AN EXPLANATION OF THE LOW NO_X EMISSION CHARACTERISTICS OF AUSTRALIAN BLACK COALS

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ABSTRACT

Australia's black coals are considered to contain above average levels of elemental nitrogen. However, in similar plants under comparable conditions, Australian coals produce lower NO_X emissions than many internationally traded coals, including coals of lower nitrogen content. Previous studies have provided no explanation of this phenomenon.

High temperature laboratory equipment developed at Casella CRE Energy has been demonstrated to accurately simulate PF-combustion conditions. It has been used to successfully reproduce the NO_X release behaviour of Australian black coals and provide reasons for the lower emissions. A pilot scale test rig has also verified the results.

Coal nitrogen exists in volatile and char-bound forms. The rate of nitrogen release was characterised for selected Australian and Internationally traded coals. The faster nitrogen is released, the greater the opportunity for it to be reduced rather than oxidised. Volatile nitrogen is the major contributor to NO_X formation in uncontrolled combustion but even when low-NO_X, burners are fitted, volatile nitrogen can be converted to NO_X if its release is slow. For the Australian coals a large proportion of the total nitrogen was released rapidly during devolatilisation. However, for the International coals, nitrogen was released more slowly or more of it was retained in the char.

The conversion of char nitrogen to NO_X was also measured. The results showed the percentage conversion to be similar for the Australian and Internationally traded coals. Overall this means that, for a given nitrogen content, the Australian black coals would produce less NO_X than the International coals, when fired under comparable conditions.

INTRODUCTION

Nitrogen oxides form a minor component of the products of combustion from all processes in which fuel is used. NO_X emissions are a potential source of acid rain or are a component in the formation of photochemical smog. Hence minimisation of NO_X emissions is important.

Coal combustion gives rise to NO_X emissions, which are largely derived from nitrogen in the fuel, however in addition, thermal NO_X is formed from atmospheric nitrogen and usually accounts for about 20% of total NO_X emissions from conventional pulverised coal burners. Fuel NO_X derived from the nitrogen contained in the coal is formed as a result of extremely complex and not completely known chemical reactions. Some of the nitrogen contained in the coal is released with the volatiles whilst the remaining nitrogen remains in the char. Conversion to NO_X is dependant on combustion factors.

Based on a comparison with International coals, many Australian bituminous coals contain relatively high nitrogen contents on a dry ash-free basis, mainly within the range 1.4-2.1%. However early Australian investigations of NO_X emissions, from a range of coal-fired combustion plant, revealed that NO_X emissions were less than for many international coals having similar or lower nitrogen contents. [1], [2], [3]. This lead to the question as to what might be the cause of this measured phenomena, which was subsequently confirmed in major international power plants and industrial sites when coal from Australia was used to replace most International coals.

Investigations were undertaken [4] in order to reproduce the low NO_X phenomena at laboratory scale. However, they were unsuccessful, probably because NO_X emissions depend on coal properties, burner design and operation. It was assumed that both combustion intensity and oxygen availability in the laboratory furnace rig overwhelmed the nitrogen release characteristics of the coal and hence masked what was happening in practice.

It was subsequently found that developments undertaken at laboratory and pilot scale at CRE were aimed at examining the formation of NO_X from the combustion of coal [5]. In consequence it was decided to undertake studies on a range of Australian bituminous coals using the techniques developed by CRE in order to explain the reason for their lower NO_X emission characteristics, compared with many International coals.

BACKGROUND

CRE have been involved in NO_X related studies for many years. Detailed studies were carried out on volatile and nitrogen partitioning and an extensive database was established, primarily concerning the behaviour of British coals. This data has been used to assist in the development of a correlation of NO_X formation with coal properties. More recent projects on coal reactivity and NO_X reduction have also been carried out. These studies have examined characteristics such as devolitilisation rates [6], NO_X reduction reactions [7] and char reactivity for a range of internationally traded coals. In particular CRE were participants in the UK Department of Trade and Industry collaborative NO_X project [8], [9]. This project was a major UK initiative aimed at improving the prediction of NO_X formation from a full sized power plant. The partners included two major UK power generators, equipment manufacturers, mathematical modellers and academic institutions. The program involved the evaluation of eight International coals and comparison of laboratory and full-sized plant tests. CRE carried out tests on each of the coals studied during this program and obtained results that compared favourably with full-scale data. For these reasons CRE were selected to undertake studies to characterise a suite of Australian coals and to hopefully explain the reason for most Australian bituminous coals producing lower NO_X emissions than their nitrogen content would indicate.

Experimental techniques chosen for the evaluation of coal performance in a pf environment at laboratory scale must be able to simulate the combustion conditions. Because of the difficulty of achieving the required high combustion temperatures, residence times and gas environments, laboratory based techniques are required to compromise in some areas of boiler simulation. However, CRE have expended significant development effort on a number of techniques and those chosen for the investigation were drop tube furnaces and flat flame burners. Both techniques have the required ability to reach representative operating temperatures, attain high particle heating rates, have controllable residence times, to use various combustion atmospheres and to provide an accurate method for particle collection.

The laboratory studies used by CRE to examine the suite of Australian coals are a Laminar Flat Flame Burner (LFFB), and Optical-Access Drop Tube Furnace (OA-DTF) and a NO_X Reduction Test Facility (NRTF). The LFFB is used to characterise the devolatilisation of the coal in detail, at volatile release rates of the coal below 35 ms. The OA-DTF is used to investigate both the devolatilisation and combustion of pulverised coals at longer residence times. The NRTF is a pilot scale test rig that was purpose built to study parameters likely to effect staged combustion processes. The test facility simulates the conditions typical of a pulverised fuel fired boiler fitted with low NO_X burners. A description of all three test rigs used in the experiments is given in detail elsewhere [10].

The coals used in the study were selected from a wide range of available coal samples on the basis of laboratory standard analysis data. The coals were selected to include a range of proximate analyses and nitrogen contents and also to include coals from different mining areas of eastern Australia.

TEST PROGRAMME

The Australian coals selected for the study were designated Alpha, Beta, Gamma, Delta, Theta, Lambda, Sigma and Omega. The coals were prepared as pf, specified as 70% passing 75 µm, representing the nominal size distribution of coal obtained from power station pulverisers. Representative pf samples were collected and prepared for the various stages of testing by oven drying at 40^{o}C. Samples of the coals were also sifted to various size fractions, for the LFFB test and for the OA-DTF tests. All prepared coal was stored below 0^{o}C after drying to prevent adsorption of moisture.

All coals were tested in the LFFB, which consists of a burner designed to produce a narrow lamina flame a few centimeters long with a controllable velocity, temperature profile and composition. Coal is injected into the centre of the flame and particles are heated rapidly at rates characteristic of a pf boiler. Only small amounts of oxygen are free to react with the coal, since most of the oxygen is consumed by the burner’s hydrogen fuel supply. Any remaining oxygen reacts preferentially with the volatiles as they are released and so there is little char combustion. Hence the main process occurring in the flame is coal devolitisation. In this study, the LFFB was used to characterise the devolitisation of the coal in detail, characterising the volatile release rates of the coal below 35 ms. By collecting and measuring the nitrogen content of the chars produced, the volatile nitrogen release rate can also be calculated.

The method used to obtain the kinetic rate data is as follows. The burner is set at the test conditions specified for the test. In each case in this study, the temperature was 1350^{o}C, which represents the temperature in the near burner field of a typical low NO_X burner and the oxygen is set to nominally 0% (i.e. a stoichiometry of 1.0 with respect to the burner’s hydrogen fuel supply). Coal is then fed while samples are collected at positions equivalent to residence time of 5, 10, 15, 25 and 35ms. For each sample the amount of volatiles released is determined by an ash tracer method and the sample is analysed for carbon, hydrogen and nitrogen. Once the total volatile release is known the amount of volatile nitrogen release can be calculated from the amount of nitrogen remaining in the char. A Weibull function curve is fitted to the data obtained for total volatiles and volatile nitrogen over the full 35 ms residence time range. Initial kinetic rates are calculated by constructing tangents to the initial 5 ms of the fitted curves produced.

The OA-DTF was used to obtain three important parameters. Firstly, a volatile yield was obtained at 150ms at 1350^{o}C. Secondly, a burn out profile was obtained at 1500 ^{o}C and at 5% oxygen. Thirdly, devolatilised char was produced at 150ms at 1350 ^{o}C, which was then re-fired at a range of temperatures, oxygen levels and residence times.

The high temperature volatile yield is a better indication of coal performance than the proximate volatile yield. 1350 ^{o}C is a representative temperature for the internal recirculation zone of a low NO_X burner. The internal recirculations zone operates at reduced stoichiometry and so nitrogen released into this zone is unlikely to be oxidised to $NO_{X.}$ Furthermore, the higher the total volatile release into this zone the lower the oxygen concentration that results. Thus the measure of the total volatile yield and the split of nitrogen between the volatiles and the resulting char is an important parameter. These parameters are measured by carrying out a furnace test with the residence time set at 150ms. It cannot be assumed that coal particles would spend more than 150 ms in a reducing zone before being mixed with tertiary air.

Burnout profiles are analogous to “burning profiles” produced by thermogravimetric techniques for characterising coal reactivity. However, instead of monitoring weight loss over a period of time, samples are collected at intervals of time and can be analysed to determine the amount of combustibles remaining in the coal. The amount of nitrogen released can also be measured. The advantage of obtaining a drop tube furnace burnout profile is that the flow through the reactor system ensures that reducing reactions, which take place in packed bed systems, do not occur and particle temperatures are related to the combustion characteristics of the coal., Drop tube tests are carried out at 1500 ^oC, which is representative of the conditions in the boiler above the burner where combustion of the volatiles is completed and most of the non-volatile material is burnt. Only the inlet oxygen is controlled and is selected at 5%, since for most coals this gives an exit oxygen level in the region of 3% that is representative of boiler exit gas concentrations. As a result, coal particle temperatures, which are heavily dependant on oxygen, also reach representative levels. Data is fitted with a curve and the coals are ranked against other coals whose performance at full scale has been characterised. In this way likely boiler burnout performance can be predicted.

The above parametres give an indication of how much coal and nitrogen can be reduced by low NO_X burners and how much of the coal remains unburnt. The additional information required to predict NO_X emission propensity is the proportion of the nitrogen released from the char during char combustion that is oxidised to NO_X. During combustion, nitrogenous species undergo complex reactions with the char surface and with carbon monoxide and hydrocarbon species surrounding the char particles. Numerous reactions can take place and nitrogen species released from the coal can take various forms depending on coal type. Chemical changes cannot be measured directly and if they could it would be at unrepresentative conditions. However, the char nitrogen to NO_X conversion provides a convenient way of measuring the overall impact of the reactions and generating a parameter that can be combined with other data obtained to estimate NO_X formation propensity. Nitrogen to NO_X conversion is calculated as the percentage of the nitrogen release from the charge during char combustion that is converted to NO_X.

The NRTF is operated with two coal combustion zones: a reducing zone for reduction of nitric oxide and a burnout zone for completion of the combustion of the fuel. The reducing zone is a simulated coal combustion gas mixture, produced by a 25kw premix propane-air burner incorporated into the upper part of the furnace. The rig permits coals to be tested at identical oxygen concentrations, temperatures and pre-combustor NO_X levels so that variations during tests are due to the coal and not to rig conditions. For this study, a temperature of 1350^oC at the inlet was selected, which gives higher temperatures in the coal burning region; again typical of a pf boiler. The measured parameters are the coal burnout, the exit NO_X level and the reduction propensity. This information is obtained for different exit oxygen levels (3% and 5%) and for different reducing zone stoichiometries.

RESULTS

Coal Analysis

Sub-samples of the coals were subjected to chemical analysis by standard laboratory methods and the results are given in Tables 1 and 2.

Coal	Inherent moisture (% ad)	Ash (% ad)	VM (% ad)	FC (% ad)	Ash (% db)	VM (% db)	VM (% daf)
Delta	2.75	8.95	19.20	69.10	9.20	19.74	21.74
Theta	3.01	10.97	34.14	51.88	11.31	35.20	39.69
Beta	7.07	7.92	26.63	58.38	8.52	28.65	31.32
Alpha	2.09	13.44	29.36	55.11	13.72	29.99	34.76
Gamma	14.26	6.24	29.64	49.86	7.28	34.57	37.28
Lambda	1.86	12.89	29.34	55.91	13.14	29.90	34.42
Sigma	1.20	17.47	21.55	59.78	17.68	21.81	26.50
Omega	6.69	11.28	39.86	42.17	12.09	42.72	48.60

Note: Volatile cake powdery for Delta, Beta and Gamma, sintered for all others

Table 1: Coal Proximate Analysis

Coal	C (% ad)	H (% ad)	N (% ad)	N by Perkin Elmer (%ad)	S (% ad)
Delta	76.94	3.76	1.64	1.6	0.38
Theta	71.36	4.63	1.57	1.6	0.89
Beta	70.58	4.18	1.56	1.6	0.34
Alpha	70.58	4.14	1.45	1.7	0.56
Gamma	60.41	4.97	1.10	1.1	0.60
Lambda	71.32	4.70	1.95	1.9	0.65
Sigma	70.37	3.89	1.62	1.7	0.60
Omega	64.17	5.51	0.86	0.9	0.42

Note: Hydrogen not corrected for inherent moisture

Table 2:Coal Ultimate Analysis (British Standard methods except where stated

Char Analysis & Determination Of Volatile Yield, Char Nitrogen Yield & Burn-Out

The coals fed to the LFFB, OA-DTF and the NRTF, together with their corresponding chars are ashed in a muffle furnace at 815°C for two hours. The dry ash content of the coals and chars are used to calculate the volatile yield or burnout (depending on the test conditions) using the equation:

$$\%\,Yield\ V_V\ or\ Burnout(\%daf) = \frac{a_2 - a_1}{a_2(100 - a_1)} x\,10^4$$

where a_1 = % dry ash content of coal
a_2 = % dry ash content of char.

The nitrogen remaining in the char, expressed as a percentage of the nitrogen in the dry ash free coal, is calculated using the equation:

$$\%\,Char\ Nitrogen\ Yield\ C_N = 100 - \frac{100\,N_1 - N_2(100 - V_V)}{N_1}$$

where N_1 = % dry ash free nitrogen content of coal
N_2 = % dry ash free nitrogen content of char.

Coals and chars are also subjected to carbon, hydrogen and nitrogen analyses using a Perkin Elmer Elemental Analyser. The instrument operates by burning the sample in oxygen, passing the combustion gases through a reducing column and measuring the products using a chromatographic column fitted with a thermal conductivity detector. The values obtained are corrected for ash. The coals are dried at 40°C prior to testing and chars are rapidly quenched after reaction. Both are stored in the presence of a desiccant to prevent adsorption of ambient moisture. The basis of the ash corrected results is thus assumed to be dry and ash free. The results are given in Table 3.

Coal	Initial volatile release rate (10^2g volatiles g^{-1} coal ms^{-1})	High temperature volatile yield, V_V (%daf)	High temperature nitrogen yield, V_N (%daf)	V_N/V_v
Alpha	0.87	51.15	81.1	1.59
Beta	2.95	57.86	75.9	1.31
Gamma	4.82	73.26	70.8	0.97
Delta	1.68	38.13	76.6	2.01
Theta	0.95	62.34	76.6	1.23
Lambda	4.32	54.72	80.3	1.47
Sigma	4.10	47.67	79.6	1.67
Omega	1.62	65.86	70.2	1.07

Initial volatile release rates were calculated from the results of LFFB tests over the range 5 to 35 ms at 0% oxygen and 1350°C. Total volatile yields (V_V) were obtained using the OA-DTF at the same oxygen and temperature conditions and a residence time of 150 ms. The data is shown in Table 3 and the volatile release profiles are plotted in Figure 1.

Table 3: Volatile Release Data from LFFB and OA-DTF

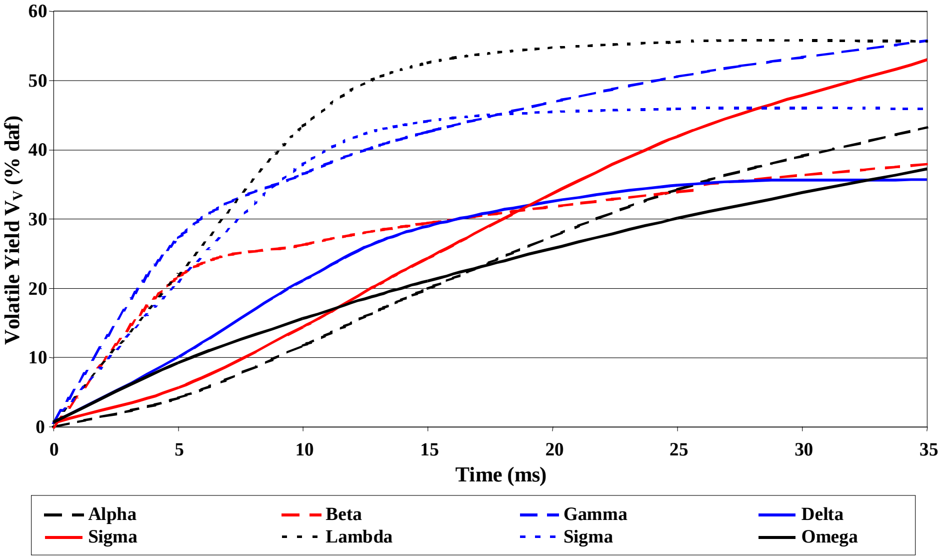

Figure 1: Volatile Release Profiles Obtained from LFFB

The measurement of V_V represents the first step in predicting levels of NO_X that the coals would be expected to produce at full scale. In terms of NO_X emissions, and flame stability, high volatile release rates are also a desirable feature. Coals having lower release rates may give rise to higher stoichiometries in the internal recirculation zone of a low-NO_X burner and thus to poorer NO_X reduction. They may also undergo less intense combustion when air becomes available, lowering temperatures locally and resulting in slower release of nitrogen. In turn this increases the chance that nitrogen will be oxidised to NO_X. Initial volatile release rates are typically in the range 3.2×10^{-2} g volatiles g^{-1} coal ms^{-1} for many high-volatile, bituminous UK coals. Volatile-release rates substantially below this are considered to be low and rates above this value are considered to be high. Similarly, V_v at 150 ms has an average value of about 51 % for a large number of high volatile UK bituminous coals and values higher than this will promote lower NO_X..

Summarising the results on the basis of total volatile release rates and total volatile yields allows the following statements to be made:

- Sigma and Lambda give average volatile yields but have high release rates, so they should give acceptable flame stability and potentially low NO_X levels.
- Beta is very much like UK coals. It has an average volatile release rate and the volatile yield is high so the coal would be expected to give acceptable NO_X levels.
- Omega and Theta have high volatile yields but the release rates are quite low. They are likely to perform acceptably overall in a boiler with low-NO_X burners.
- Gamma has a high volatile yield and volatile release rate and should perform well.
- Alpha and Delta have medium and low volatile yields respectively and low release rates. NO_X levels for these coals cannot be predicted on this evidence alone.

In order to make accurate predictions both the total volatiles and the volatile nitrogen must be taken into account. Table 3 also summarises the volatile nitrogen yields (V_N) obtained together with the ratio of V_N to V_V. The data is plotted in Figure 2. The values for total volatile nitrogen yield at 150 ms are all above the average value of about 54 % for a large number of high volatile UK bituminous coals. Omega and Gamma have notably lower volatile nitrogen yields than might have been expected by considering the total volatile yields alone. However the values obtained for V_N are still favourable compared with many Northern Hemisphere coals which are known to give acceptable levels of NO_X. Alpha, Lambda and Sigma exhibit exceptionally high values of V_N. Lambda (1.95) and Sigma (1.62) have quite high levels of nitrogen in the coals which might be of considerable concern when considering the coals for power station use. These levels of V_N do not suggest that NO_X should be a problem.

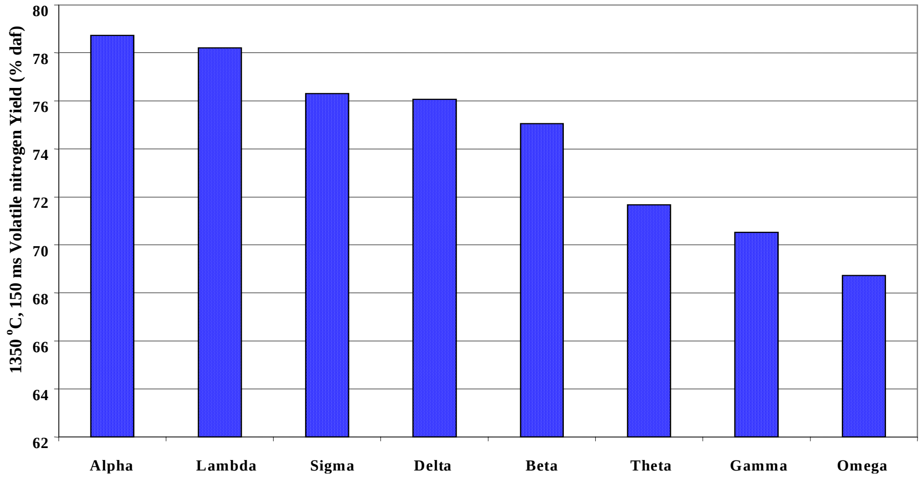

Figure 2: High Temperature Volatile Nitrogen Yields from OA-DTF

It is possible to measure the nitrogen conversion of the coals. Nitrogen-to-NO_X conversion is simply calculated by a mass balance from OA-DTF data, using as input the mass of char entering the rig per unit time, gas flow rates, the concentration of NO_X in the exit gases and the amount of nitrogen remaining in the unburnt residue collected at the outlet. The values obtained depend on the conditions at which the measurements are made and so a range of conditions is studied. For illustrative purposes the results obtained at 1350°C and 5% oxygen are presented in Figure 3 but the full data set includes data for additional tests at 1200 and 1500°C at 5% oxygen and at 1350°C at 10% oxygen.

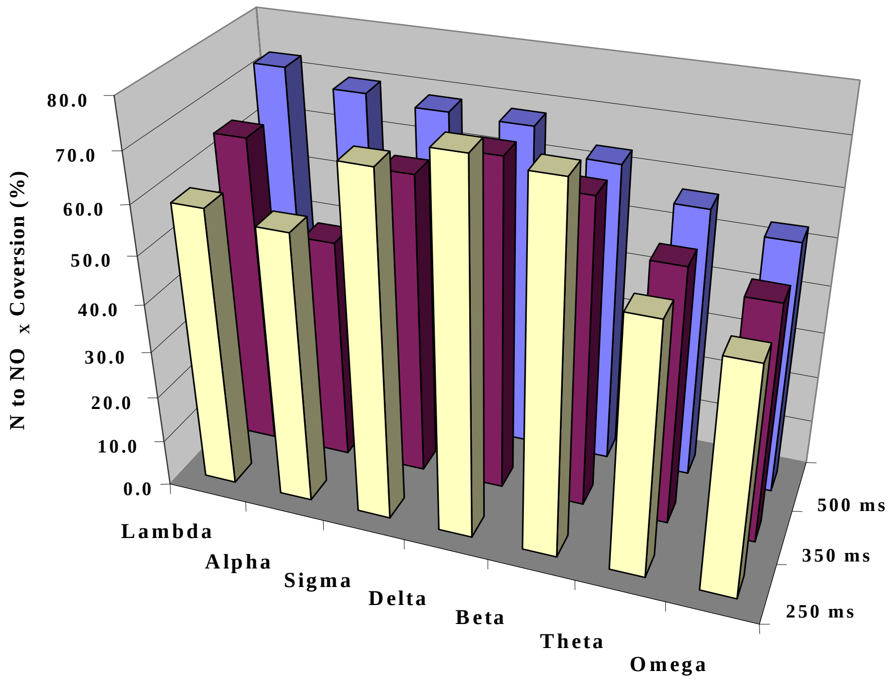

Figure 3: Char Nitrogen to NO_X Conversion Values at Various Residence Times

Taken with volatile nitrogen yields and release rate data, the results show that the overall performance of most of the coals studied is very good. A slow nitrogen release rate has been shown to result in up to 20% of the nitrogen released between 35 and 150 ms being converted to NO_X. This explains the lower nitrogen to NO_X conversions of Alpha, Theta and Omega. The slow release rate of Delta has little effect on its overall nitrogen to NO_X conversion because the V_N to V_V ratio is very high and nitrogen is preferentially released with the volatiles.

In the NRTF three of the coals predicted to produce differing levels of NO_X were tested with the reducing zone stoichiometry varied between 0.7 and 0.9, whilst the excess oxygen and NO_X level in the pre-combustor flame was kept constant. Burnout air was added to achieve exit-oxygen levels of either 3% or 5%. The results clearly showed that Omega gave the lowest overall NO_X levels at both excess oxygen levels. The NO_X levels obtained were in the range 300 to 320 ppm, some 50 to 60 ppm lower than for Alpha at 3% oxygen, which equates to a 15% difference in NO_X. Lambda gave the highest NO_X level at 3% excess oxygen, producing approximately 20 ppm more NO_X than Alpha. All three coals gave higher levels of NO_X at 5% excess oxygen than at 3% excess oxygen but the ranking order of the three coals remained the same.

Standard laboratory analyses suggest that Lambda should give up to a third more NO_X than Alpha but the overall conversion derived from volatile release rates, nitrogen partitioning and char nitrogen to NO_X conversion suggest only a difference in the region of 5%, much closer to the pilot scale test results. Conversely, based on nitrogen content, Alpha would be expected to give almost 70% more NO_X than Omega. However, the mean nitrogen conversions for test results at all conditions are 53.7% for Omega and 59.1 for Alpha. Thus the difference in NO_X emissions is predicted from laboratory scale to be only about 10%, much closer to the 15% difference observed at pilot scale.

CONCLUSIONS

It was demonstrated that the advance laboratory scale techniques for the prediction of NO_X and burnout used by CRE can be successfully applied to Australian coals. All of the Australian coals could be evaluated and ranked for their propensity for NO_X emission and carbon burnout using the laboratory scale techniques. However, one of the Australian coals was outside of the normal range of coals generally accepted for pf power station boiler use, due to its low volatile matter content and as a result some uncertainty remained concerning its ultimate carbon burnout.

For the coals studied the yield of volatile nitrogen is the most significant factor in contributing to the prediction of low NO_X emissions. Char nitrogen to NO_X conversion was also shown to be highly significant in the assessment of one coal, which may indicate that the forms of nitrogen present in Australian coals play a significant role in the nature of nitrogenous species formed during combustion. Measurement of these two parameters alone should give a good initial indication of the suitability of other untested Australian coals for use in pf boilers fitted with low NO_X burners.

The use of the CRE laboratory and pilot scale test methods enable predictions to be made from small coal samples for NO_X emission propensity and carbon burnout. It was confirmed that NO_X emissions from Australian coals cannot be predicted from fuel nitrogen contents and that most Australian bituminous coals produced lower NO_X emissions than the nitrogen content of the coal would indicate. For the Australian coals a large proportion of the total nitrogen was released during devolitilisation. However, for the International coals tested, nitrogen was released more slowly or more of it was retained in the char. Conversion of char nitrogen to NO_X showed the percentage conversion to be similar for both the Australian and the Internationally traded coals. Overall this means that for a given nitrogen content the Australian black coals would produce less NO_X than the International coals, when fired under comparable conditions.

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