

# A comparison between ignition behaviours of 7 different UK and World-Traded coals in air, and in a mixture of oxygen and carbon dioxide gases representative of oxy-combustion conditions

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## 1. Abstract

A 20L ignition test chamber has been used to test suspensions of 7 UK and world traded coals in air and O<sub>2</sub>/CO<sub>2</sub> mixtures typical of oxy-combustion conditions. The coals varied in rank from sub-bituminous to bituminous and were tested in varying concentrations from the ignition limit to 400g/m<sup>3</sup>. Following each successful test the combustion residue was collected, weighed and analysed within a thermogravimetric analyser, allowing Q factors to be estimated.

The ignition limit varied slightly in air, but was mostly around the 200g/m<sup>3</sup> level. With the exception of Coal C, which was much harder to ignite, it correlated roughly with coal rank. The ignition limit changed significantly with O<sub>2</sub> concentration when in mixed O<sub>2</sub>/CO<sub>2</sub> gases. Only a few high volatile coals ignited in 21% O<sub>2</sub>/CO<sub>2</sub> v/v, and then only with a 2500 J, rather than a 1000J, igniter.

An increase in O<sub>2</sub>/CO<sub>2</sub> levels to 30 or 35% gave ignition patterns similar to those carried out in air with a further increase to 40% having little additional effect. In addition the minimum ignition concentration decreased with increase in O<sub>2</sub>.

Heterogeneous combustion or gasification of the coal by CO<sub>2</sub> appears to be confirmed by comparing weight loss results for air and O<sub>2</sub>/CO<sub>2</sub> mixtures respectively for an equivalent peak pressure rise.

## 2. Introduction

Oxy-combustion consists of the combustion of coal in either oxygen or more likely O<sub>2</sub>/CO<sub>2</sub> mixtures. This produces a flue gas with high levels of CO<sub>2</sub> and low levels of inert gases, particularly nitrogen. Oxyfuel combustion therefore reduces the cost and energy penalty inherent in actually capturing CO<sub>2</sub>; however, there is a trade off due to the

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requirement for an air separation unit to generate the necessary oxygen. Oxyfuel is currently less well-developed than IGCC and post combustion capture, with oxyfuel burner design a key factor affecting the technical feasibility of the process.

The recycling and replacement of the oxidant stream's atmospheric nitrogen with flue gases will have a major effect upon combustion. Diatomic nitrogen has a lower molar heat capacity than the mainly triatomic flue gas. This is the primary reason for the higher O<sub>2</sub> concentrations shown by this study to be necessary. The ability to accurately model the ignition behaviour of pulverised coal, and to predict the behaviour of different coal types, will be critical for oxyfuel burner and development.

### 3. Samples and experimental programme

Seven coals from a range of sources, but mostly typical of those burned in UK power stations have been tested in a 20 litre ignition chamber. The set of coals covered those with volatile matter contents ranging from below 30 to ca. 50 wt% daf – spanning the range of bituminous to sub-bituminous coals. These coals and their proximate analysis are listed in Table 1. All coal samples were supplied in pulverised form.

The following were investigated:

- Effect of coal rank,
- Effect of coal concentration (from the ignition limit to 400g/m<sup>3</sup> in O<sub>2</sub>/CO<sub>2</sub> gases, and the ignition limit to 600g/m<sup>3</sup> in air),
- Effect of oxygen concentration (air, 21.5, 26.5, 30.2, 34.9 and 40.0% v/v O<sub>2</sub> in CO<sub>2</sub> were studied for all coals except Coal F, which was only studied in air)

Table 1 - Proximate analysis of the coal samples in rank order

Coal	Moisture (% ar)	VM (% db)	FC (% db)	Ash (% db)	VM (% daf)	FC (% daf)
A	7.4	44.4	45.9	9.7	49.1	50.9
B	3.3	36.7	51.9	11.4	41.5	58.5
C	2.6	33.0	53.1	13.9	38.3	61.7
D	1.7	32.7	52.7	14.6	38.3	61.7
E	1.6	30.3	49.4	20.3	38.0	62.0
F	-	34.7	60.2	5.1	36.6	63.4
G	1.7	24.8	57.5	17.8	30.1	69.9

The 20 litre ignition chamber has been described previously [Cashdollar, K.L. (1996)]. Briefly, the chamber is near spherical with a hinged top, which is bolted down during experiments (Figure 1a). The coal dust is weighed and placed on the dispersion nozzle at the bottom of the chamber. An electrically fired igniter is connected to the control system and the lid closed.

For tests in air the chamber was partially evacuated to ca. 0.14 bar a before a short blast of dry air 0.3 s at 7.6 bar(g) dispersed the dust and raised the chamber pressure to about 1 bar(a). After an additional delay of 0.1 s the igniter was fired. Tests in the various O<sub>2</sub> in CO<sub>2</sub> mixtures were almost identical, except that prior to ignition the chamber was completely evacuated before being back filled to ca. 0.14 bar(a) with the chosen gas.

A desktop computer sampled a strain gauge (which measures the pressure within the chamber) at a maximum rate of 9 kHz throughout the firing process. This allows calculation of the peak pressure ratio and rate of pressure rise.

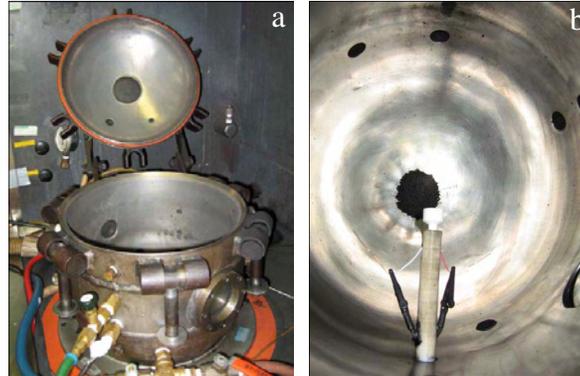


Figure 1 - NIOSH 20 litre ignition chamber a) outside b) inside (with coal and igniter)

The chemical igniters were manufactured by Fr. Sobbe, Germany. When activated a large number of hot particles are generated with little increase in gas volume. The majority of tests, using both air and O<sub>2</sub>/CO<sub>2</sub> atmospheres, were conducted using 1000J igniters. A series of additional tests were conducted with all coals except Coal F using 2500J igniters in 21.3% v/v O<sub>2</sub> in CO<sub>2</sub>. The only difference between the different igniters was the mass of chemical charge inside the capsule unit.

After the ignition experiments were completed the char residues were collected, weighed and stored. Micro-proximate analysis was then undertaken for each char sample at Imperial College with a Mettler-Toledo TGA/SDTA851e thermogravimetric analyser. Between 10 and 30mg of each sample was placed in a 75 $\mu$ l alumina crucible, before being heated under nitrogen. The samples were first heated to 105°C, before volatile content was measured by further heating the sample (at 20K/min) to 900°C. After 45 minutes, air was introduced to burn off the fixed carbon leaving just the ash and igniter residue.

Weight loss from the ignition process was calculated by collecting the sample and comparing with initial mass – as previously completed by Man et al. (2007). This method still has a number of problems, including accounting for the igniter residues and the need to determine collection efficiency. A vacuum cleaner filter was designed to aid collection. Non-woven bags were used to collect the sample, with the filter being weighed prior to, and after, utilisation. This dramatically increased the speed of experimentation.

The collection efficiency was established by dispersing and then collecting coal without igniters at concentrations varying from 100 – 600 g/m<sup>3</sup>. Separately igniters were fired on an empty chamber before coal was loaded and dispersed. Collection efficiency was then measured. The results were pooled and found to be strongly linear (and nearly identical) with a coefficient of determination about a correction equation (eqn. 1) of 0.999.

$$\text{Corrected mass} = 1.035 \times \text{mass collected} + 0.2957 \quad (1)$$

Weight loss, assumed to be from devolatilisation, can then be readily calculated to give a dry ash free value.

The Q factor can then be evaluated. It is known to be a multiplier representing the greater efficiency of volatile production/release during rapid heating compared to the standard proximate analysis [Gibbins et al. (1993)]. It is defined in eqn. 2.

$$Q_{\text{factor}} = \frac{\text{weight loss from devolatilisation (wt\%daf)}}{\text{Coal VM (wt\%daf)} - \text{Char VM (wt\%daf of original coal)}} \quad (2)$$

#### 4. Results and discussion

Whilst the higher rank coals were more difficult to ignite, all ignited in air using a chemical igniter of 1000 J. The minimum explosibility limit (MEL) in air varied, but was around 200g/m<sup>3</sup>. In this study ignition is defined as: a pressure ratio, PR > 2 and pressure rise rate, K<sub>st</sub> > 1.5 bar m/s (where PR is the maximum pressure divided by initial ignition pressure, and K<sub>st</sub> is the rate of pressure rise normalised to the explosion chamber volume).

The ignition limit, on the other hand, changed significantly with O<sub>2</sub> concentration. Only a few of the high volatile coals ignited in 21% O<sub>2</sub>/CO<sub>2</sub> and all of these required a 2500 J igniter. However, an increase in O<sub>2</sub>/CO<sub>2</sub> levels to 30 or 35% gave ignition patterns similar to those when air was the oxidant. In addition, the minimum ignition concentration decreased by about 100g/m<sup>3</sup> with increase in O<sub>2</sub> compared to the corresponding runs in air. Interestingly, further increase in O<sub>2</sub>/CO<sub>2</sub> from 35 to 40% did not significantly change the minimum ignition concentration. Above the minimum ignition concentration, the peak PR for any given O<sub>2</sub> concentration had a tendency to form a plateau independent of coal rank.

The devolatilisation results show a similar trend to the pressure ratio data. There is sharp increase in weight loss at the minimum explosibility limit that appears broadly independent of, or falls slightly with increase in coal loading. In air there appears to be a maximum weight loss of around 50 to 60% daf for most of the coals. However, there is a general trend for weight loss to increase with increase in oxygen concentration. For some coals, the weight loss in 40% O<sub>2</sub>/CO<sub>2</sub> can be as much as 20% higher than the corresponding experiment carried out in air (i.e. constant coal loading and igniter strength).

Comparing the weight loss data with oxygen concentration, it appears that the ignition pattern carried out in air are similar to those done in the ca. 30% O<sub>2</sub>/CO<sub>2</sub> mixtures depending on the coal. The low volatile coals appear to need a slightly higher amount of

oxygen (up to 35%) to match the data in air whereas the higher volatile coals in general only ‘need’ about 30% oxygen. However, this trend is difficult to discern with certainty.

The char residues from the ignition tests exhibited a fairly narrow range of apparent Q-factors (apparent, as this assumes all the weight loss was in fact due to devolatilisation alone). The lower rank coals (such as Coal A and B) have low Q-factors typically around 2. The higher rank coals (Coal C, D, E and G) exhibit slightly higher Q-factors, which range from about 2.5 to 3.5. All of the coals show a Q-factor with a definite negative slope with coal loading (shown for Coal F and G in Figure 3), though this effect is more pronounced in the higher rank coals.

The effect of O<sub>2</sub> concentration is far from clear but there does appear to be a trend of increasing Q-factor with higher oxygen concentration. It is unclear from the data how this trend is affected by variation in coal rank. The Q factor is known not to be affected by the extent of weight loss in situations where only devolatilisation is taking place. Heating rates (which would also affect Q factor) are likely to be similar in all cases, and as such it appears that the actual Q factor for the coals in these experiments is at, or lower than, the minimum value observed (allowing for experimental scatter). The recorded higher apparent Q factors then indicate heterogeneous combustion of some of the sample, proceeding in parallel with devolatilisation of other particles in the sample (and possibly, but not necessarily, in parallel with devolatilisation of the heterogeneously-combusted particles).

Although the 20-L chamber only attains temperatures suitable for heterogeneous combustion to take place for a short period of time, a small amount of char combustion cannot be ruled out given the significantly enhanced oxidation potential of an O<sub>2</sub>/CO<sub>2</sub> atmosphere compared to an O<sub>2</sub>/N<sub>2</sub> atmosphere.

To visualise the effect of oxygen on coal ignition, the data has been plotted (Figure 2) so that the effect of coal loading and rank is removed and the O<sub>2</sub> / CO<sub>2</sub> data is shown relative to that carried out in air.

Table 2 shows the oxygen concentration for performance similar to air. It also illustrates how similar the coals were in ignition behaviour.

Table 2 - Level of CO<sub>2</sub> for ignition equivalent to air

Coal	TGA VM (% daf)	% O <sub>2</sub> in CO <sub>2</sub> equivalent to air
A	44.4	30
B	33.7	25 to 30
C	32.6	35 to 40
D	30.8	30 to 35
E	31.8	30 to 35
G	25.8	30 to 35

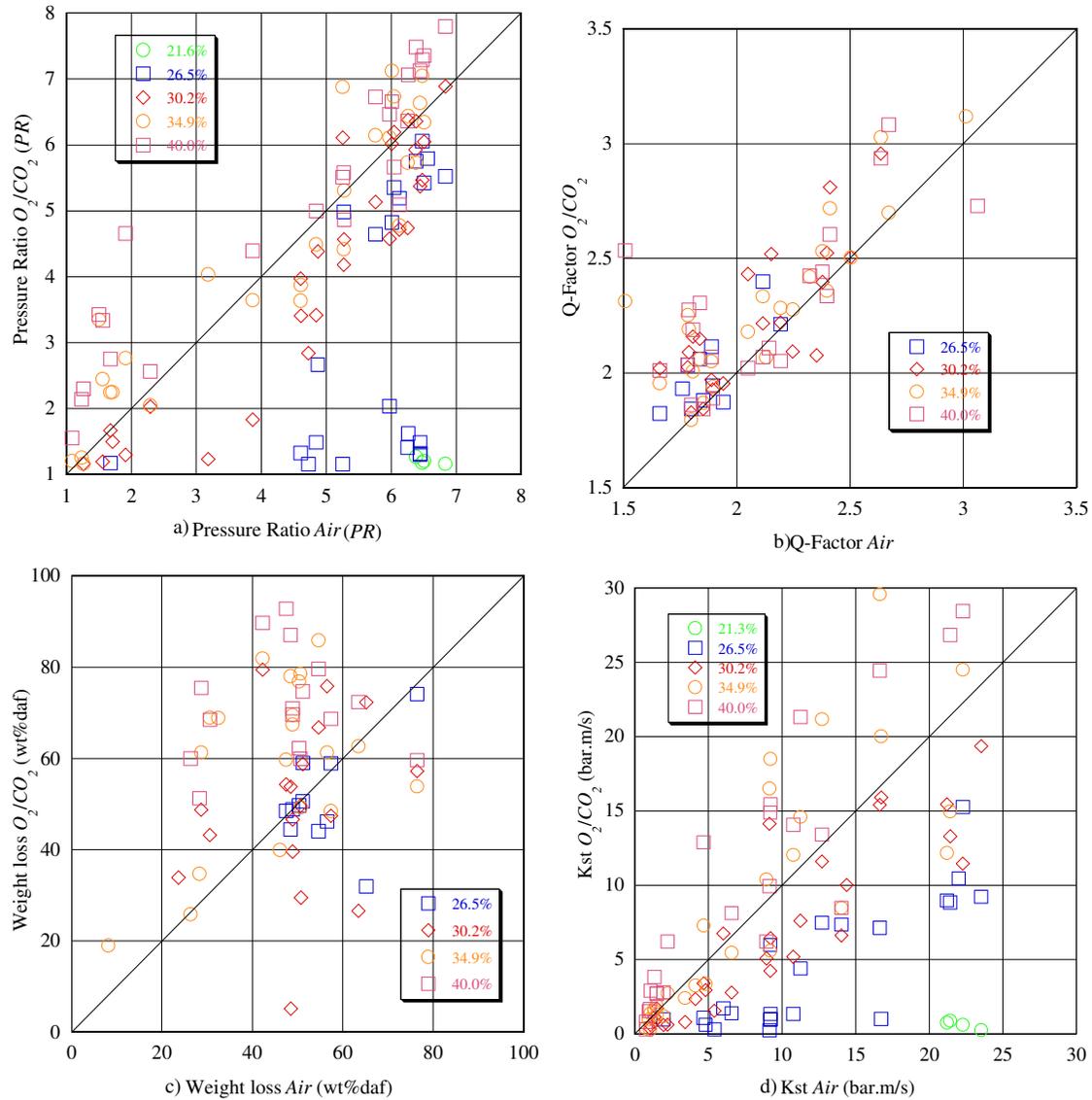


Figure 2 - Effect of oxygen concentration on ignition - cumulative results for all coals – a) pressure ratio, b) Q-factor, c) weight loss and d)  $K_{st}$

An ignition index has been applied (eqn. 3) [Man et al. (2007)], in an attempt to make the process less subjective.

$$\text{Ignition index (in air)} = \text{Max weight loss from ignition (wt\%ad)} \times PR \quad (3)$$

The results are shown in Table 3, where higher rank coals clearly have a higher ignition index. The index also rises with  $O_2$  concentration, which is consistent with the expected and observed rise in explosibility with  $O_2$  levels.

The one anomaly apparent is Coal C which apparently requires at least 34.9%  $O_2/CO_2$  to give satisfactory ignition. Apart from this a clear trend of increasing explosibility with falling rank is seen.

Table 3 - Coal ranking by ignition index

Coal	TGA VM (%daf)	Ignition Index				
		Air	26.5% O2	30.2% O2	34.9% O2	40.0% O2
A	44.4	371.3	294.5	353.0	394.1	377.5
F	36.6	293.6	-	-	-	-
B	33.7	285.7	294.8	285.0	438.5	473.5
C	32.6	182.9	no ignition	193.2	277.9	294.6
D	30.8	270.6	309.8	360.9	377.2	485.1
E	31.8	291.2	234.8	302.3	380.0	543.3
G	25.8	258.3	no ignition	215.6	327.0	407.2

The effect of coal particle size has also been examined for Coals F and G. The coals were sieved into 6 different fractions, split around 38 $\mu$ m, 53 $\mu$ m and 75 $\mu$ m. It is recognised that this process may have caused selective maceral enrichment (more likely for Coal G), although this has not been studied. The samples were tested in air at different concentrations. Evaluated Q-factors are shown in Figure 3.

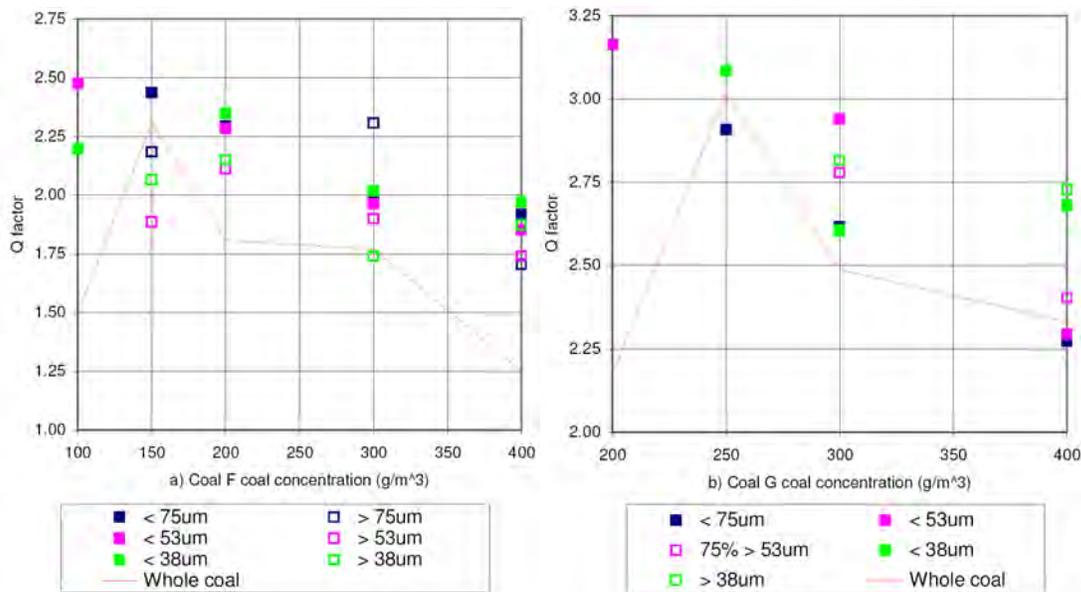


Figure 3 – Effect of particle size on Q-factor of a) Coal F and b) Coal G

The results show that for both coals, whilst the pressure ratio, K<sub>st</sub> and weight loss increase with the increasing percentage fines used and decrease with increasing sieve size the Q-factor does not appear to be greatly affected. Rather the Q-factor appears to depend more upon the coal concentration – indeed heterogeneous combustion appears to be taking place more at low coal concentrations in the Coal G case.

## 5. Conclusions

From a large number of ignition experiments it is possible to rank the coals in order of ease of ignition and to establish the effect of O<sub>2</sub> in CO<sub>2</sub> at various concentrations from 21 to 40% v/v. The ignition order followed the general trend of rising explosibility with falling rank as would be expected. The differences between the coals at higher O<sub>2</sub> levels were relatively small, although the low volatile coals showed higher than expected weight losses. An increased Q-factor is also seen where the concentration of O<sub>2</sub> in CO<sub>2</sub> increases above 21% - although there appears to be little evidence that the Q-factor continues to increase with increasing oxygen concentration. The Q-factor is also not seen to significantly vary with particle size.

The concentration of O<sub>2</sub> in CO<sub>2</sub>, which gave a similar ignition comparable to that in air, was established to be between 25 and 35%. This is a slightly broader range than reported by Tana et al. (2006) - 28% to 35% who used heat flux data; and by Man et al. (2007), 30% to 35%, whom conducted similar tests with high rank coals.

None of the coals ignited in 21% O<sub>2</sub> in CO<sub>2</sub> using a 1000J igniter, and only four of the six tested ignited using a 2500J igniter. This means the risk of explosion in mills and associated pipework is less than for air if the O<sub>2</sub> concentration in the primary stream is consistently maintained at a value no higher than that for air (i.e. ~ 21% v/v).

## 6. Acknowledgements

The activities were completed under Phase 1 of OxyCoal-UK. This was supported by the Engineering and Physical Sciences Research Council (EPSRC), whose funding is gratefully acknowledged.

The authors gratefully acknowledge the assistance of Mr. Greg Green and Mr. Jon Jobko from the CDC/NIOSH Pittsburgh Research Laboratory.

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