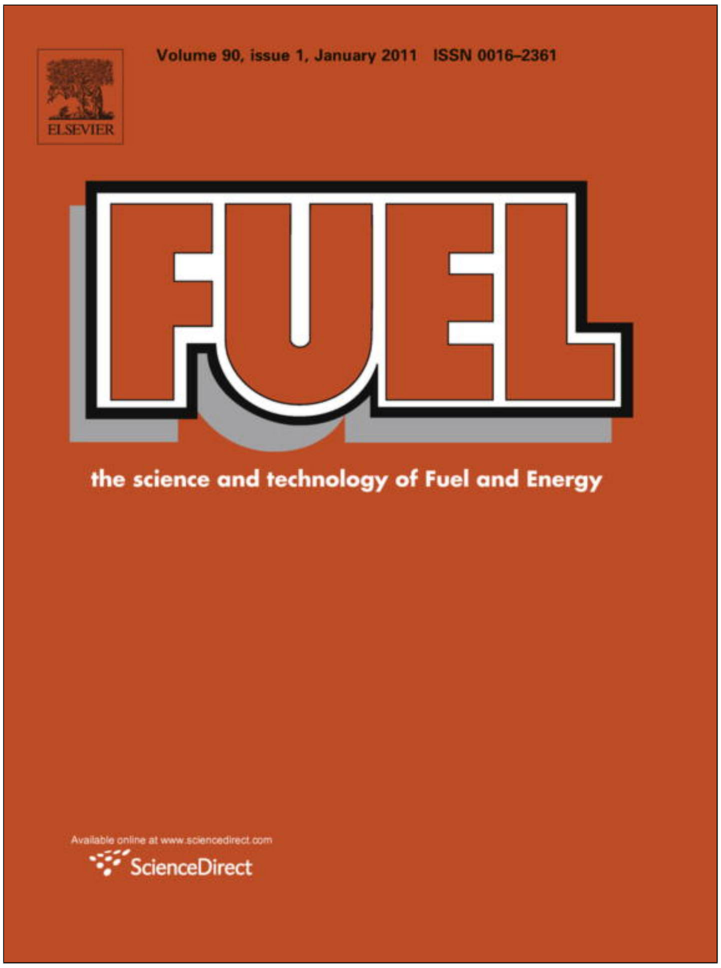
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Factors affecting coal particle ignition under oxyfuel combustion atmospheres

C.K. Man *, J.R. Gibbins

Department of Mechanical Engineering, Imperial College London, Exhibition Road, London SW7 2AZ, UK

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ABSTRACT

A set of 13 coals of different rank has been tested for ignition propensity in a 20-L explosion chamber simulating oxyfuel combustion gas conditions. Their char residues were also analysed thermogravimetrically. The effects of coal type, coal concentration (from 100 to 600 g/m^3), O_2 in CO_2 atmospheres (up to 40% v/v) and particle size were investigated.

The higher rank coals were significantly more difficult to ignite and mostly required higher energy chemical igniters (1000 or 2500 J) whereas the lower rank coals could be ignited with a 500 J igniter even at low coal dust concentrations.

The minimum explosibility limit/ignition concentration in air varied slightly around a value of 200 g/m^3, a little higher for low volatile coals and a little lower for high volatile coals.

The ignition limit changed significantly, however, with O_2 concentration in CO_2, where coals required more oxygen to ignite. Most coals failed to ignite at all in 21% v/v O_2 in CO_2, but an increase to 30 or 35% v/v O_2 gave ignition patterns similar to those in air. In addition, the minimum ignition concentration decreased with increase in O_2. However, a further increase to 40% v/v O_2 did not generally affect the minimum ignition concentration.

Particle size had a non-linear effect on coal ignition. The fine particles (<53 μm) behaved almost identical to the whole coal. However, the larger size fraction (>53 μm) was generally more difficult to ignite and exhibited a much lower weight loss.

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1. Introduction

Oxyfuel combustion, also known as oxyfiring, of pulverised fuel (PF – also known as pulverised coal) in relatively conventional supercritical boilers has been estimated potentially to be able to achieve cost and performance levels with CO_2 capture that are comparable to those for alternative pre-combustion (using integrated gasifier combined cycle – IGCC) and post-combustion (amine) options [1–3]. Currently the largest oxyfuel trials are at around 30–40 MW thermal scale, at Schwarze Pumpe in Germany [4] and Renfrew in Scotland [5]. The latter work is, however, developing a single burner which in multiple units could be used in any size of plant.

In oxyfuel combustion systems based on air-fired plant recycled flue gas, consisting mainly of CO_2 and water vapour, is used to replace nitrogen from the air and to moderate flame temperatures [6,7]. Under oxyfuel combustion conditions formation of both SOx and NOx has been reported to be reduced [8,9]. In addition, high levels of SOx and NOx removal may be possible by relatively simple processing as the CO_2 is compressed for transport and storage [10].

While oxyfuel burners have similarities to conventional air burners in some areas, they differ significantly in others such as different levels of O_2 will be encountered in primary and secondary gas streams. In particular, it is possible that oxygen would not be added to the primary flue gas recycle stream passing through the mills for safety reasons (i.e. the certainty of serious explosions if oxygen levels were accidentally elevated in the mills), or only be added up to air concentrations (i.e. 21% v/v). In contrast, in conventional bituminous coal burners the incoming fuel is ignited using oxygen pre-mixed with the fuel in the primary air. In oxyfuel burners with recycled flue gases as primary 'air', oxygen must be mixed with the fuel after injection giving potentially quite different combustion patterns. The recycled flue gases replacing the nitrogen in air would also contain much higher levels of CO_2 and water vapour, both triatomic gases with significantly higher heat capacities than diatomic nitrogen, and possibly also higher dust loadings.

The ability to assess the ignition behaviour of pulverised coal under oxyfuel combustion conditions, and to predict the behaviour of different coal types, is therefore important for oxyfuel burner development and design, and also for oxyfuel plant safety

* Corresponding author. Present address: Pittsburgh Research Laboratory, National Institute for Occupational Safety and Health, PO Box 18070, Pittsburgh, PA 15236, USA. Tel.: +1 412 386 6628; fax: +1 412 386 6595.

E-mail address: cman@cdc.gov (C.K. Man).

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considerations related to PF handling. This paper examines the effect of coal type and oxygen/CO_2 concentrations on ignition in a laboratory apparatus and an analysis of the properties of the residual char. The principal objectives of the study were to establish ignition patterns comparable to air and to investigate the effect on coal rank.

2. Experimental

2.1. Coals

Thirteen coals were used in this study covering a wide range of coal rank. Six were from North America (USA) and the remainder from China (CN), the UK, South Africa (SA) and Vietnam (VNM). All coal samples were supplied in pulverised form, typically 75% passing through a 75 μm screen. Table 1 shows the basic coal characteristics.

The effect of particle size was investigated using three of the coals (Freeport, Pittsburgh and Sewell). Samples were sieved by hand at 53 μm to give large and small particle fractions. This cut off size was chosen because it gave roughly an equal 50:50 split in the pulverised fuel, which is normally ground to 75–80% below 75 μm. In addition, particle size distributions for the coals were determined using a Gilsonic Autosiever ™. The particle size distributions, i.e. grind quality for all the coals are illustrated in Fig. 1.

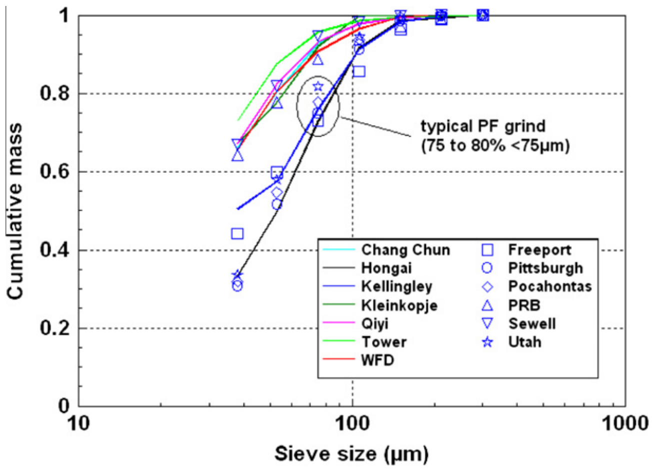

Fig. 1. Cumulative particle size distributions.

2.2. Coal ignition apparatus and test procedures

The Pittsburgh Research Laboratory (PRL) 20-L ignition chamber (Fig. 2) has been described in detail elsewhere [11–13]. It is almost spherical in shape and made of stainless steel with a pressure rating of 21 bar (g). The top of the chamber is hinged and opens across the whole chamber diameter, thus allowing easy access to the interior for sample loading and cleaning after each test. For tests it is secured to the main body with six bolts. A strain gauge pressure transducer is used to measure the explosion pressure and rate of pressure rise. As this measures absolute pressures, it can also be used to monitor the evacuation of the chamber prior to the addition and dispersion of the test gas. The sapphire windows serve as viewports and permit temperature measurements using infrared pyrometers (not used in these tests). Pressure data (and temperature if required) can be sampled at a maximum rate of 9 kHz using a desktop computer. An in-house computer program allows the data to be processed, printed and stored.

Initial ignition/deflagration tests were carried out in air using whole PF samples. The pulverised coal dust samples were carefully weighed and placed on the dispersion nozzle at the bottom of the chamber (Fig. 3). The coal concentration for any test (in g/m^3) is equal to the mass divided by the chamber volume. After the coal and igniter have been placed in position, the lid was bolted securely and the chamber partially evacuated to 13.8 kPa (2 psi).

Fig. 2. The 20-L ignition apparatus.

Table 1
List of coals and properties (n/a = not analyzed, VM = volatile matter, GCV = gross calorific value).

Coal name	Coal origin	Moisture %ar	VM %ar	FC %ar	Ash %ar	VM %daf	GCV MJ/kg	C %daf	H %daf	S %daf	Cl %daf	N %daf	O %daf
Chang Chun	CN	0.6	12.2	72.7	14.5	14.4	30.1	90.1	4.3	0.4	0.0	1.7	3.4
Hongai	VNM	0.7	6.5	68.4	24.4	8.7	26.1	93.3	3.5	0.7	0.0	0.9	1.5
Kellingley	UK	3.3	30.3	50.9	15.5	37.3	28.0	83.1	5.3	2.3	0.4	1.8	7.2
Kleinlopje	SA	3.6	21.6	56.2	18.6	27.8	25.8	83.9	5.0	0.4	0.0	1.7	9.0
Qiyi	CN	0.5	13.0	67.8	18.7	16.1	28.4	89.1	4.5	0.3	0.5	1.4	4.2
Tower	UK	0.5	9.1	84.1	6.3	9.8	33.4	91.8	3.9	0.9	0.0	1.3	1.9
WFD	UK	0.7	8.4	84.6	6.3	9.0	33.4	92.5	3.8	0.9	0.0	1.3	1.4
Freeport	USA	0.6	22.8	69.1	7.5	24.8	n/a	88.0	4.8	1.2	0.0	1.5	4.5
Pittsburgh	USA	1.8	35.4	57.5	5.3	38.5	n/a	84.0	5.5	1.2	0.0	1.7	7.7
Pocahontas	USA	0.9	18.9	74.8	5.4	20.2	n/a	90.5	4.1	0.9	0.0	1.2	3.2
PRB	USA	11.3	37.5	45.4	5.9	45.2	n/a	74.2	4.8	0.3	0.0	1.1	19.7
Sewell	USA	1.4	29.7	62.0	6.9	32.4	n/a	86.1	4.8	0.8	0.0	1.7	6.6
Utah	USA	2.2	38.5	52.9	6.4	42.1	n/a	81.7	5.5	0.8	0.0	1.9	10.2

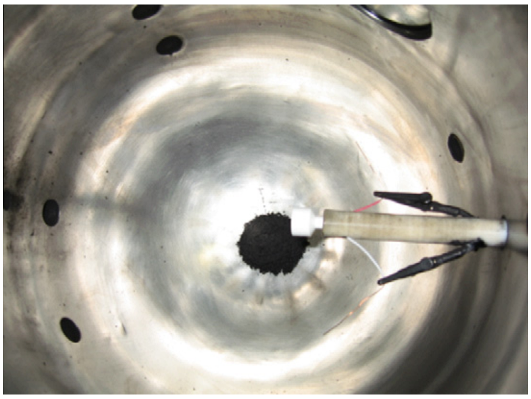

Fig. 3. Inside of ignition chamber with coal and igniter in position.

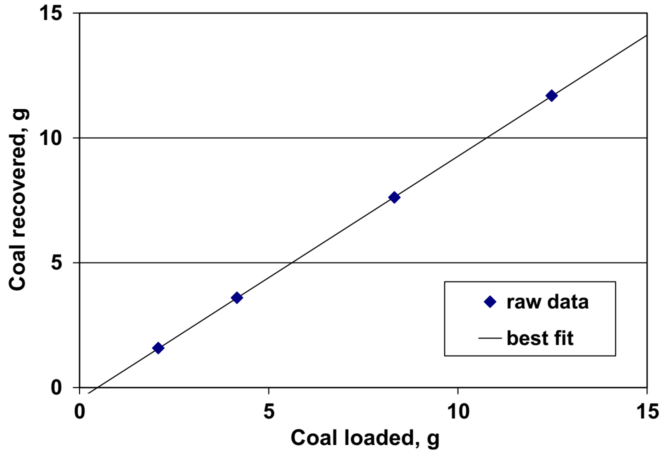

Fig. 4. Coal loading and collection correlation.

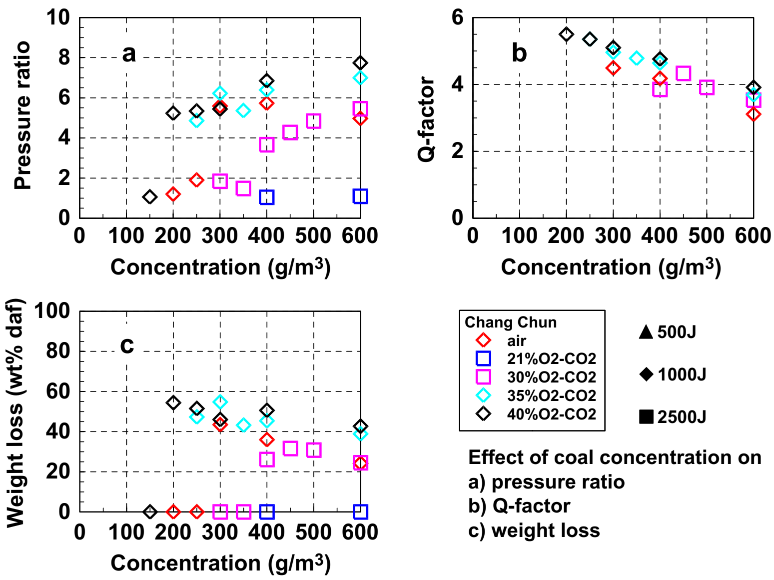

Fig. 5. Chang Chun coal – effect of coal loading.

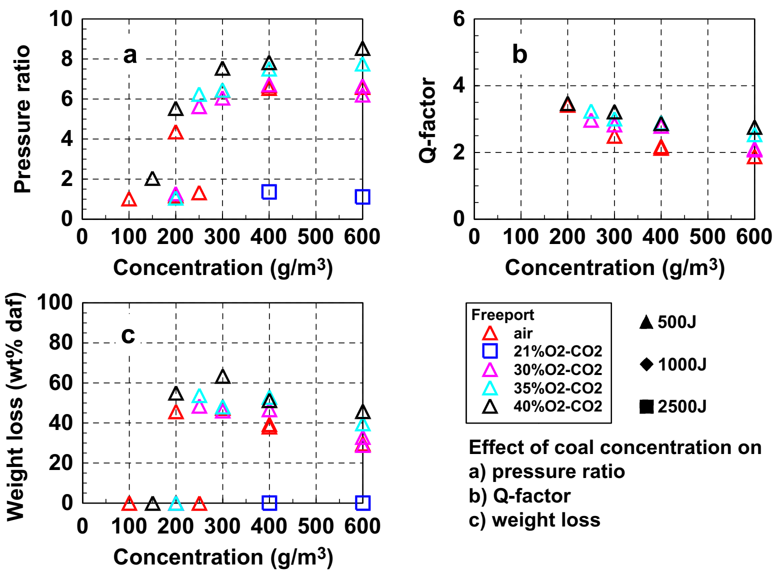

Fig. 6. Freeport coal – effect of coal loading.

A short blast of dry air (0.3 s at 758.4 kPa (110 psi) from a 16 L reserve tank) was use to disperse the dust and raise the chamber pressure to about 1 bar (a). After an additional delay of 0.1 s the igniter was activated electrically.

Ignition tests were also carried out in 21, 30, 35 and 40% v/v O_2 in CO_2. For tests using O_2/CO_2 mixtures, pre-mixed cylinders of the appropriate concentrations were obtained. The switch from using air to other gases required a few small modifications to the normal ignition chamber test procedure. The reserve tank was flushed out several times before being filled with the gas mixture used to disperse the coal. In addition, the chamber was evacuated totally before it was back filled to 13.8 kPa (2 psi) with the gas mixture followed by dispersion and ignition.

2.3. Igniters

The pyrotechnic igniters were manufactured by Fr. Sobbe, Germany and consist of 40 wt% zirconium, 30 wt% barium nitrate and 30 wt% barium peroxide. These generate a large number of hot particles with minimal increase in gas volume through the following reaction:

$$3Zr + Ba(NO_3)_2 + BaO_2 \rightarrow 2BaO + 3ZrO_2 + N_2$$

The igniters are activated electrically with an internal fuse wire with a delay of about 0.01 s. A 2500 J igniter can be considered to be equivalent to about 20 matches all ignited at once.

2.4. Weight loss measurements in ignition tests

Weight loss i.e. devolatilisation from the ignition process can be calculated in two ways. One is by the 'ash tracer' method, which assumes the ash material from the coal to be inert.

$$\text{Ash tracer weight loss, wt\%db} = 100 \times (1 - \text{coal ash/char ash})$$

However, this method is often unreliable [14,15], especially for coals with low ash contents where the error bars are very wide. Igniter residue material, barium oxide and zirconium oxide, is also

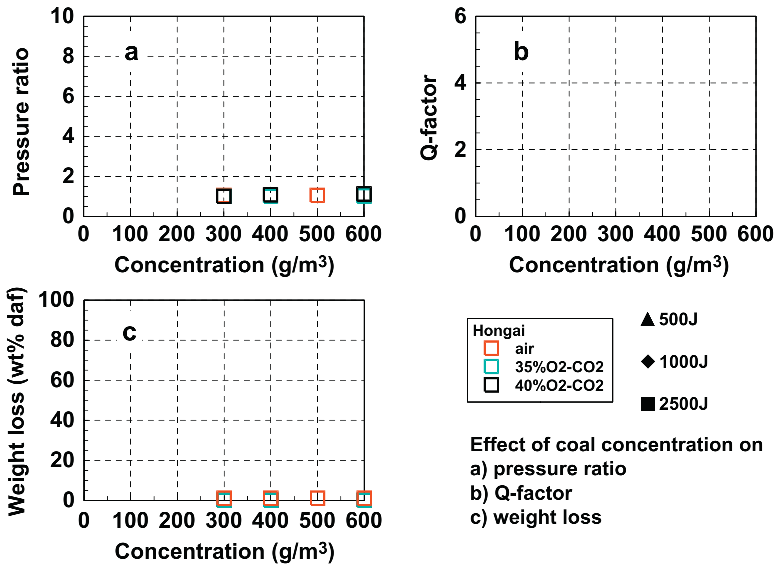

Fig. 7. Hongai coal – effect of coal loading.

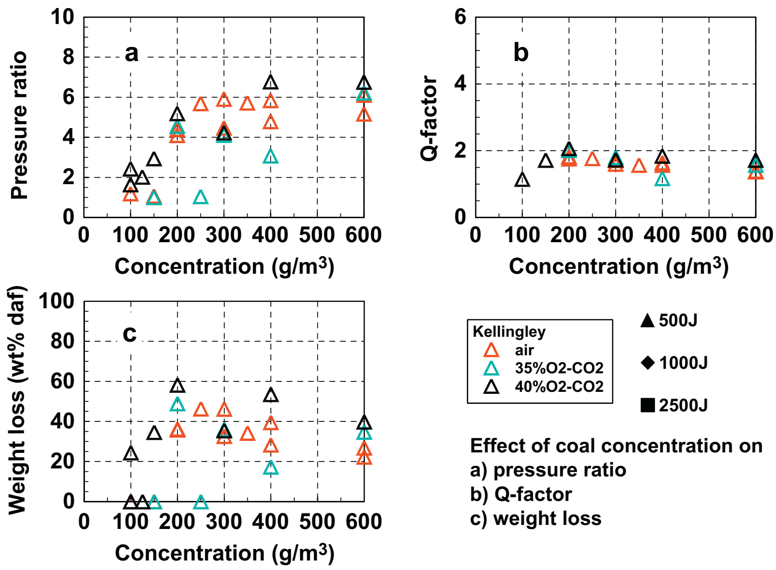

Fig. 8. Kellingley coal – effect of coal loading.

not readily distinguished from the coal ash in the samples collected from ignition tests.

An alternative collection of the char residue for direct weighing to derive the weight loss was investigated. It is still necessary to correct for the igniter residue, but this can be done by simple chemical stoichiometry; a 500 J igniter contains 120 mg of reactants and larger igniters pro rata. In addition, since it is not possible to collect absolutely all of the material, a collection factor, essentially a collection efficiency, was found to be necessary. A formula for this was established by carrying out 'blank' runs where coal samples were dispersed in the 20-L chamber but without any igniters using about 2, 4, 8 and 12 g of coal which is equivalent to dust concentrations of 100, 200, 400 and 600 g/m^3 respectively. A good correlation was obtained between the coal loaded and the coal collected (Fig. 4) and a linear collection factor correction equation was derived, as shown below:

$$\text{Corrected mass recovered} = \text{mass collected} \times 1.029 + 0.458\ \text{g} - \text{igniter residue}$$

From this, the weight loss from the ignition experiments can easily be obtained by the equation below.

$$\text{Weight loss, wt\%ad} = 100 \times (1 - \text{corrected mass recovered}/\text{coal loaded})$$

Finally, this can be converted to a dry-as-free basis by the following equation:

$$\text{Weight loss, wt\%daf} = \frac{100 \times \text{weight loss(wt\%ad)} - \text{moisture}}{100 - (\text{coal ash} + \text{moisture})}$$

2.5. Char sample TGA tests, residual volatile matter and Q factor calculations and interpretation

All the residual char samples from the ignition tests were analysed in a Mettler–Toledo TGA/SDTA851e thermogravimetric analyser to give micro-proximate data [16]. Samples (typically 10 mg) were heated in a 75 µl alumina crucible inside a TGA furnace with a flow of high purity nitrogen to 105 °C then to 900 °C

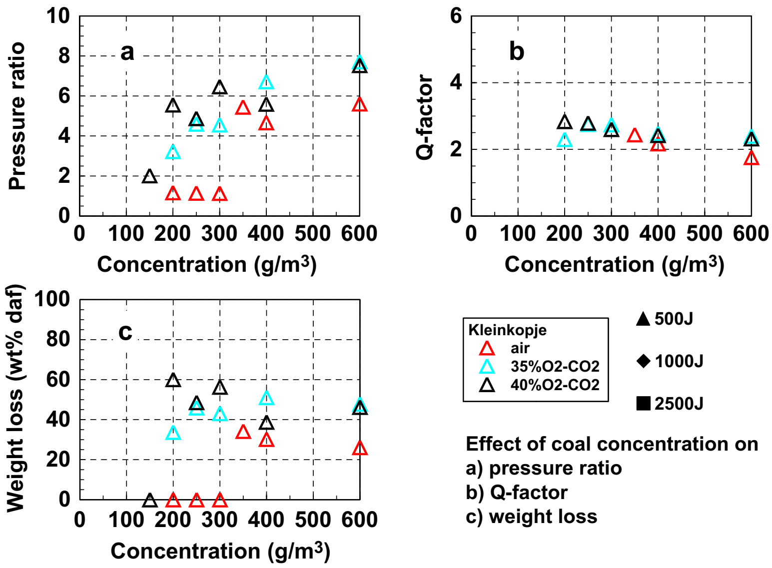

Fig. 9. Kleinkopje coal – effect of coal loading.

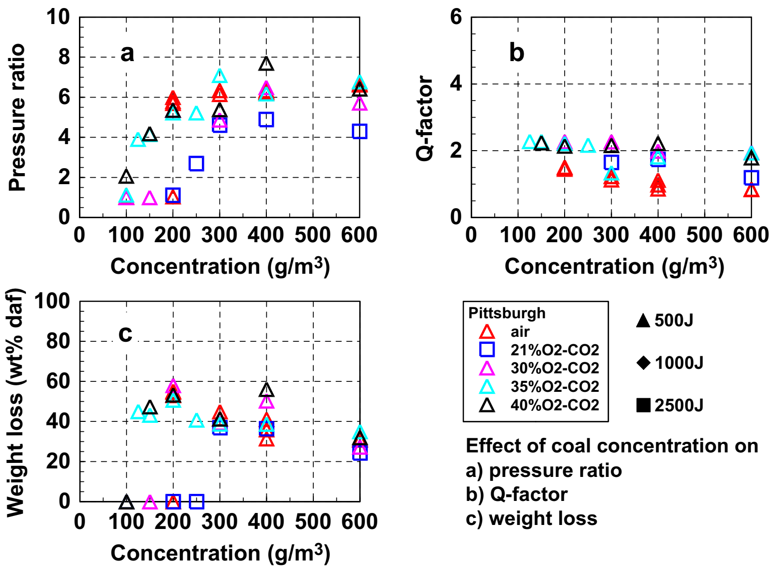

Fig. 10. Pittsburgh coal – effect of coal loading.

(at 30 °C/min), to give values for moisture and volatile matter content respectively. Finally, introduction of air resulted in 100% burn-off and allowed the fixed carbon and ash values also to be calculated.

Micro-proximate ash contents can be used to estimate weight loss during the ignition test but, as discussed above, this appears to be a less reliable method than direct weighing. Residual volatile matter contents can be used to estimate the extent to which the potential volatile release from the coal has been completed.The *Q*-factor is the ratio of coal volatile matter release, which represents the efficiency of volatile release compared to standard proximate analysis [17,18].

$$\text{Q-factor} = \frac{\text{weight loss from devolatilisation(\%daf coal)}}{\text{Coal VM(\%daf)} - \text{Char VM(\%daf of original coal)}}$$

This should not be confused with the *R*-factor which is simply the ratio of measured volatiles divided by the proximate volatiles [17,19].

$$\text{R-factor} = \frac{\text{weight loss from devolatilisation(\%daf coal)}}{\text{Coal VM(\%daf)}}$$

3. Results and discussion

The relationship between coal loading on ignition and the effect of air and oxygen concentration in CO_2 for all 13 coals are shown in Figs. 5–17. A pressure ratio (PR) of two and above i.e. a doubling of the pressure inside the chamber can be regarded as a positive ignition test. It was possible to ignite all coals in the 20-L chamber

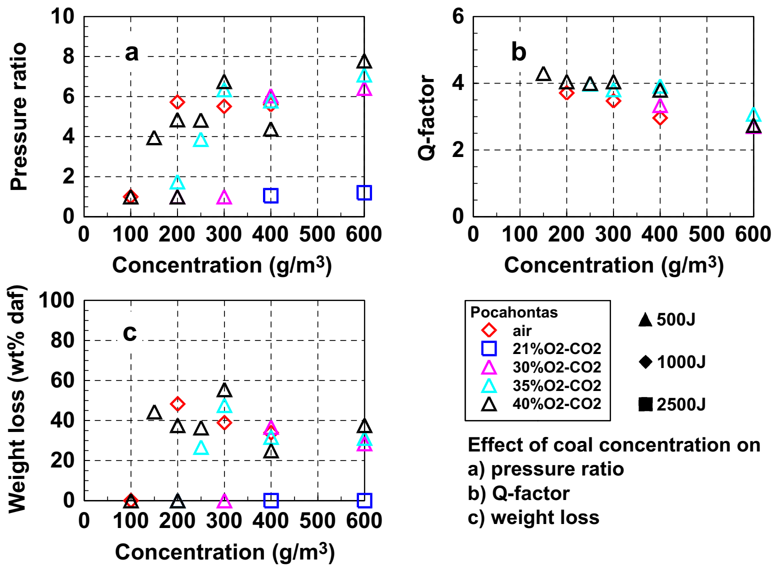

Fig. 11. Pocahontas coal – effect of coal loading.

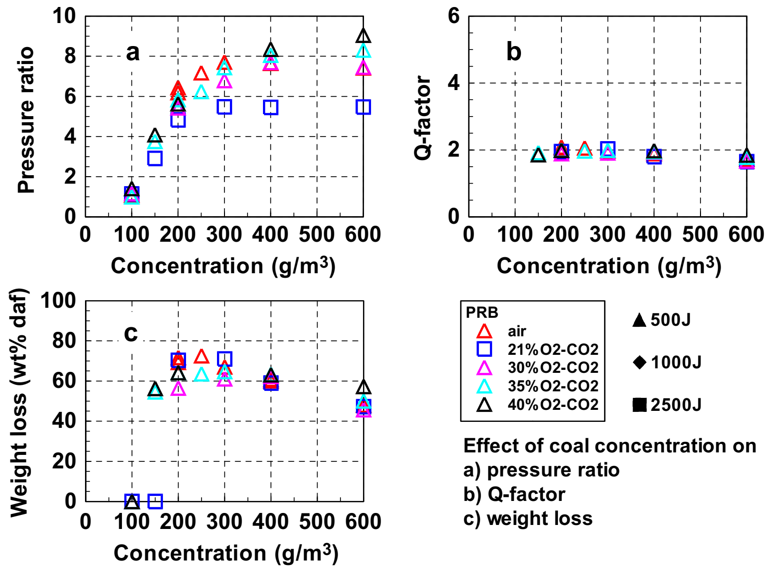

Fig. 12. PRB coal – effect of coal loading.

using air as the dispersal medium, with the exception of Hongai coal (Fig. 7) which did not ignite even with the 2500 J igniter.

The higher rank coals were significantly more difficult to ignite, as one might expect [20], but most ignited in air using the higher energy chemical igniters (1000 or 2500 J) whereas the lower rank coals could be ignited with a 500 J igniter even at low coal dust concentrations.

The minimum explosibility limit (MEL)/ignition concentration in air varied slightly for each coal. This was mostly around the 200 g/m^3 level, a little higher for low volatile coals and a little lower for high volatile coals. The ignition limit, on the other hand, changed significantly with O_2 concentration.

Only the high volatile coals Pittsburgh (Fig. 10), PRB (Fig. 12), Sewell (Fig. 14) and Utah (Fig. 16) ignited in 21% O_2/CO_2 and all required the large 2500 J igniters (the Kellingley coal was not tested under these conditions). However, an increase in O_2/CO_2 levels to 30 or 35% gave ignition patterns similar to that carried out in air for all coals. In addition, the minimum ignition concentration decreased by about 100 g/m^3 with these higher O_2 levels compared to the corresponding runs in air. Interestingly, further increase in O_2/CO_2 from 35 to 40% did not have much additional effect on the minimum ignition concentration.

Above the minimum ignition concentration the peak PR for any given oxygen concentration had a tendency to only increase slightly if at all and this appears to be independent of coal rank. However, there appeared to be a slight increase in PR with increasing oxygen concentration.

The char residues from the ignition tests showed a wide range of *Q*-factors. Low rank, high volatile coals such as Pittsburgh (Fig. 10), PRB (Fig. 12) and Utah (Fig. 16) have low *Q*-factors, typically around 2. In addition, this value appears to be almost constant for all tests for these coals, regardless of coal loading and oxygen concentration. A value of 2 and the invariance suggests that mainly devolatilisation is taking place.

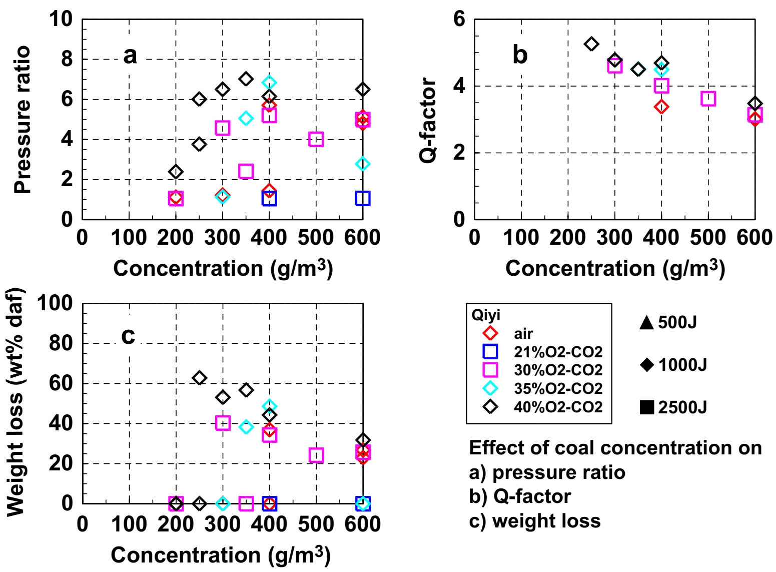

Fig. 13. Qiyi coal – effect of coal loading.

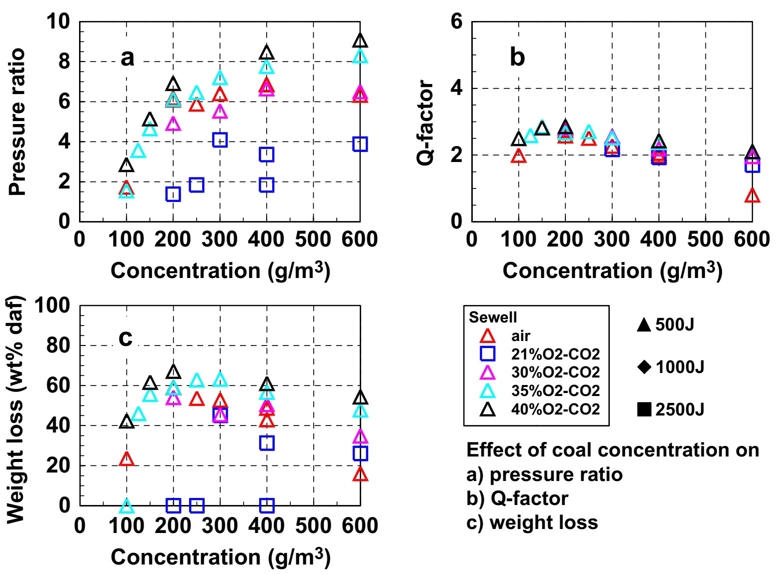

Fig. 14. Sewell coal – effect of coal loading.

The medium rank coals Chang Chun (Fig. 5), Pocahontas (Fig. 11), and Qiyi (Fig. 13) exhibited higher *Q*-factors which range from about 3 to 5 and are again in a narrow band, but these show a definite negative slope with coal loading. The high rank low volatile coals Tower (Fig. 15) and WFD (Fig. 17) have very high *Q*-factors which spread from below 5 to over 10. Again there is a negative slope with coal loading. For the Tower coal there appears to be an additional trend of increase in *Q*-factor with oxygen concentration.

The weight loss results show a similar trend to the pressure ratio data (Figs. 5–17). There is sharp increase in weight loss at the minimum explosibility limit but this often levels off or falls slightly with increase in coal loading.

There appears to be a maximum weight loss of around 40–50%daf for many of the coals. This is certainly not unexpected or unusual for high volatile coals such as Pittsburgh (Fig. 10), Sewell (Fig. 14) and Utah (Fig. 16). This equates to *R*-factors of about 1.3, which has been confirmed in other studies [19] as being a reasonable increase over proximate VM yields for rapid heating. However, the low volatile coals such as Chang Chun (Fig. 5) and Tower (Fig. 15) give almost the same amount of weight loss, typically around 30–40%daf, which appear high relative to their proximate volatile matter contents. For these coals, the *R*-factors are about 3.

There is a general trend for weight loss to increase with increase in oxygen concentration. For some coals, weight loss in 40% O_2/CO_2 can be as much as 20% higher than the corresponding experiment carried out in air (i.e. constant coal loading and igniter strength).

One possible explanation for this extraordinarily high weight loss is heterogeneous combustion in addition to devolatilisation. Although the 20-L chamber only attains temperatures suitable for heterogeneous combustion to take place (approximately >400–500 °C) for a short period of time (estimated to be about 1 s), a small amount of char combustion cannot be ruled out. Perhaps even more surprising is that this occurs across the

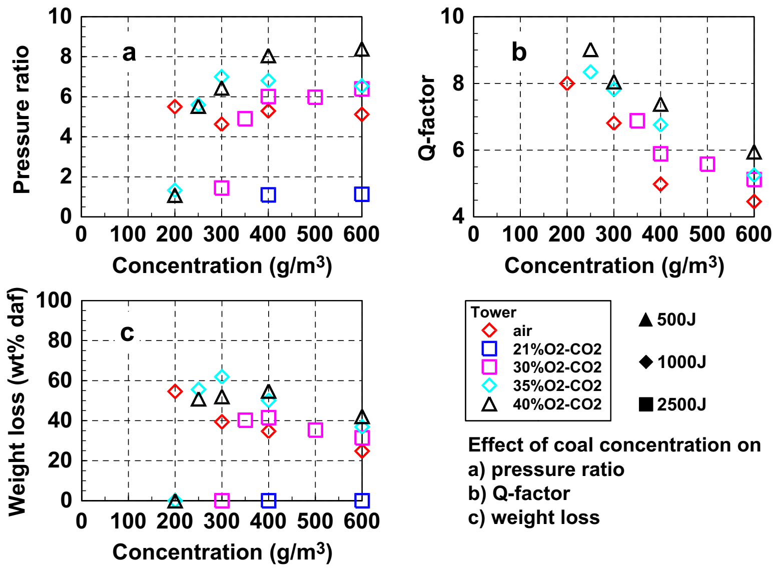

Fig. 15. Tower coal – effect of coal loading.

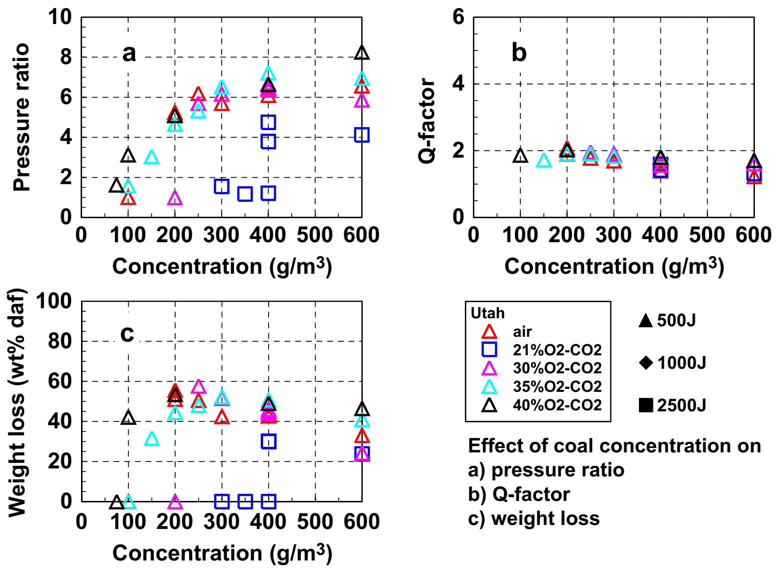

Fig. 16. Utah coal – effect of coal loading.

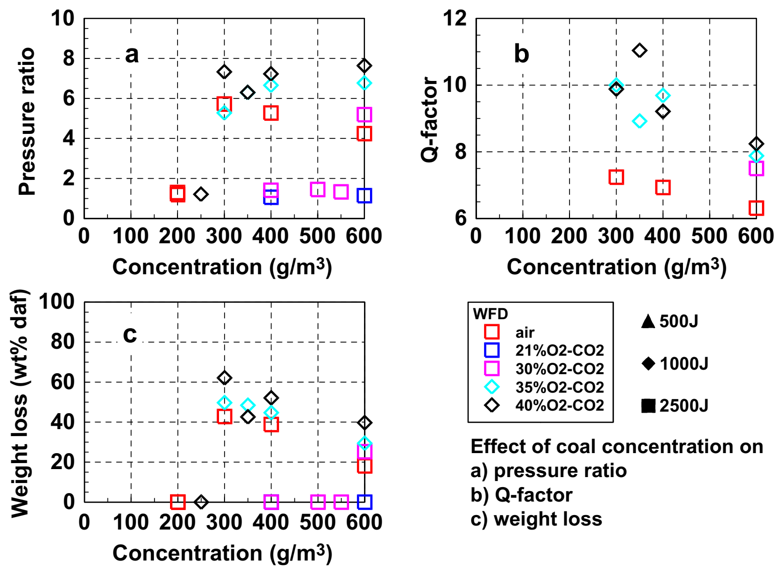

Fig. 17. WFD coal – effect of coal loading.

set of coals tested, even the low volatile coals which are generally considered to be less reactive.

Comparing the weight loss data with oxygen concentration, it appears that the ignition pattern for tests carried out in air are similar to those done in the 30 or 35% O_2/CO_2 mixtures depending on the coal. The low volatile coals appear to need a slightly higher level of oxygen i.e. 35% to match the data in air whereas the higher volatile coals in general only 'need' about 30% oxygen. This "ignitability" data has been summarized in Table 2 which shows this trend more clearly.

Low volatile coals are known to be difficult to ignite. Therefore, the five low volatile coals have been ranked in order of ignitability from the ignition experiments carried out in air. To minimize external influences, the data chosen consisted of tests using a common strength of igniter (1000 J). Hongai (Fig. 7) did not ignite with the 2500 J igniter so it was not tested with the 1000 J igniter. It has been included in this comparative study for completeness. All factors were considered, including pressure ratio, minimum explosive limit, and weight loss. Table 3 shows the five coals listed in order of ease of ignition (with the easiest first).

The order generally follows coal rank with the highest volatile matter content coal being the easiest to ignite. Chang Chun appears to be the easiest coal to ignite with fairly high PR values and a MEL of about 300 g/m^3. Tower was a close second using these criteria.

Table 2
Ignition "goes" under different oxygen concentrations (◯ = no ignition, ◐ = 50% ignition, ● = 100% ignition, empty space means no data).

Coal	Oxygen concentration (% v/v)			
	21 in N2	30 in CO2	35 in CO2	40 in CO2
Chang Chun				
Freeport				
Hongai				
Kellingley				
Kleinlopje				
Pittsburgh				
Pocahontas				
PRB				
Qiyi				
Sewell				
Tower				
Utah				
WFD				

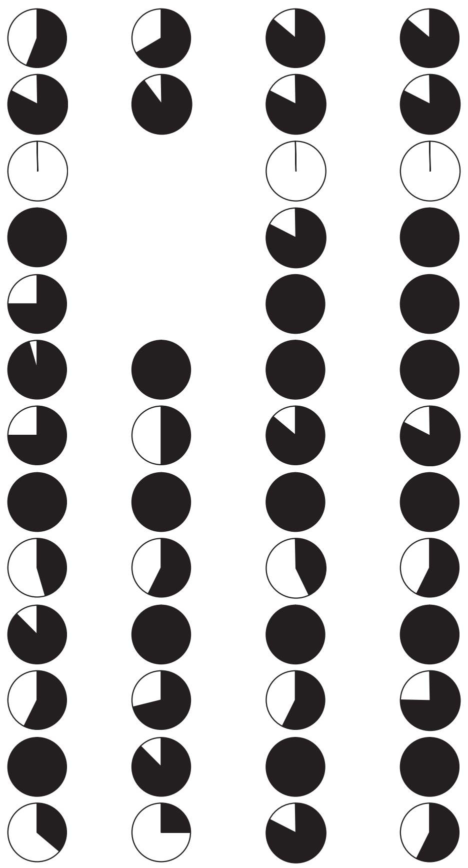

Table 3
Low volatile coal ignitability ranking.

Coal ignitability (top = most easy)	TGA prox VM%daf	Wt.%daf TGA volatile released at, °C		
		500	550	600
Chang Chun	15.5	2.49	5.47	8
Tower	11.0	2.01	3.01	4.4
Qiyi	16.1	2.9	6.29	8.95
WFD	9.1	0.77	1.55	2.92
Hongai	8.3	2.33	3.29	4.05

Table 4
Coal ranking by ignition index.

Coal ignitability (top = most easy)	TGA prox VM%daf	Ignition index for two igniter energies		
		1000 J	2500 J	Mean
Chang Chun	15.5	243	306	274
Tower	11.0	300	243	271
Qiyi	16.1	211	232	221
WFD	9.1	176	245	210
Hongai	8.3	0	0	0

The ignition at 200 g/m^3 may be an anomaly because it was not repeatable using the 2500 J igniter. Therefore, its MEL was roughly the same as Chang Chun at about 300 g/m^3.

Tower was judged to be better than Qiyi in ignition, despite having a lower volatile matter content from the TGA results (which might have suggested the opposite). Again this assessment was based on the respective PR, weight loss and minimum explosive limits, which mostly places Tower to be easier to ignite of the two coals. The MEL for both Qiyi and WFD is about 400 g/m^3. However WFD gave lower PR values and it did not ignite at 600 g/m^3 with the 1000 J igniters. This further weakened its ignition propensity ranking. In general, WFD required the 2500 J igniters to give a successful ignition in air. As mentioned before, Hongai did not ignite even with 2500 J igniters. This is attributed to its low volatile content and relatively coarser grind (about 72% below 75 μm, Fig. 1) – a poor combination of factors.

The partial TGA volatile release values at three different temperatures shown in Table 3 have been added to evaluate whether the amount of volatiles released at relatively low temperatures had an effect on the coals ignitability. The general trend appears similar to the overall volatile matter content.

An attempt to quantify this coal ignition ranking has been made by introducing an ignition index [21]. This is intended to make the ignition ranking process less subjective.

$$\text{Ignition index(in air)} = \text{maximum weight loss from ignition(wt\%daf)} \times \text{PR}$$

The results are shown in Table 4. Interestingly the order has not changed but this way of ranking the coals does have the advantage of allowing coals to be both compared and contrasted from an ignition point of view. From the mean values, two of the coals, Chang Chun and Tower, appear to have similar ignitability. Two other coals Qiyi and WFD also appear to be comparable in ignitability from this evaluation method.

Particle size appears to have a small effect on coal ignition. Overall, the <53 μm fraction behaved very similar to the whole coal, i.e. there was no additional 'fines' effect with this sample. This might suggest that the ignition process is almost insensitive to particle size as the weight losses are very close between the whole coal and the <53 μm fraction for the three coals studied (Figs. 18–20). However, the large size fractions (>53 μm) of a coal were generally slightly more difficult to ignite and always resulted in a significantly lower weight loss compared to its corresponding small size fraction (<53 μm). As with the whole coal ignition tests, the weight loss peaks at the minimum explosibility level (around 200 g/m^3 for two of the set of three coals examined) and gradually decreases with increase in coal loading. Pittsburgh was slightly different (Fig. 19). Although the <53 μm fraction had a MEL level of about 200 g/m^3 like the others, the large particle size fraction was more difficult to ignite and the explosibility limit was increased to about 300–400 g/m^3.

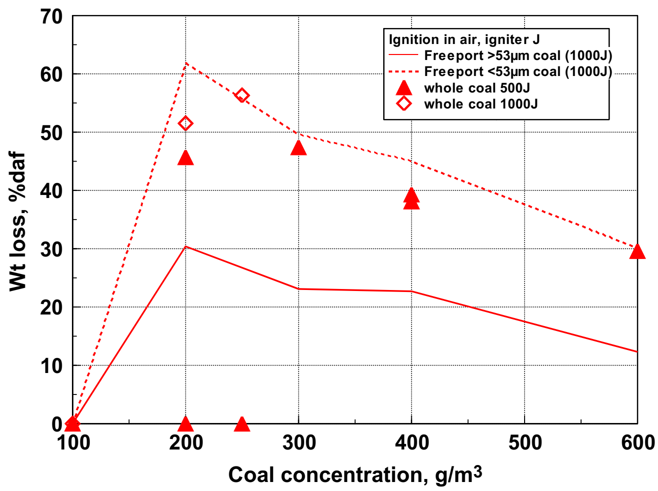

Fig. 18. Effect of particle size on weight loss for Freeport coal.

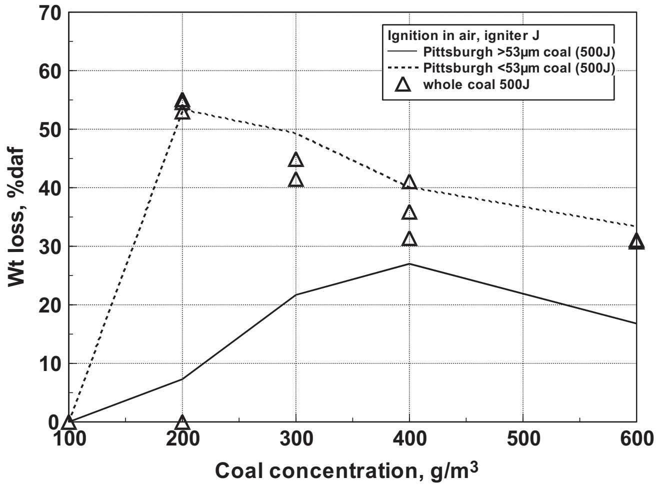

Fig. 19. Effect of particle size on weight loss for Pittsburgh coal.

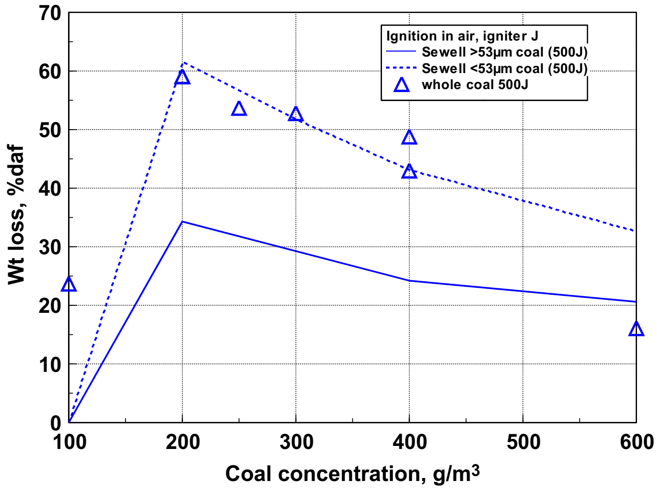

Fig. 20. Effect of particle size on weight loss for Sewell coal.

4. Conclusion

The PRL ignition apparatus can be used to assess and rank coals in order of ease of ignition in air and in O_2/CO_2 mixtures from 21 to 40% v/v. The ignition order generally followed coal rank. There were relatively smaller differences than expected between the different coals at higher O_2 levels since the low volatile coals showed higher than expected weight losses.

Almost all the coals ignited in O_2 in CO_2 at some point. The concentration of O_2 in CO_2 that gave a similar ignition comparable to that in air was established to be between 30 and 35%. This is consistent with recent data reported by CANMET, which concluded that the heat flux from oxyfuel experiments between 28 and 35% O_2 in CO_2 was comparable to that carried out in air [22]. Few coals ignited in 21% O_2 in CO_2 even with the large 2500 J igniter. This has important safety implications for coal mills in an oxyfuel power plant, since it could be dangerous if coal ignited inside the mills or in the adjacent pipework.

Particle size had a small but nevertheless notable effect on coal ignition with the finer particles behaving almost identical to the bulk coal but 'large' particles (>53 μm) tended to be more difficult to ignite.

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