volume), rendering it unsuitable for direct sequestration in a supercritical state by compression, a process which requires a high concentration of CO_2 [4, 5].

As an alternative, oxy-fuel combustion is considered as a promising new approach for CO_2 capture. Oxy-fuel combustion technology involves the combustion of pulverized coal in a mixture of oxygen (as opposed to air) and recirculated flue gas (mainly CO_2 and H_2O). The advantages of this approach are that it offers a way of avoiding the excessively high flame temperatures characteristic of fuel combustion in oxygen, carries the heat through the boiler, reduces the net volume of flue gas and substantially increases the concentration of carbon dioxide in the flue gas. A flue gas consisting mainly of CO_2 (80–95% by volume) and water vapour is then generated. The water

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vapour can be easily removed by condensation, making it easier to purify the remaining CO_2 [6].

The successful implementation of O_2/CO_2 technology in conventional pulverized coal boilers depends on a full understanding of the changes that result from replacing N_2 with CO_2 in the oxidizer stream [7]. Although many published works have focused on the oxy-fuel combustion process [8–12], the effect of adding steam to the oxy-fuel combustion atmosphere has hardly been studied [13].

Alternatively, biomass is a renewable fuel which can be used for reducing CO_2 emissions. This source of energy is considered carbon neutral because the carbon dioxide released during biomass utilization is recycled as an integral part of the carbon cycle. The combination of oxy-fuel combustion with biomass could be used as a sink for CO_2, and therefore more knowledge about cofiring coal and biomass under oxy-fuel conditions is needed [14].

Thermogravimetric analysis (TG) is one of the most common techniques used to rapidly investigate and compare thermal events during the combustion and pyrolysis of solid raw materials, such as coal, woods, etc. [15–22]. Although extrapolation to other devices at a larger scale cannot be performed directly, the information obtained from the combustion profiles in the TG could be used for an initial evaluation of the behaviour of the combustion on an industrial scale [23]. This would contribute to an understanding of coal combustion behaviour and prove very useful both from a fundamental viewpoint and for the comparison of samples [24].

In this work, TG was used in order to study the effect of replacing N_2 with CO_2 in a combustion atmosphere and of enhanced levels of O_2 in an oxy-fuel combustion atmosphere upon the thermal characteristics of different rank coals and coal/biomass blends. Oxy-fuel combustion with steam addition was also evaluated in order to study the effect of the wet recirculation of flue gas.

Materials and methods

Materials

Ten coals of different rank [25] were used in this work: an anthracite (AC), a semi-anthracite (HVN), two medium-volatile bituminous coals (UM and SAA), and six high-volatile bituminous coals (DAB, SAB, M6N, BA, CAB, and NZ). A biomass, olive waste (OW), was also employed. The samples were ground and sieved to obtain a particle size fraction of 75–150 μm. The proximate analysis and the high heating value of the samples are presented in Table 1.

Apparatus and procedure

The techniques employed in this study were TG and derivative thermogravimetry (DTG). Non-isothermal TG was performed using a Setaram TAG24 analyser. Approximately, 5 mg of sample was used for each experiment. A small amount of sample and a slow heating rate were used to avoid heat transfer limitations and to minimize mass transfer effects. The samples were heated at a heating rate of 15 °C min^{-1} in different atmospheres from room temperature to 1000 °C. The simulated air combustion atmosphere was 21%O_2/79%N_2. Three O_2/CO_2 oxy-fuel combustion conditions were used (21, 30, and 35% of O_2). The addition of 10 and 20% of steam (as a replacement for CO_2) to an oxy-fuel combustion atmosphere containing 21 and 30% of O_2 was also evaluated in order to

Table 1 Proximate analysis and high heating value of the coals

Sample	AC	HVN	UM	SAA	DAB	SAB	M6N	BA	CAB	NZ	OW
Origin	Spain	Spain	Mexico	South Africa	China	South Africa	Mexico	Spain	Colombia	New Zealand	Spain
Rank	an	sa	mvb	mvb	hvb	hvb	hvb	hvb	hvb	hvb	–
Proximate analysis[a]											
Moisture content/wt%	2.3	1.1	0.4	2.6	2.9	2.4	1.8	1.2	4.3	11.5	9.2
Ash/wt%, db	14.2	10.7	21.1	14.9	10.9	15.0	30.2	6.9	4.4	2.9	7.6
V.M./wt%, db	3.6	9.2	23.7	25.6	28.8	29.9	30.6	33.9	39.6	47.9	71.9
F.C./wt%, db[b]	82.2	80.1	55.2	59.5	60.3	55.1	39.2	59.2	56.0	49.2	20.5
High heating value/kJ kg^{-1}, db	29160	31818	27826	27952	28825	27780	23057	33081	30965	27943	19905

an anthracite, *sa* semi-anthracite, *mvb* medium-volatile bituminous coal, *hvb* high-volatile bituminous coal, *db* dry basis

[a] The proximate analysis was conducted in a LECO TGA-601

[b] Calculated by difference

study the effect of the wet recirculation of flue gas. The total flow rate of all the mixtures of gases was 150 cm^3 min^{-1}. The steam generator consisted of a CEM® (Controlled Evaporator and Mixer), in which water and O_2/CO_2 were mixed and heated up to 175 °C. Liquid and mass flow controllers were used to control the flow rates of the water, O_2, CO_2, and N_2. The derivative curves (DTG) of the samples were represented as a function of temperature.

Determination of the combustion parameters

From the combustion profiles, the following thermal parameters were calculated for characterizing the combustion process: initial temperature (T_i), final temperature (T_f), peak temperature (T_{peak}), temperature corresponding to a conversion degree of 50% (T_{50}), maximum rate of mass loss (DTG_{max}), average rate of mass loss (Average DTG), and coal reactivity index (R_C). The initial temperature (T_i) and the final temperature (T_f) were taken as the temperature values (after the initial loss of moisture) at which the rate of mass loss was 1% min^{-1}. The peak temperature (T_{peak}) corresponds to the temperature at which maximum rate of mass loss occurs [26]. The temperature value at the maximum rate of mass loss is usually considered inversely proportional to the reactivity and combustibility of the sample [27], whereas the maximum rate of mass loss is considered directly proportional to the reactivity of the sample [28]. The average rate of mass loss (Average DTG) is the ratio between the burnout fraction and time during the temperature range of initial to final temperature. The coal reactivity index (R_C) was used to evaluate the burning performance of pulverized coal, defined as:

$$R_C = 1/m_0(\mathrm{d}m/\mathrm{d}t)_{max} \qquad (1)$$

where $(\mathrm{d}m/\mathrm{d}t)_{max}$ is the maximum combustion rate, and m_0 is the initial mass of ash free sample. It should be noted that the greater the R_C value, the higher the combustion reactivity.

Results and discussion

Combustion characteristics of pulverized coal under air and O_2/CO_2 atmospheres

DTG curves for the AC, HVN, DAB, and NZ coal samples under the different combustion and oxy-fuel combustion atmospheres are shown as example in Fig. 1. The DTG curves for the other coals (not represented) showed similar behaviour. From the combustion profiles (Fig. 1), it can be seen that coal combustion in the O_2/N_2 atmosphere differed from that in the O_2/CO_2 atmosphere at the same oxygen concentration (21%). For all the coals studied in the $21\%O_2/79\%CO_2$ atmosphere, the curves of mass loss rate shifted to higher temperatures in relation to those in the $21\%O_2/79\%N_2$ conditions. This implies that there was a delay in the burning process, which may have been due to the higher specific heat of CO_2 compared to N_2, leading to comparatively lower gas temperatures and in turn to a reduction of the fuel particle temperatures during oxy-fuel combustion in comparison to combustion in air. Li et al. [6] found lower rates of mass loss and higher burning times for a bituminous coal after replacing N_2 with CO_2 from an air atmosphere, indicating that the devolatilization rate of coal was lower under a CO_2 atmosphere than under a N_2 atmosphere. These authors thus concluded that replacing N_2 with CO_2 was unfavourable to coal combustion.

On the other hand, if the oxygen concentration was increased under the oxy-fuel combustion atmospheres ($30\%O_2/70\%CO_2$ and $35\%O_2/65\%CO_2$), the DTG curves moved to lower temperatures, and the rate of mass loss increased whereas the burning time decreased in comparison to the values achieved in the $21\%O_2/79\%N_2$

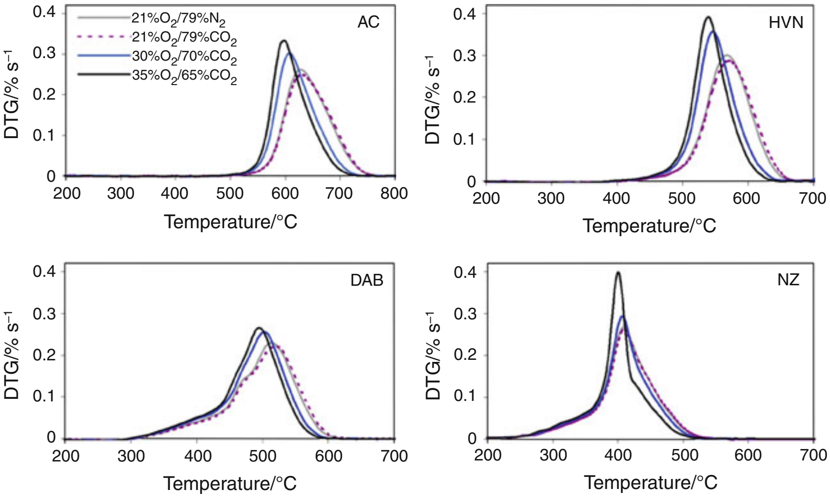

Fig. 1 DTG curves for coal combustion under different atmospheres

combustion atmosphere (Fig. 1). This may be because a higher oxygen concentration enhances the devolatilization and combustion of coal samples. Under a lower oxygen concentration, the ignition of both the volatile matter and char would be retarded [8].

Table 2 summarizes the combustion parameters for the AC, HVN, DAB, and NZ coals, whose results are similar to those of the other coals studied. It can be seen that the initial temperature, T_{i}, for the coal samples was similar or slightly higher under oxy-fuel combustion atmosphere with 21% of O_2 compared to air. However, T_{i} decreased as the O_2 concentration increased in the O_2/CO_2 atmosphere. The same tendency is observed for the final temperature, T_{f}, the peak temperature, T_{peak}, and the temperature corresponding to a conversion degree of 50%, T_{50}. In contrast, the Average DTG and $\mathrm{DTG_{max}}$ values followed the opposite tendency. They were slightly lower under the air atmosphere than under the oxy-fuel atmosphere containing 21% of O_2, but they increased as the O_2 concentration increased in the O_2/CO_2 atmosphere. Table 2 also shows that, in general, the coal reactivity index (R_{C}) in the $21\%O_2/79\%CO_2$ atmosphere was slightly lower than that in the $21\%O_2/79\%N_2$ atmosphere, but was higher in the $30\%O_2/70\%CO_2$ and $35\%O_2/65\%CO_2$ atmospheres, these two atmospheres showing similar values of coal reactivity.

If the characteristic temperatures for all the different rank coals are compared, the order is the following: AC > HVN > UM = BA > SAB = SAA ≥ DAB > CAB ≥ M6N > NZ. This order approximately coincides with that of coal rank (Table 1).

Combustion characteristics of biomass and coal/biomass blends under air and O_2/CO_2 atmospheres

Figure 2 shows the DTG curves for the biomass sample, OW, and the coal/biomass blends under the oxy-fuel combustion atmospheres. From the combustion profiles (Fig. 2), it can be seen that the combustion of the OW, 90%HVN + 10%OW, and 80%HVN + 20%OW samples in the O_2/N_2 atmosphere differed from that of the O_2/CO_2 atmosphere at the same oxygen concentration (21%). As in the case of the coals, in the $21\%O_2/79\%CO_2$ atmosphere, the curves of mass loss rate shifted to higher temperatures in relation to those in the $21\%O_2/79\%N_2$ conditions, indicating that there was a delay in the combustion process. However, for the biomass and coal/biomass blend samples, when the oxygen concentration was increased under the oxy-fuel combustion atmospheres ($30\%O_2/70\%CO_2$ and $35\%O_2/65\%CO_2$), the DTG curves moved to lower temperatures, the rate of mass loss increased and the burning time decreased with respect to the corresponding parameters in the $21\%O_2/79\%N_2$ combustion atmosphere.

Figure 2 also shows that an initial mass loss occurred between 25–105 °C for all the samples, due to moisture evaporation, after which two mass loss stages occurred in the OW and blend samples, compared to a one stage mass loss for the HVN sample (Fig. 1). The second mass loss stage in the OW sample was possibly due to the release of volatiles and their combustion, while the final mass loss may have been due to char oxidation. In the case of the blends, the second mass loss was very small, due to the low

Table 2 Combustion parameters of the coals under different atmospheres

Sample	Atmosphere	T_{i}/°C	T_{f}/°C	T_{peak}/°C	T_{50}/°C	$\mathrm{DTG_{max}}$/% s^{-1}	Average DTG/% s^{-1}	R_{C}/% kg^{-1} s^{-1}
AC	$21\%O_2/79\%N_2$	560	732	625	634	0.261	0.141	53996
	$21\%O_2/79\%CO_2$	561	734	633	636	0.248	0.139	50849
	$30\%O_2/70\%CO_2$	545	707	608	613	0.303	0.146	65117
	$35\%O_2/65\%CO_2$	540	696	600	603	0.333	0.154	71177
HVN	$21\%O_2/79\%N_2$	482	644	567	566	0.301	0.149	60252
	$21\%O_2/79\%CO_2$	484	650	570	569	0.287	0.147	55352
	$30\%O_2/70\%CO_2$	470	627	547	547	0.358	0.157	78894
	$35\%O_2/65\%CO_2$	465	616	540	539	0.392	0.163	77706
DAB	$21\%O_2/79\%N_2$	357	588	516	499	0.229	0.105	48817
	$21\%O_2/79\%CO_2$	360	593	520	503	0.222	0.103	45983
	$30\%O_2/70\%CO_2$	351	576	501	490	0.255	0.109	55768
	$35\%O_2/65\%CO_2$	346	567	495	483	0.265	0.111	56410
NZ	$21\%O_2/79\%N_2$	293	504	410	405	0.267	0.104	49585
	$21\%O_2/79\%CO_2$	294	506	411	405	0.260	0.103	49821
	$30\%O_2/70\%CO_2$	288	497	406	400	0.293	0.105	57537
	$35\%O_2/65\%CO_2$	283	486	395	393	0.399	0.107	73462

T_i initial temperature, T_f final temperature, T_{peak} peak temperature, T_{50} temperature corresponding to a conversion degree of 50%, DTG_{max} maximum rate of mass loss, *Average DTG* average rate of mass loss, R_C coal reactivity index

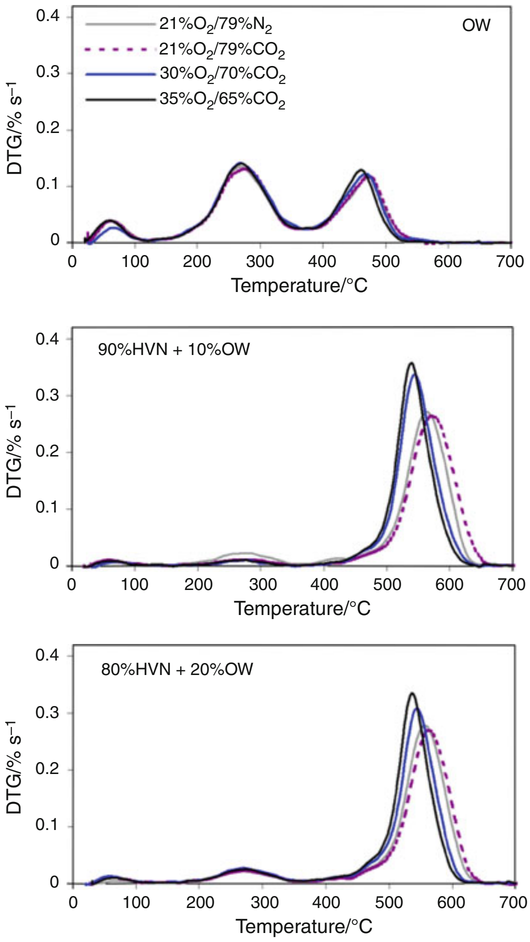

Fig. 2 DTG curves for the combustion of biomass and coal/biomass blends under different atmospheres

proportion of biomass in the blend, the last peak closely resembling that of the coal sample, presumably because coal was the predominant component of the blend.

The combustion parameters for these samples are summarized in Table 3. In the case of the OW and blend samples, the parameters were calculated only taking into consideration the last mass loss stage, so that they could be then compared with those of the HVN coal sample. For all the combustion and oxy-fuel combustion atmospheres, it can be observed that the highest values of T_i, T_f, T_{50}, DTG$_{max}$ and Average DTG corresponded to the HVN samples, followed by those of the 90%HVN + 10%OW and the 80%HVN + 20%OW samples in that order. Small differences were found in the T_{peak} and R_C values of the samples. The addition of biomass to the coal caused a decrease in the combustion and oxy-fuel combustion temperatures. Biomass is a fuel with high reactivity and high volatile matter content (Table 1), which can lead to a faster reaction and to an improvement in the combustion behaviour of a coal in agreement with the results of other works [18, 29, 30].

Effects of steam addition upon the oxy-fuel combustion characteristics of pulverized coal

Figure 3 compares the DTG curves for the AC, HVN, DAB, and NZ coal samples under the 21%O_2/79%CO_2 and 30%O_2/70%CO_2 oxy-fuel combustion atmospheres with those corresponding to the same atmosphere after the replacement of CO_2 with 10 and 20% of steam. It can be observed that the addition of steam shifted the DTG curves

Table 3 Combustion parameters of the biomass and the coal/biomass blends under different atmospheres

Atmosphere	Sample	T_i/°C	T_f/°C	T_{peak}/°C	T_{50}/°C	DTG$_{max}$/% s^{-1}	Average DTG/% s^{-1}	R_C/% kg^{-1} s^{-1}
21%O_2/79%N_2	HVN	482	644	567	566	0.301	0.149	60252
	90%HVN-10%OW	465	641	565	561	0.282	0.132	62291
	80%HVN-20%OW	455	623	559	546	0.278	0.129	58044
	OW	367	520	475	302	0.118	0.064	24636
21%O_2/79%CO_2	HVN	484	650	570	569	0.287	0.147	55352
	90%HVN-10%OW	469	645	570	564	0.264	0.130	54590
	80%HVN-20%OW	460	628	565	552	0.269	0.127	55948
	OW	370	522	475	305	0.118	0.064	23807
30%O_2/70%CO_2	HVN	470	627	547	547	0.358	0.157	78894
	90%HVN-10%OW	453	619	545	542	0.337	0.142	69394
	80%HVN-20%OW	452	608	545	535	0.308	0.131	61221
	OW	371	514	467	308	0.122	0.067	31640
35%O_2/65%CO_2	HVN	465	616	540	539	0.392	0.163	77706
	90%HVN-10%OW	453	611	540	536	0.358	0.145	76386
	80%HVN-20%OW	448	601	535	529	0.335	0.137	65442
	OW	365	509	461	302	0.129	0.067	25639

T_i initial temperature, T_f final temperature, T_{peak} peak temperature, T_{50} temperature corresponding to a conversion degree of 50%, *DTG*$_{max}$ maximum rate of mass loss, *Average DTG* average rate of mass loss, R_C coal reactivity index

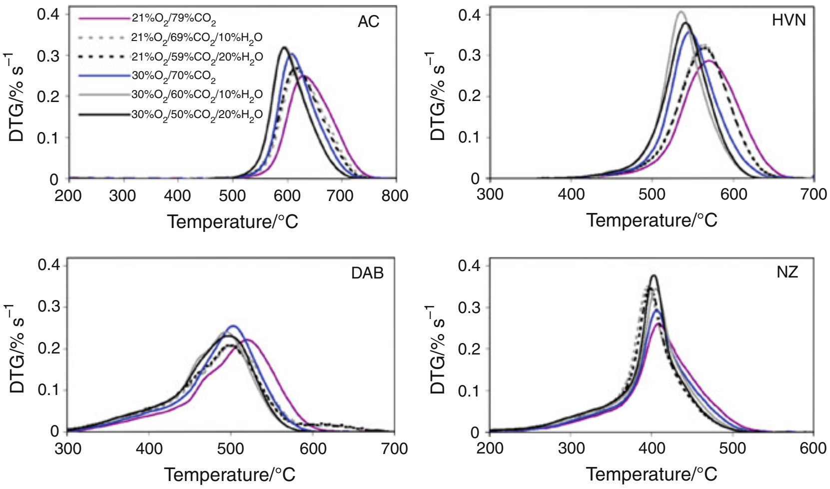

Fig. 3 DTG curves for coal combustion under oxy-fuel atmospheres with 21 and 30% of O_2 containing steam

to lower temperatures and generally caused an increase in the rate of mass loss and a decrease in combustion time with respect to those in the 21%O_2/79%CO_2 or 30%O_2/70%CO_2 oxy-fuel combustion atmospheres, probably due to the fact that the specific heat capacity of H_2O is lower than that of CO_2 [31], as a result of which the combustion stage was brought forward. However, no relevant differences were observed from the addition of 10 and 20% of steam (Fig. 3).

Conclusions

The different coals studied exhibited different combustion behaviours under O_2/CO_2 oxy-fuel conditions with respect to air combustion conditions. The coal, biomass, and coal/biomass blend samples showed slightly higher reactivities under air compared to oxy-fuel conditions at the same O_2 level, since the replacement of O_2 by CO_2 caused the rate of mass loss to decrease and the burning process occurred at higher temperatures. However, once the O_2 concentration had been increased to 30 and 35% in the oxy-fuel combustion atmosphere, the rate of mass loss rose and the burning process was brought forward and shortened. The addition of biomass to coal caused a decrease in the combustion and oxy-fuel combustion temperatures. The replacement of 10 and 20% of CO_2 with steam in the oxy-fuel combustion atmosphere of the coal resulted in an increase in the rate of mass loss and a decrease in the burning time, no significant differences being observed between the two steam concentrations.

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