Ignition behaviour of different rank coals in an entrained flow reactor

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Abstract

An experimental study to determine the temperature and mechanism of coal ignition was carried out by using an entrained flow reactor (EFR) at relatively high coal feed rates (0.5 g min\(^{-1}\)). Seven coals ranging in rank from subbituminous to semianthracite, were tested and the evolved gases (O\(_2\), CO, CO\(_2\), NO) were measured continuously. The ignition temperature was evaluated from the gas evolution profiles, and it was found to be inversely correlated to the reactivity of the coal, as reflected by the increasing values of the ignition temperature in the sequence: subbituminous, high volatile bituminous, low volatile bituminous and semianthracite coals. The mechanism of ignition varied from a heterogeneous mechanism for subbituminous, low volatile bituminous and semianthracite coals, to a homogeneous mechanism for high volatile bituminous coals. A thermogravimetric analyser (TGA) was also used to evaluate coal ignition behaviour. Both methods, TGA and EFR, were in agreement as regards the mechanism of coal ignition. From the SEM micrographs of the coal particles retrieved from the cyclone, it was possible to observe the external appearance of the particles before, during and after ignition. The micrographs confirmed the mechanism deduced from the gas profiles.

Keywords: Ignition; coal; entrained flow reactor; SEM
1. Introduction

Ignition is the substantial burning of a combustible material. This requires that the loss of heat be balanced by the heat release at the ignition temperature [1,2]. 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. The reactivity and ignition behaviour of coal particles is of considerable importance for designing the boiler and controlling the combustion process. Coal particles can ignite either homogeneously, where the initial step is the pyrolysis and ignition of the volatiles followed by the ignition of the char, or heterogeneously, which involves the direct attack of oxygen on the whole sample particle [3,4].

It has been established that neither temperature nor the mechanism of ignition are inherent properties of coal, and that they depend on the sample mass, the heating rate of the particle, the surrounding gas, and the ignition indicator [5]. In general, ignition depends on the type and reaction conditions of the test apparatus [6], and there is no widely accepted method for determining ignition temperatures. This is reflected by the diversity of experimental techniques that have been used to study coal particle ignition [2,5]. Thermogravimetric techniques (TGA/DTG) and drop tube furnaces are among the most common methods used to study coal ignition. Although TGA techniques have been widely employed to determine the ignition behaviour of coal [7-11] they operate at very different conditions to those encountered in a pulverised coal combustor. Thus, other bench equipment such as drop tube furnaces and entrained flow reactors, which simulate more closely the combustion conditions of industrial pf combustors (i.e., continuous coal feeding, particle residence time, heating rate), have been used [12-13]. There are, however, some shortcomings associated with the use of this equipment such as the use of sized fractions of coal or the operation under laminar flow conditions.
Nevertheless, the pilot- and full-scale tests are costly to operate and the above techniques can help in the understanding of coal ignition behaviour.

In addition, different measurement techniques have been employed to determine the ignition temperature of pulverised coal. In this regard, some of the most common approaches used to estimate the temperature of ignition are based on the visual observation of the number of flashes emitted [14], detection of the light emissions with photomultipliers [15], measurement of the particle temperature by means of thermocouples [16], or following the evolution of the gaseous products during coal particle ignition and burnout [17,18].

This work is concerned with determining the temperature and mechanism of ignition of different rank coals under conditions where particle interactions take place. To this end, a laminar entrained flow reactor, operating at high coal feed rates was used in order to promote interactive effects between the particles. Thus, group behaviour rather than single particle behaviour applies [19,20], and this is important for properly approximating the particle interactions that occur in industrial coal combustion and gasification processes. It should be noted that the ignition temperatures reported in this paper correspond to the gas temperature at which ignition takes place. In addition, due to the lack of a standardised methodology for determining coal ignition temperature and the wide variety of equipment and operating conditions used for its evaluation, the values should be regarded as trends rather than absolute values [5].

2. Experimental

An electrically heated, laminar entrained flow reactor with a reaction zone of 170 cm and an internal diameter of 4 cm, capable of reaching a maximum temperature of 1100 °C, was used for the ignition studies. Coal was entrained in the primary carrier gas, at
ambient temperature, by a rotary feed system and introduced into the furnace through an
air-cooled injection probe. The secondary gas was preheated before being introduced
into the furnace through two concentric flow straighteners, located in the annular space
between the injector and the furnace wall. The gas flow rates in the furnace were
adjusted to ensure the specified gas residence time. A water-cooled collecting probe was
inserted into the reaction chamber from below. The reaction products were quenched by
supplying nitrogen. The char samples were removed by a cyclone, and a 1 µm glass-
fibre filter. Upon removing the moisture, the exhaust gases were monitored by a battery
of infrared (SO2, CO, CO2, NOx and N2O) and paramagnetic (O2) analysers.
The ignition tests in the entrained flow reactor were conducted at atmospheric pressure.
Coal with a particle size fraction of 53-106 µm was fed in continuously at a rate of 0.5 g
min⁻¹. The reactor was heated at 15 °C min⁻¹ from 500 to 800 °C. The experiments were
conducted under stoichiometric conditions, in air, and 100% excess oxygen. The gas
flow rate employed was that necessary to ensure 2 seconds of residence time at 500 °C
(10 cm³ min⁻¹ at 25 °C and 1 atm).

Seven coals of varying rank and, therefore, different volatile matter content (which
exerts a strong influence on ignition behaviour), were used: PE (Chile), IN (Indonesia),
GU(Venezuela), LK (USA), CA, LD, HV (Spain). The main characteristics of these
coals are given in Table 1.

For the purposes of comparison the coal ignition temperatures were also determined in a
differential thermogravimetric analyser [7, 21]. In all the pyrolysis and combustion tests
5 mg of sample was heated at 15 °C min⁻¹ from room temperature to 850 °C. A gas flow
rate of 50 cm³ min⁻¹ was used; the inert gas for the pyrolysis experiments was argon
whereas air was used as the reactive gas for the combustion tests. The experimental
conditions from a previous work [22] were employed so as to ensure consistently reproducible results.

3. Results and discussion

The repeatability of the ignition tests in the entrained flow reactor was evaluated using coal PE. Figure 1 shows the variation in the concentration of different gases (CO, CO₂, NO and O₂) during three continuous experiments with coal PE in air; the gas evolution profiles for one of the high volatile coals (LK) are also included in this figure for comparison purposes. It can be seen that the repeatability of the temperature at which the ignition event occurs is very good, whichever gas is used for its determination. The results presented in the remaining part of this work correspond to the mean value of at least two experiments. At low temperatures, the production of CO increases, that of CO₂ and NO shows a slight increase, and there is some O₂ consumption due to the evolution of coal volatiles at low temperatures. However, the heat generation is not sufficient to sustain ignition and higher temperatures are necessary for ignition to occur. As can be seen in Figure 1, ignition is characterised by a rapid decrease in CO production, a significant O₂ consumption, and an increase in the production of CO₂ and NO.

The definition of ignition temperature is somewhat arbitrary [14,17]. In this work the criterion for determining the ignition temperature was based on the first derivative curves of the gases composition. The ignition temperature was taken as the temperature where the derivative curves, normalised with respect to the maximum derivative value, reached a value of 10%. This is illustrated in Figure 2 for the evolution of CO₂ corresponding to an ignition test for coal PE in air.

In order to study the effect of oxygen on ignition behaviour, a series of tests was conducted for PE at a fixed coal feed rate of 0.5 g min⁻¹, the oxygen concentration
varying from the stoichiometric value to 300% excess oxygen. The results obtained are depicted in Figure 3, which shows that increasing the excess oxygen leads to a clear decrease in the ignition temperature, due to the greater amount of oxygen available for the coal particles.

The ignition mechanism of the different rank coals was inferred from the evolution curves of the gases. The four high volatile bituminous coals (IN, GU, CA, LK) display a similar behaviour, i.e., two changes can be seen in their gas composition profiles. The first change corresponds to the combustion of the volatiles and the second to char combustion. This was confirmed by the constant increase in the CO$_2$ produced after the ignition event. Thus, under the experimental conditions used in this work, the ignition of high volatile bituminous coals takes place via a homogeneous ignition mechanism, with the sequential ignition of volatiles and char.

A different behaviour was observed for the subbituminous coal, PE. The CO concentration decreased dramatically until nearly zero value, whereas that of CO$_2$ increased drastically after ignition, maintaining thereafter a constant value. This behaviour is in accordance with a heterogeneous ignition mechanism, which involves the simultaneous ignition of volatiles and char. In the case of the subbituminous coal PE, this can be explained by the high reactivity of its char. Coal LD also exhibits a heterogeneous ignition mechanism, but in this case the ignition temperature is much higher than that of coal PE. Temperatures below 700 °C are not high enough for LD to ignite. However, after the ignition event (709 °C in air), there is a steep increase in the CO$_2$ evolution profile as a consequence of the relatively high char reactivity. The semianthracite, HV, is clearly the least reactive coal in accordance with its rank and the more aromatic structure of higher rank coals.
The influence of coal rank on ignition behaviour was also evaluated for the seven coals under stoichiometric, air, and 100% excess O₂ conditions. The results summarised in Figure 4 verify the mutual dependence of ignition temperature and rank, since the values of Figure 4 indicate a decreasing trend of reactivity from PE (subbituminous) to HV (semianthracite). The ignition temperatures of the high volatile coals (IN, GU, CA and LK) remain practically constant. In all cases an increase in excess oxygen produces a decrease in the ignition temperature. It should also be taken into account that for coal PE the ignition tests conducted in air are equivalent to an oxygen excess of 300%, while for the other coals the experiments in air correspond to an excess of oxygen in the 140-160% range.

The ignition temperatures evaluated from the TGA tests are also included in Figure 4 for purposes of comparison. In this work, the ignition temperatures determined by TGA (static method) are almost 300 °C lower than those evaluated in the entrained flow reactor (dynamic conditions), in accordance with the results summarised by Su et al. [2]. Although the operating conditions in the TGA and the EFR are very different, in this work a similar trend for the variation of ignition temperature with coal rank was obtained. In addition, the ignition mechanism of different rank coals analyzed on the basis of the TGA results of a previous work [10], coincides with the mechanism determined in this work in the entrained flow reactor. However, the mechanism and the temperature of ignition depend on the experimental technique, and other results on the ignition of coal blends have highlighted the differences in the ignition mechanism determined in the TGA and in the EFR [10, 23].

Furthermore, the morphological surface of the solid particles separated in the cyclone during the ignition tests in the entrained flow reactor was examined with a Zeiss DSM 942 scanning electron microscope. These observations provided additional confirmation
of the mechanisms of ignition evaluated from the gas evolution profiles. The SEM micrographs shown in Figure 5 illustrate the events that take place before, during and after ignition for coals with a heterogeneous mechanism (exemplified by coal PE in Figures 5a to 5c), and for coals that present homogeneous mechanisms (represented by coal LK in Figures 5d to 5f). The surfaces of the particles of coal PE before ignition (Figure 5a), retain their original angular shape, and most of the particles do not show fissures. Microcracks, and also fissures, are more evident in Figure 5b which depicts the morphology of PE during ignition. In this case the original angular shape is retained but with the appearance of small pores. These features are more pronounced after ignition, once the combustion of coal PE takes place. This is clear from Figure 5c, which shows the irregularly reticulated exterior of the char particles from coal PE, with numerous openings.

In contrast, the behaviour displayed by the particles of the LK bituminous coal is markedly different to that of coals PE, HV or LD, which have a rather different structure and ignition mechanism. It can be observed in Figure 5d that the particles of the high volatile coal start to swell and to show signs of plasticity before ignition, and that externally they begin to show a rounded shape. The structure of coal LK during ignition is presented in Figure 5e; the particles which are of a cenospheric type, present a swollen structure and plastic features with some small degasification holes on their outer surface. The formation of these structures is typical of high volatile bituminous coals with plastic properties. However, contrary to the behaviour of coal PE, most of the particles of LK do not show signs of oxygen attack on the solid surface (Figure 5e). A SEM micrograph of the LK particles retrieved from the cyclone after ignition is shown in Figure 5f. It is clear that in the case of the high volatile bituminous coals there is a first stage, which involves devolatilisation followed by the ignition and combustion of
the volatiles, and a second stage where the heterogeneous attack of oxygen on the char surface occurs (homogeneous ignition mechanism).

The method applied in this work for the assessment of coal ignition behaviour is much more rapid and cheaper than the tests in pilot-scale plants. Although the operating conditions are still different, they are certainly closer to industrial conditions than in the case of TGA. This methodology therefore will be used in order to evaluate the ignition behaviour and the interactive effects that might arise in the case of coal blends, for which the number of studies conducted in entrained flow reactors has been much smaller than in thermogravimetric systems.

4. Conclusions

The ignition behaviour of seven coals varied depending on their rank. The mechanism of ignition, evaluated in a laminar entrained flow reactor, changed from heterogeneous for subbituminous, low volatile bituminous and semianthracite coals, to homogeneous for high volatile bituminous coals. The results obtained in a TGA for the ignition mechanism and for the variation of the ignition temperature with coal rank were the same as those obtained in the EFR, although the more realistic conditions in the EFR make the results of this method more suitable for predicting the ignition behaviour of coal in industrial boilers. Observation by SEM of the morphological structure of the coal particles before, during and after the ignition event, provided additional confirmation of the ignition mechanism of the different rank coals.

Acknowledgements

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Figure 4. Effect of coal rank on ignition temperature.

Figure 5. SEM micrographs of particles from coals PE and LK: a, d) before; b, e) during; c, f) after ignition.
Table 1. Main characteristics of the coals used.

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db: dry basis; daf: dry ash free basis; mmf: mineral matter free basis; sb: subbituminous; hvb: high volatile bituminous; lvb: low volatile bituminous; sa: semianthracite.
Figure 1.
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Figure 2.
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Figure 3.
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