

The Effect of Dewatering on the Combustion Behaviour of Loy Yang Coal in a Drop Tube Furnace

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Abstract

The Mechanical Thermal Expression (MTE) process is an efficient means of removing water from low-rank coal, however the effect of dewatering on the combustion properties of lignite is largely unknown. This work examines the burnout performance of both dewatered lignite and untreated lignite in a drop tube furnace. The results show that combustion of the MTE-processed lignite is approximately 20% slower under conditions relevant to pulverised-fuel flames. This is apparently due to the reduction in pore volume which occurs during coal dewatering by the MTE process.

1 Introduction

Large deposits of lignite in Victoria are an important energy resource for electricity generation in Australia. However, the high water content of these coals results in low process efficiencies and causes high greenhouse gas emissions from conventional pulverised-fuel boilers.

Mechanical Thermal Expression (MTE) is an efficient means of pre-drying lignite, utilising mild temperatures and pressures to remove water as a liquid. Utilisation of low-moisture MTE coal in current lignite-fired power plants could yield substantial reductions in greenhouse gas emissions, however retrofit modifications to the boilers would be necessary due to the changed properties of the dewatered coal. To investigate the impact of MTE on coal combustion, and to optimise potential boiler modifications, a mathematical furnace model is currently under development [1]. Knowledge of char combustion is a vital part of this model.

Previous studies using Victorian and South Australian lignite indicate that the MTE process removes 25 to 60% of sodium, and 5 to 50% of magnesium from the coal, with the higher removal achieved for lignite with higher initial inorganic content [2, 3]. Since the concentration of alkaline and alkaline earth species has been shown by many researchers to increase intrinsic char reactivity [4], it is likely that MTE-coal will have a lower intrinsic reactivity. Other work within our Centre has also shown that the MTE process compresses the pore structure of the coal causing a reduction in macro-pore volume with a corresponding increase in meso-pore volume [5]. This change in pore volume may reduce the rate of char combustion since the rate of oxygen transport limits the combustion rate at high temperatures. Under pulverised-fuel combustion conditions both intrinsic reactivity and oxygen transport play an important role in determining the combustion rate [6]. Thus it is expected that the combustion rate of MTE coal would be slower than unprocessed lignite due to the combined effects of reduced catalytic species and slower oxygen transport through the pores.

Drop tube furnaces have been used extensively in the literature to study coal combustion [6, 7] since they reproduce conditions similar to the combustion environment in a pulverised-fuel flame and they are relatively simple to operate under controlled conditions. In this work, a drop tube furnace has been used to investigate the combustion rate of both MTE-processed, and unprocessed, Loy Yang coal under pulverised fuel conditions.

2 Experimental methodology

2.1 Measurement of conversion for low-ash lignite

Carbon conversion in a drop tube furnace can be determined using coal ash as a tracer. Thus the conversion (%) is calculated from an ash balance of the coal [8] so that:

$$W = \frac{10^4(A_1 - A_0)}{A_1(100 - A_0)} \quad (1)$$

Where W is the conversion (%), A_0 is the ash content of the feed coal (%), dry basis, and A_1 is the ash content of the collected char (%), dry basis.

This technique is problematical when using low-ash coals since the uncertainty associated with the ash measurements (A_0 and A_1) in Eqn. 1 leads to a large uncertainty in the conversion (W). Further errors also occur with this technique for Victorian brown coals due to the high content of volatile non-mineral inorganic species since these species are not completely accounted for in the solid ash. To overcome these potential errors, a modified ash tracer method has been developed which uses the addition of artificial ash into the low-ash coal as a tracer. The development and validation of this method is reported elsewhere [9].

2.2 Coal Preparation

Expanded perlite (Ausperl P200, Australian Perlite) is used as an artificial ash and is added to the coal. The perlite has a similar density to the char particles and is an inert solid at the temperatures used in this study. The expanded perlite was screened to 90-125 μm and dried in an oven for an hour at 110°C and then cooled in a desiccator. The perlite was found to contain 0.25 % moisture, and 97.6 % of the sample was retained as ash (dry basis) on heating to 800°C.

The coal used in this study is Loy Yang coal from Australia. The typical properties of the coal are shown in Table 1. Samples of coal were prepared from run of the mine coal milled to less than 3mm in a Van Gelder swing mill. The samples were either air dried, or dried using a small MTE continuous rig followed by further air drying. The operation of the MTE apparatus are described in detail elsewhere [10],

and the operating conditions used were 150°C process temperature, 12MPa effective pressing force with a press time of 1 minute. Dried coal samples were crushed in a rotary mill to less than 500µm and then screened to a size fraction of 90 to 125µm.

Moisture	58%
Ash	0.97 % db
Volatile matter	50.5 % daf [11]
Fixed Carbon	49.5 % daf [11]

Table 1: Proximate analysis of Loy Yang coal (db = dry basis, daf = dry, ash-free).

2.3 Experimental Apparatus

The drop tube furnace used in this work is located at ACIRL in Brisbane. A schematic of this facility is shown in Figure 1. The furnace consists of a heated tube of 50mm internal diameter and 1000mm length. Water-cooled collection and feed probes are inserted into the furnace to give an effective tube length which can be varied between 121 and 400 mm.

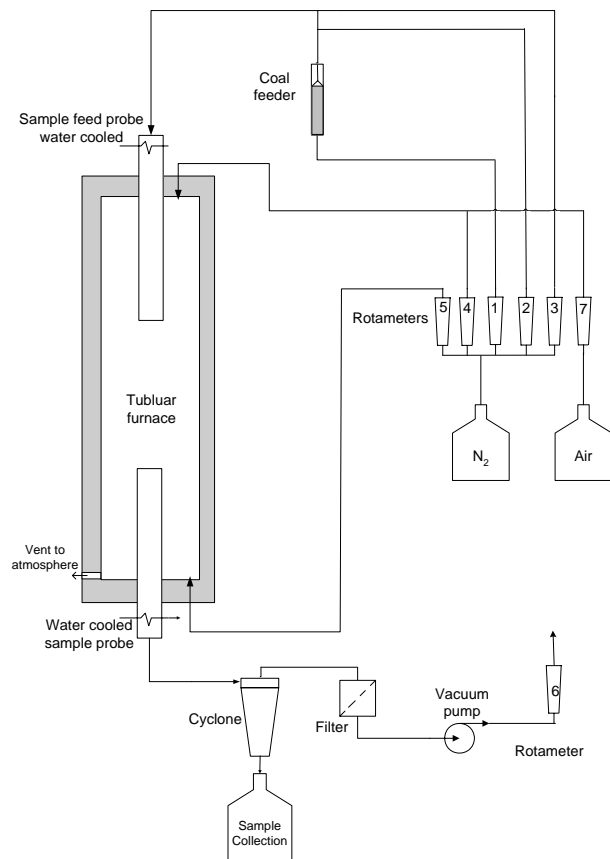


Figure 1: Schematic diagram of the ACIRL drop tube furnace.

A coal and perlite mixture (~5 g) was prepared and mixed thoroughly with a spatula. The feed mixture consisted of screened coal, either MTE processed or unprocessed, with approximately 10% perlite added and was fed to the furnace using the customised feeder. The feeder consists of a cylindrical bed of coal, with a slow flow of nitrogen through the bed (rotameter 1). A fixed cone is located at the top of the bed to collect particles and entrain them in the coal feeding line. The coal feed rate was controlled by regulating the speed at which an electric motor raised the coal bed. A vibrating

device was attached to the coal feeder to encourage an even flow of coal. Additional flows of nitrogen were blended into the feed line. The coal feed rate was measured before and after each test by weighing the mass of particles collected over a given time from the coal feed line, and was compared to the total mass of coal fed to the furnace during each experiment. The coal feed rate was maintained at between 6.3 and 7.5 g/hr for each of the experimental runs. In each experiment, all of the coal feed mix was fed through the furnace to ensure accurate closure of the mass balance. The coal feed was injected into the furnace through the water-cooled sample feed probe.

The majority of gas is added to the furnace through the annular region at the top of the furnace thus allowing for the gas to be preheated to the furnace temperature. The relative flow rates of air and nitrogen were such that the initial oxygen concentration in the feed to the furnace was 5 vol%.

Char was collected by the water-cooled sample probe and then separated from the gas in a cyclone separator. A paper filter unit is installed after the cyclone, however minimal char was collected by the filter paper. Gas flow through the char collection probe was maintained by a vacuum pump, and adjusted such that the flow rate through the line slightly exceeded the combined flow rate of gas supplied to the top of the furnace and the feed probe.

Additional nitrogen was supplied to the furnace through the base (rotameter 5). The purpose of this flow was to prevent the ingress of oxygen due to the slightly negative pressure at the bottom of the furnace caused by the operation of the sample pump. Excess nitrogen supplied at this point exits via the vent in the bottom of the furnace.

The furnace temperature was varied from 900 to 1100°C, typical temperatures for brown coal combustion. The effective furnace length was varied from 120 to 400mm, corresponding approximately to residence times between 250 and 1000 ms.

Coal conversion due to combustion in the drop tube furnace is calculated using Eqn. (1). The char collected from the furnace was tested for ash content (A_f) using the method outlined in Australian Standards 2434.8-2002 [12]. The ash content of the feed coal-perlite mix (A_0) was calculated using Eqn. (2).

$$A_0 = (1 - x_p)A_c + x_p A_p \quad (2)$$

Where x_p is weight fraction of perlite in the feed coal-perlite mix, A_c is the ash content of the parent coal (% dry basis), as measured by the standard ash test for low rank coal [12], and $A_p = 97.6\%$ is the ash content of the perlite (dry basis) allowing for the 2.4% loss on heating.

2.4 Char characterisation

The drop tube furnace was also used to produce char by reducing the air flow in the furnace and replacing it with additional nitrogen to reduce the oxygen concentration to 0.5-1 vol%. Screened samples of both MTE and unprocessed coal were fed to the furnace without the addition of perlite. The furnace temperature for these runs was 1100°C and the residence time was approximately 250ms. The pore structure and micro-porous surface area of these chars were measured using mercury intrusion porosimetry and CO₂ adsorption, respectively.

3 Results and Discussion

The conversion of coal due to combustion in the drop tube furnace, as a function of furnace temperature, is shown in figure 2 for a residence time of 460ms. It can be seen that the conversion of the MTE coal is approximately 20% less than that of the unprocessed lignite at each furnace temperature investigated. Figure 3 also clearly shows that the measured combustion rate of the MTE coal is less than that of the raw lignite.

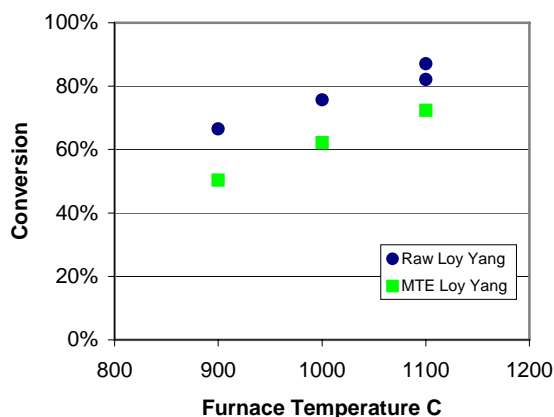


Figure 2: Measured conversion of tested coals at residence time of 460ms

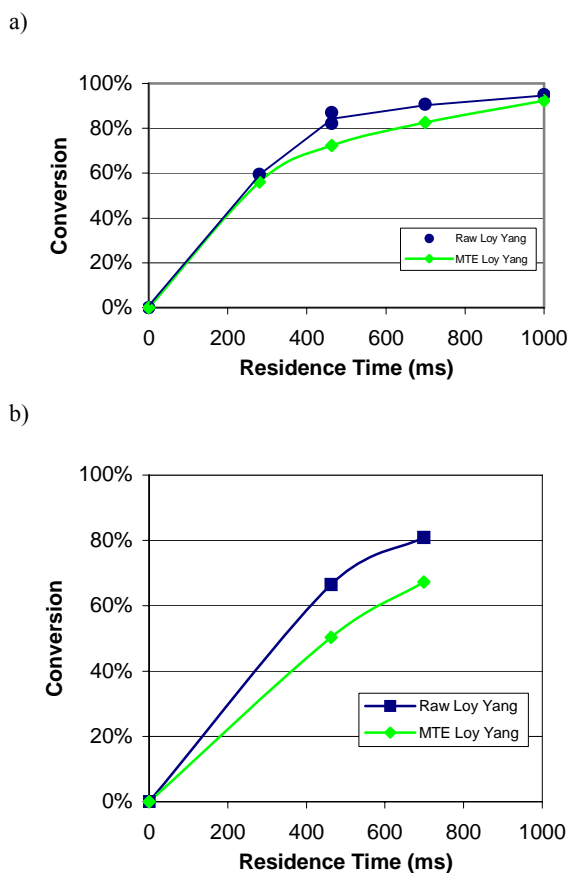


Figure 3: Conversion profile of MTE-processed and unprocessed Loy Yang coal with furnace temperature: a) 1100°C, b) 900°C.

Figure 3a shows that the measured conversion at the lowest residence time at 1100°C is similar for both the MTE and the unprocessed lignite. This suggests that the pyrolysis behaviour of both coals is similar. While the same behaviour is not observed in figure 3b, the shortest residence time studied for the 900°C test is almost double that of the 1100°C test. Due to the nature of the tests, both char combustion and pyrolysis occur during the experiment. Future work will investigate the pyrolysis separately to the char combustion by examining the conversion in the drop tube using an oxygen-free gas stream.

It can also be observed from figure 3 that at 1100°C the extent of conversion is similar for both coals after approximately 1000ms. This could be due to thermal deactivation of the char, which has been shown by others to significantly slow the rate of combustion as conversion nears completion [13]. This suggests that as the chars approach full conversion the char reactivity drops to such an extent that the conversion versus time curves begin to converge.

It appears from the combustion profiles of figure 3 that the two coals behave similarly during pyrolysis. This suggests that the near flame region of a pf-coal flame might be similar for both coals. However, since the combustion rate of the MTE coal is slower than the untreated lignite, it is expected that in the far-flame region the MTE coal will result in a lower heat flux and a longer flame length.

Mercury intrusion porosimetry tests were performed on chars made from the two coals and the results are shown in figure 4. It can be seen that the pore volume of the char from raw Loy Yang coal has a higher pore volume than that of the char made from the MTE coal. This difference in pore structure will result in slower oxygen transport within the MTE char and is consistent with the reduced combustion rate observed for the MTE coal relative to that of the unprocessed coal.

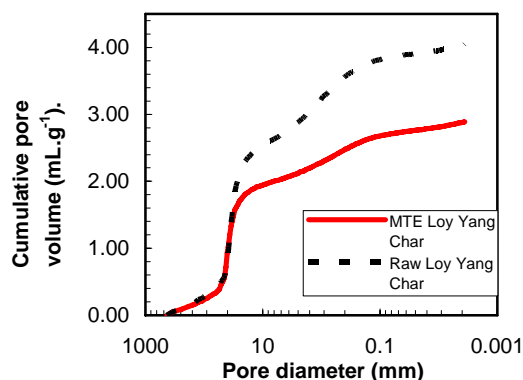


Figure 4: Cumulative pore intrusion of char samples obtained from mercury intrusion tests.

The micro-porous surface area of the two chars was investigated using CO₂ adsorption and calculated using the Dubinin equation [14]. For both the chars the measured surface area was essentially the same at approximately 600m²/g. This suggests that while the MTE coal has the same surface area available for reaction as the unprocessed lignite this surface is less accessible to oxygen at the elevated temperatures investigated and hence a lower combustion rate is observed. There may also be an effect from the reduced catalytic species in the MTE coal, however other work done in our laboratory suggests that this is negligible [15].

Coal pore structure has a major influence on char reactivity at higher temperatures. Knill et al [7] have modelled char combustion in a drop tube furnace with reasonable accuracy for a suite of coals using the char pore structure as measured using mercury intrusion porosimetry, N₂ BET surface area, and particle size as the only inputs to the model, and assuming a constant char intrinsic reactivity. The model of Knill has been applied to the pore structures measured in this work, with an assumed N₂ BET surface area of 110 m²/g for both chars. Figure 5 shows the results of this model for the chars derived from MTE and raw Loy Yang coal. The model assumes a constant particle temperature and that particles burn with reducing diameter according to equation 3.

$$d_p = d_{po} (1 - X)^{0.25} \quad (3)$$

Where d_{po} is the initial particle diameter, and X is the fractional conversion.

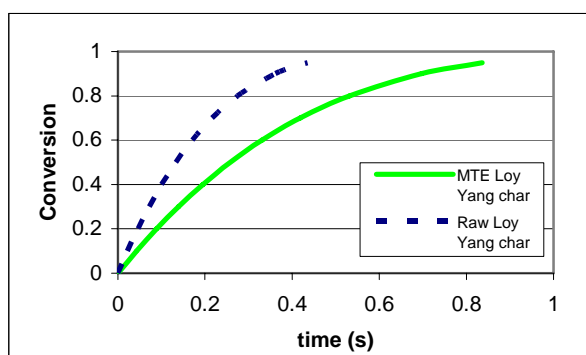


Figure 5: Predicted coal conversion using the model of Knill; based on measured mercury intrusion data and an assumed particle temperature of 1100°C.

Figure 5 shows that the combustion rate of the MTE char is predicted to be considerably slower than that of the char from the unprocessed lignite, based on the measured mercury intrusion data. While the model predictions qualitatively support the trends observed in figure 3a, a number of assumptions have been made in applying the model. Furthermore, the model was formulated for the combustion of pre-pyrolysed chars and not for coals, as used in this work. Further experimental data is required to apply to this model directly to the combustion results of this work.

4 Conclusion

A drop tube furnace has been used to measure the combustion behaviour of both MTE dewatered lignite and unprocessed lignite. Results show that the MTE process decreases the reactivity of the coal under conditions which approximate those in pulverised-fuel furnaces. This decrease in reactivity is apparently due to the reduction in pore volume that occurs during with the MTE process.

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6 References

- [1] Ashman, P.J. and Mullinger, P.J., Research issues in combustion and gasification of lignite. *Fuel*, 84, 2005, 1195-1205.
- [2] Favas, G. and Chaffee, A., MTE Processing of South Australian Lignites., CRC for Clean Power from Lignite report no. 01009. 2001
- [3] Qi, Y. and Chaffee, A. Effects of Processing Conditions on the Nature of Product Water from a Novel Coal Drying Process, *Eighth Annual Conference of the CRC for Clean Power from Lignite*. 2001.
- [4] Quyn, D.M., Hongwei, W., Hayashi, J., and Li, C.-Z., Volatilisation and catalytic effects of alkali and alkaline earth metallic species during the pyrolysis and gasification of Victorian brown coal. Part IV. Catalytic effects of NaCl and ion-exchangeable Na in coal on char reactivity. *Fuel*, 82, 2003, 587-593.
- [5] Hulston, J., Allardice, D., Huynh, D., Huynh, S., and Chaffee, A. Effect of MTE on the PhysicoChemical, Combustion and Storage Properties of Loy Yang Brown Coal. *11th Annual Conference of the Cooperative Research Centre for Clean Power form Lignite*. 2004.
- [6] Laurendeau, N.M., Heterogeneous kinetics of coal char gasification and combustion. *Progress in energy and combustion science*, 4, 1978, 221-270.
- [7] Knill, K.J., Maalman, T.F.J., and Morgan, M.E., Development of a combustion characterization technique for high volatile bituminous coals. International Flame Research Foundation report no 88/a/10. 1989
- [8] Marney, D., Investigation of the Pyrolysis of Two Low-Rank Coals at Elevated Pressures, *Ph.D. Thesis*, Swinburne University of Technology. 2002
- [9] Ballantyne, T.R.H., Mullinger, P.J., and Ashman, P.J., A new method for determining the conversion of low-ash coals using synthetic ash as a tracer. submitted to *Fuel*. 2005.
- [10] Clayton, S., Hoadley, A., Tiu, C., Huynh, S., and McIntosh, M. Development of a Laboratory Scale Continuous MTE Process. *Tenth Annual Conference of the Cooperative Research Centre for Clean Power from Lignite*. 2003.
- [11] Brockway, D.J. and Higgins, R.S., Brown Coal Sampling, Analysis and Composition, in *The Science of Victorian Brown Coal*, R.A. Durie, Editor. Butterworth Heinemann, 1991.
- [12] Australian Standard 2434.8-2002 Methods for the analysis and testing of lower rank coal and its chars Part 8: Lower rank coal - Determination of ash. *Standards Australia*. 2002
- [13] Hurt, R.H. Structure, Properties, and Reactivity of Solid Fuels. *Twenty-Seventh Symposium (International) on Combustion* 1998.
- [14] Gregg, S.J. and Sing, K.S.W., *Adsorption, Surface Area and Porosity*, Academic Press, 1967.
- [15] Ballantyne, T.R.H., Ashman, P.J., and Mullinger, P.J. Reactivity and structure of chars from dewatered lignite. *11th Annual Conference of the Cooperative Research Centre for Clean Power form Lignite*. 2004.