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Adiabatic testing procedures for determining the self-heating propensity of coal and sample ageing effects

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Abstract

An adiabatic testing procedure for determining the self-heating propensity of coal has been evaluated using New Zealand coals. Repeatability of the self-heating rate (R_{70}), obtained by the method, is less than ±5% of the average value for tests performed consecutively over 3–4 days. For longer time periods, even using samples under controlled storage, the self-heating rate value decreases noticeably due to pre-oxidation effects. This pre-oxidation effect can be described by the equation $R_{70}(t) = at^b$, where t is the time in days, b a constant dependent on storage method and particle size and a the R_{70} value of fresh coal. When stored samples are used for analysis, knowledge of this relationship can be used to extrapolate a fresh coal R_{70} value by doing repeated tests over time. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Several methods have been developed to study factors affecting the self-heating propensity of coal. The most common are as follows [1–4]:

1. Adiabatic heating — where a sample is placed in a reaction vessel, which is designed to minimise heat loss. The vessel is placed inside an adiabatic oven [1,3,5] or oil bath [6]. Oxygen or air is allowed to pass though the sample after it is preheated inside the oven. The oven temperature is controlled automatically to equal that of the sample, thus minimising heat losses. This method is used to directly measure self-heating rates.
2. Ignitability method (often referred to as the crossing point temperature method) — where a coal sample is placed inside an oven or controlled temperature bath and preheated air is allowed to pass though the sample while the oven temperature continues rising at a rate of 2°C/h [7]. When the temperature of the sample starts to become higher than that of the oven, this temperature is defined as the crossing point temperature.
3. Static isothermal method (also referred to as the oxygen adsorption method) — where the propensity of coal to self-heat is indicated by either the rate of oxygen consumption [8] or the rate of heat generation [9] at a constant starting temperature.
4. Dynamic method — where the desorption of gases resulting from oxidation of coal under flow conditions at a constant temperature are determined. Dry compressed air is preheated and allowed to pass through the coal sample. CO and

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CO_2 liberation are measured after reaching a steady state value [10].

All these methods have their advantages and disadvantages. The adiabatic test method of Humphreys [1] provides a full temperature history of the self-heating process at the various stages of generation. Unfortunately, few published data exist on this test method as Australian Coal Industry Research Laboratories (ACIRL) have mainly used it on a commercial service basis for mine operations in Australia.

Low rank New Zealand coals are known to be highly reactive in thermal processes [11–13] and have a history of being extremely prone to self-heating. At the end of the 1970s, a number of coal samples from the Kupakupa seam (Huntly East Mine) were sent to ACIRL in Australia to be tested for their propensity to self-heat [14]. The adiabatic test method was used and the average self-heating rate of the coal between 40 and 70°C was measured. This self-heating rate, which is known as R_{70}, is considered as a measure of the coal's propensity to self-heat [1]. A coal with an R_{70} value below 0.5°C/h is considered to be low risk, 0.5–0.8°C/h medium risk, while coals with an R_{70} value higher than 0.8°C/h are considered to be highly prone to self-heating [15]. The Kupakupa seam results ranged from 10.0 to 16.5°C/h and the seam was therefore considered to be extremely prone to self-heating [14].

Clemens et al. [9] studied the characteristics of a range of New Zealand coal samples varying from lignite to bituminous using isothermal differential thermal analysis (DTA) and temperature-programmed in situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS). Their results showed that subbituminous samples were more prone to self-heating than the lignites and bituminous samples tested. Vance et al. [5] and Vance [16] showed the effect of moisture content on New Zealand subbituminous coals under adiabatic conditions. Effectively, the rate of self-heating increased as moisture content decreased.

A more recent study [17] has been completed on the propensity of New Zealand coals to self-heat using the adiabatic method of Humphreys [1]. This paper presents the results obtained to evaluate the reliability of the adiabatic testing procedure for self-heating of coal, specifically looking at the repeatability between tests on the same sample, as this has not been documented before. Initially a repeat test was performed on a lignite sample, and it became apparent that ageing effects due to sample storage were having an impact on the self-heating results. To evaluate this more closely, a large coal lump was obtained from a fresh mining face and 11 replicate samples were obtained from this for analysis. One sample was used for proximate analysis and 10 for adiabatic self-heating tests. In addition repeat analyses were made of two other coals supplied from the Coal Research Ltd. sample bank.

2. Experimental

2.1. Preparation and storage of coal samples

A selection of coal samples was obtained from the Coal Research Limited (CRL) sample bank. These samples represented different coal ranks ranging from lignite to high volatile bituminous and were supplied as crushed (<5 mm) run-of-mine coal. These coals also covered two different annual surveys of New Zealand mines, namely 1996 and 1998. The samples were delivered to The University of Auckland on the 1st of May 1998 in airtight plastic bags containing approximately 250–300 g of coal. All samples were kept in the laboratory cold storage unit to reduce the effect of oxidation prior to testing.

A 2.5 kg lump of run of mine subbituminous coal was sampled from the working face of Huntly East Mine and sent directly to the university for testing. This sample was received on 28 May 1998. It was crushed to −5 mm the day after it was received. The crushed coal was split into 11×200 g samples and placed in airtight bags. One sample was sent to CRL for a proximate analysis, sulphur content and specific energy determination. The remaining samples were stored in the laboratory coal storage unit for the adiabatic testing. Each sample was crushed to −212 μm just prior to testing in the adiabatic oven, in order to minimise the effects of oxidation on fresh surfaces created by the grinding of the coal.

2.2. Adiabatic oven

The adiabatic oven consists of three parts: the oven; the reaction vessel; and the controller [18]. The oven

Fig. 1. Interior view of the adiabatic oven.

and the door are double skinned and contain kaowool and high density fibreglass for insulation. Heating is provided by a 1.6-kW element. A fan, located at the rear of the oven (Fig. 1), is used to ensure efficient temperature distribution inside the oven. A 16 m length of copper pipe is fitted on the door of the oven to ensure that the temperature of the inlet gas passing through it is the same as the oven when it reaches the sample (Fig. 2). The oven technical details are as follows:

Internal dimensions (mm)	400 wide×400 high× 400 deep
External dimensions (mm)	600 wide×1600 high× 1150 deep
Oven rating	1.6 kW, 230 V, single phase
Fan switch	0.375 kW, single phase
Insulation	75 mm high-density fibreglass

The reaction vessel is a 250 ml vacuum flask that is fitted with a teflon plug for sealing (Fig. 3). Two glass tubes pass through the teflon plug. The longer tube has a 3 mm inner diameter and is 150 mm long. It is used to pass gas, either oxygen or nitrogen into the vessel. It also has a pointed end to allow easy insertion though the ground coal sample. The smaller tube is used as an exhaust to collect effluent gas from above the coal sample and pass it out of the oven.

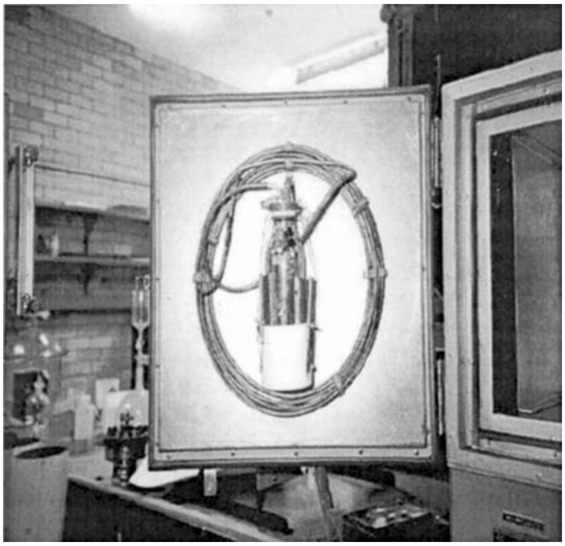

Fig. 2. Reaction vessel attachment to the oven door (note copper tubing wound in a coil).

The teflon plug is held in place by a brass clamp and two springs attached to clips supporting the reaction vessel and prevents gas leaks. Three thermocouples also pass through the teflon plug and are fastened at the end of the longer pipe to monitor and control the temperature of coal and oven. The vessel is fitted into the oven door via a support bracket, to allow easy access to the vessel and attachments (Fig. 2).

An MDC10 controller of SIGMA Controls Ltd. is used to govern the temperature of the oven and coal. It has three settings: manual; automatic; and remote. In the automatic mode the oven is set to a certain temperature and maintains this temperature until programmed to do otherwise. In the remote mode the controller monitors the oven and the coal temperature and keeps the oven temperature following that of the coal.

Six sets of type K thermocouples are used for monitoring temperature. Three of these are fitted inside the reaction vessel and connected to the data logger, controller and the limit switch, respectively. Two more thermocouples are fitted inside the oven and connected to the data logger and the controller to monitor and control the oven temperature. The last

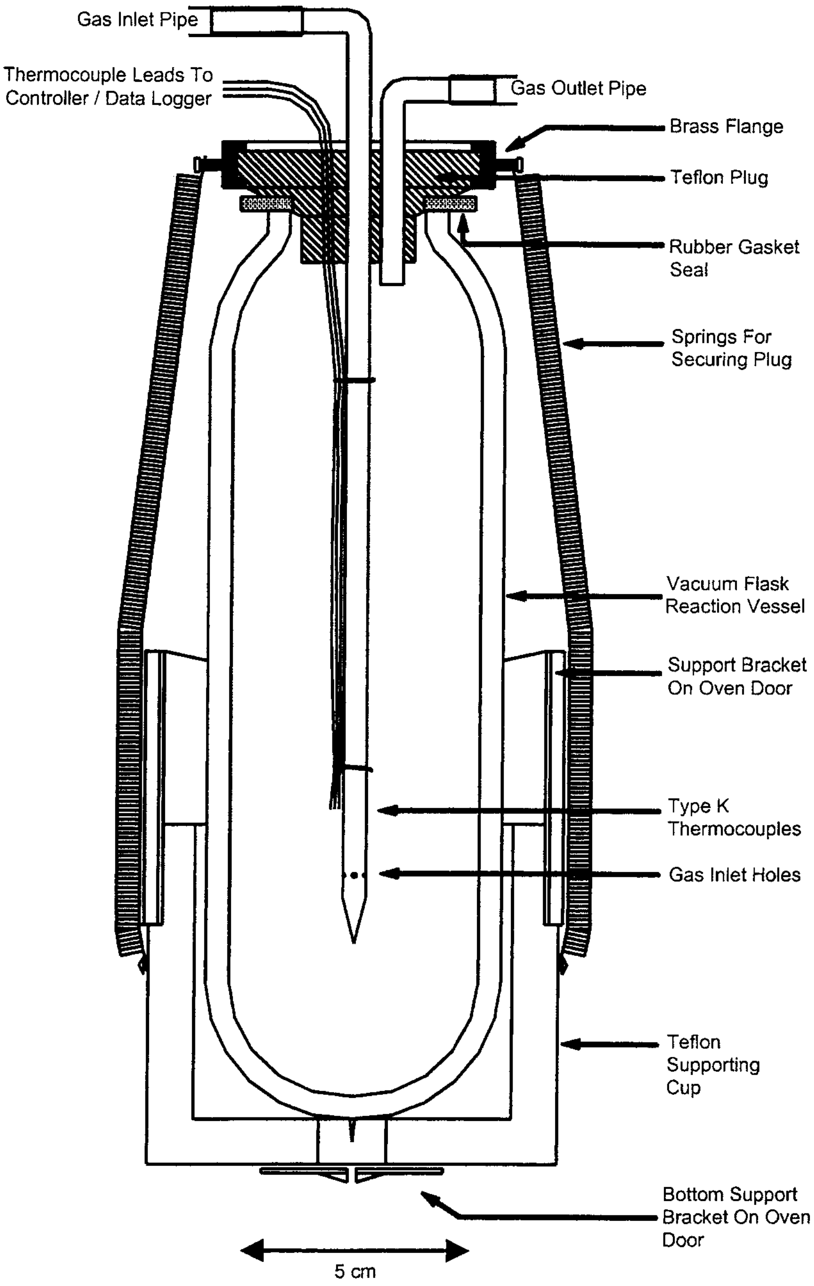

Fig. 3. Schematic diagram of the reaction vessel (modified from Humphreys et al. [3]).

one is positioned outside the oven to record the ambient air temperature.

Two solenoid valves are used to cut off gas supply when the limit switch shuts down the power or a power failure occurs. The limit switch is used to cut power off when the temperature inside the reaction vessel reaches a set limit. A data logger type DATA TAKER DT 100 is used to record coal and oven temperature and store data in a file using a PC and a special data logging program. The data file can later be analysed and the information graphed to obtain a self-heating curve. The graph can be monitored on the computer screen during the test, which allows continuous monitoring of the experiment. Once started the apparatus is designed to run unattended and the test will automatically terminate when the coal temperature reaches that fixed by the limit switch [18].

2.3. Testing procedure

All coal samples were dried under nitrogen at 105°C for approximately 15 h to ensure complete drying of the sample. This was performed using a drying tube arrangement described by Scott [18]. At the completion of drying the tube and coal were placed into the adiabatic oven at a temperature of 40°C and allowed to equilibrate. Once this was achieved the coal sample was transferred into the reaction vessel and left to stabilise at 40°C, with nitrogen passing through it. As soon as the sample temperature had stabilised, the oven was switched to remote monitoring mode and the gas selection switch turned to oxygen with a constant flow rate of 50 ml/min. The temperature change with time of the coal was recorded by a datalogging system for later analysis. The oven limit switch was set to cut-off the power to the oven and stops the oxygen flowing when the sample temperature reached 180°C. After the oven cooled down, the sample was removed from the reaction vessel and it was cleaned in preparation for the next test.

2.4. Self-heating curve analysis

All tests are started from 40°C, and the heating rate increases at a constant rate until approximately 70°C, after which there is a dramatic increase in the temperature rise rate leading to uncontrolled self-heating, until the temperature trip switch activates. Chamberlain [19], also noted that at a temperature of about 70°C a fundamental change takes place in the coal substance. He proposed that this appears to have the effect of increasing the available surface area and consequently accelerates greatly the rate of oxidation. Humphreys [1] uses the linear part of the adiabatic self-heating curve from 40 to 70°C to define what he calls the self-heating rate index R_{70}. This value is obtained as a simple ratio of the time taken to reach 70°C from a starting temperature of 40°C, and is expressed in units of °C/h. The results reported in this study have been obtained in this manner to enable direct comparison with the limited R_{70} values quoted in the literature.

3. Results

Analytical data for each coal tested are contained in Table 1. These coals range in rank from lignite A to subbituminous B. They are low in sulphur and low in ash. The corresponding self-heating rate index (R_{70}) values for each test are summarised in Table 2. Fig. 4 shows a plot of sample temperature and oven temperature against time for the initial test on the lignite sample (58/106). It clearly demonstrates the oven's ability to track the self-heating of the coal and minimise heat losses during the process. The minor deviation between 12 and 16 h is most likely an artefact of the oven temperature thermocouple, as it returns to the coal temperature beyond this time. The results of the repeat tests performed on the lignite sample (58/106) are shown in Fig. 5. Both tests show reasonable agreement with respect to the R_{70} values obtained, which were 2.26 and 1.89°C/h, respectively (Table 2). However, given the time difference between tests of

Table 1
Analytical data for New Zealand coal samples (air-dried basis, except for 62/148 on an as-received basis)

CRL Ref.	Mine	Moisture (%)	Ash (%)	Volatile matter (%)	Fixed carbon (%)	Calorific value (MJ/kg)	Sulphur (%)	ASTM rank
58/106	New Vale	25.0	3.1	39.5	32.4	19.42	0.40	Lignite A
61/814	Kopako	19.2	7.2	35.6	38.0	22.08	0.21	SubC
62/148	Huntly East	19.6	2.5	34.9	43.0	23.65	0.18	SubB
61/810	BBL	12.2	7.6	36.2	44.0	24.90	0.40	SubA

Table 2
Self-heating rate repeatability test results

CRL Ref.	Storage particle top size (mm)	Days elapsed since sampled	R_{70} (°C/h)
New Vale			
58/106	5	600	2.26
		609	1.89
Kopako			
61/814	5	20	17.23
		76	12.69
BBL			
61/810	5	21	14.91
		77	8.86
Huntly East			
HE1	5	2	19.53
HE2	0.212	3	15.20
HE3	5	6	16.55
HE4	5	7	15.12
HE5	5	8	15.67
HE6	5	12	13.88
HE7	5	13	13.47
HE8	5	14	14.39
HE9	5	16	14.80
HE10	5	33	13.47

nine days it could not be discounted that part of the reason for the lower value obtained in the second test could be attributed to oxidation during storage.

A selection of the self-heating curves for the Huntly East coal tested over a 33 day period are shown in Fig. 6. Not all the tests are plotted to avoid cluttering the figure. The R_{70} values for all of the tests range from 19.53 to 13.47°C/h (Table 2), which is consistent with the original ACIRL results of 16.5–10.0°C/h. The lower values obtained by ACIRL are most likely due to pre-oxidation effects during sample transport from New Zealand to Australia. It is interesting to note that these R_{70} values are an order of magnitude greater than the highest value recorded by Humphreys et al. [3], namely 1.45°C/h.

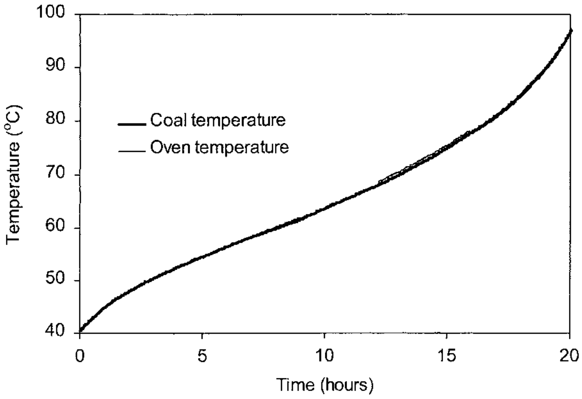

Fig. 4. Comparison of coal and oven temperatures for initial testing of New Vale sample 58/106.

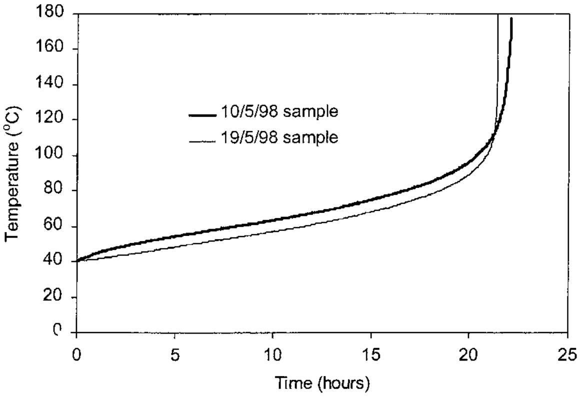

Fig. 5. Repeat self-heating profiles for New Vale sample 58/106.

When the R_{70} values for the repeat Huntly East samples are plotted against time since sampling (Fig. 7), it is clear that an oxidation effect due to ageing has taken place, even though the samples were stored under controlled conditions. This is also confirmed by the results for the repeat tests on samples from Kopako and BBL mines (Figs. 8 and 9), which show substantial drops in R_{70} of 4.54 and 6.05°C/h, respectively, (Table 2) over a 56 day period between tests. Test runs completed in consecutive days give a reasonable indication of the repeatability of the procedure. For example, samples HE3–HE5 (Table 2) have a mean R_{70} of 15.78 ± 0.72°C/h (4.56%), whereas samples HE6–HE9 (Table 2) tested the following week have a mean value of 14.14 ± 0.58°C/h (4.10%). A major part of this deviation is most likely the result of uneven particle size distribution from sample to sample.

The rapid drop in R_{70} value for sample HE2 (Fig. 7, Table 2) is primarily a combination of particle size and ageing effect. HE1 and HE2 were prepared to −212 μm at the same time. However, as HE1 was being tested adiabatically HE2 was stored at the reduced particle size instead of 5 mm, hence it tended to oxidise more readily prior to testing. These results show that at a stored particle size of −212 μm, R_{70}

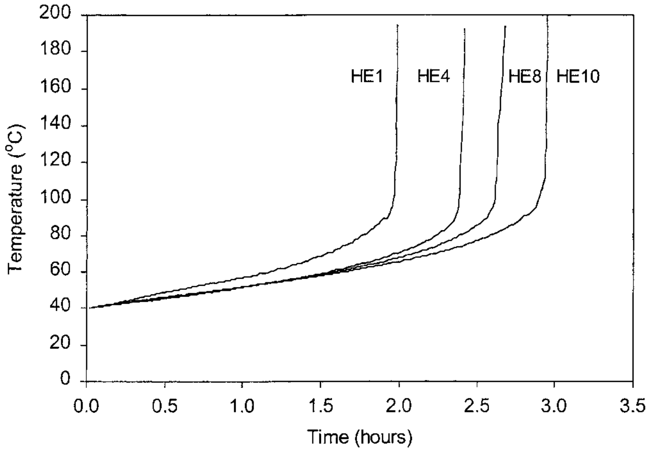

Fig. 6. Repeat self-heating profiles for Huntly East sample 62/148.

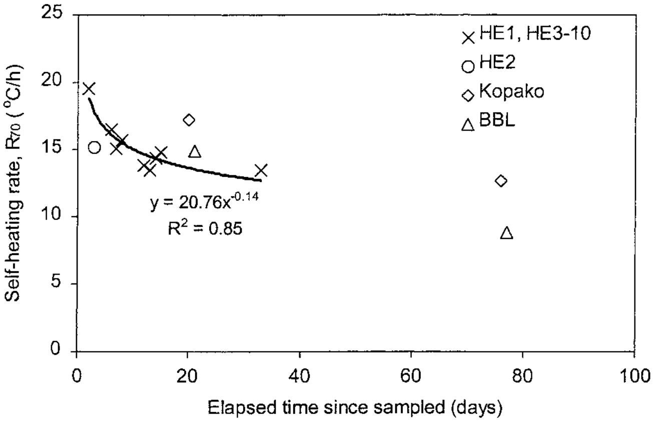

Fig. 7. Decreasing trend in self-heating rate with sample storage time.

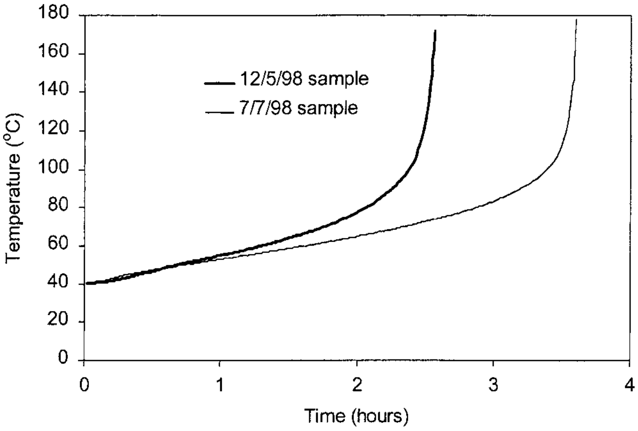

Fig. 8. Repeat self-heating profiles for Kopako sample 61/814.

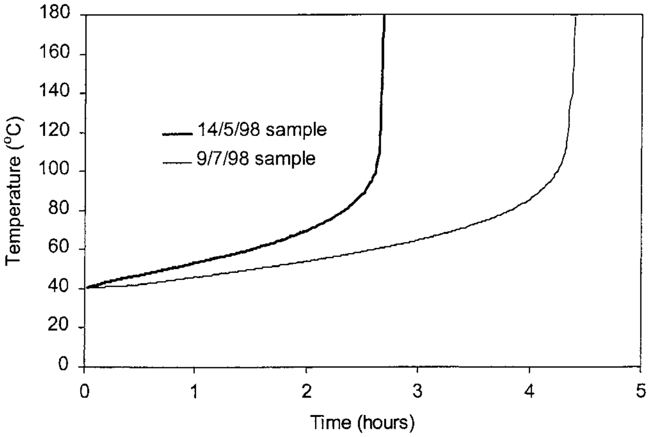

Fig. 9. Repeat self-heating profiles for BBL sample 61/810.

dropped by 4°C/h in 1 day, whereas the same drop at the larger particle size of −5 mm takes 5–6 days.

4. Discussion

The decay curve for the R_{70} values of the Huntly East repeat samples with elapsed time from sampling to testing (Fig. 7) appears to be of the form $R_{70}(t) = at^b$. The constant a is equal to the R_{70} value at day 1, or initial testing, for it is experimentally impossible to test at the time of sample recovery, i.e. $t = 0$. b is also a constant that appears to be dependent on stored sample particle size and storage method. Evidence of the particle size effect is shown by sample HE2 (Fig. 7), which, if all subsequent tests had been performed on stored (−212 μm) coal, indicates a much higher value for b at this smaller particle size. Conversely, a is independent of stored sample particle size and storage method, so that a fresh coal value for R_{70} could be obtained by doing more than one test on the same coal stored for some time at the same particle size.

Schmidt and Elder [8] proposed an equation of the form $r = kt^{b-1}$, which related the effect of time and extent of oxidation on rate of oxidation. The particle size used by Schmidt and Elder [8] to study the ageing effects was −6.4 mm, which is similar to this study. However, their test method was not adiabatic, but static isothermal and the rate values were determined from the rate of oxygen consumption with time. Nevertheless, it appears that both methods produce a similar ageing effect result, in other words they are recording the same effect. The value for b of −0.14 obtained from the line of best fit (Fig. 7) is lower than the average value of −0.2 obtained by Schmidt and Elder [8]. This is most likely due to the storage method used in this study, which retards the oxidation effect to some extent or alternatively the rank of coal may be having some influence on this parameter. The Kopako and BBL coals indicate slightly different b parameters as shown in Fig. 7.

5. Conclusions

Adiabatic testing procedures for determining self-heating rates of coal provide valuable information on the entire process of spontaneous combustion. The self-heating rate parameter (R_{70}) defined by Humphreys [1] has an acceptable repeatability of less than ±5% of the average value if tests are conducted consecutively over a short time period (<3–4 days). Given the extremely high reactivity of the coal used in these tests, this is an excellent result and is attributable to the ability of the oven to consistently simulate adiabatic conditions over sustained periods. The minor deviations recorded could easily be accounted for by slight differences in particle size distribution of the prepared samples.

Testing of stored replicate samples after 1 week show a significant decrease in R_{70} which is due to pre-oxidation of the coal. This ageing effect is even more noticeable as storage time increases, and appears to follow a power function of the form, $R_{70}(t) = at^b$, where t is the time in days since sampling the coal, a is the fresh coal value of R_{70} and b a constant dependent on storage method and particle size. Consequently, if fresh samples are not obtained or are unavailable for testing, repeat testing after at least 1 week (preferably 2 weeks) will provide data necessary to extrapolate back to a fresh coal result.

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References

[1] D.R. Humphreys, A Study of the Propensity of Queensland Coals to Spontaneous Combustion, ME Thesis, The University of Queensland, Brisbane, Australia, 1979, 108 pp.
[2] D. Cliff, D. Rowlands, J. Sleeman, Spontaneous Combustion in Australian Underground Coal Mines, in: C. Bofinger (Ed.), Safety in Mines Testing and Research Station, Brisbane, Australia, 1996, 165 pp.
[3] D. Humphreys, D. Rowlands, J.F. Cudmore, Proc. Ignitions, Explosions and Fires in Coal Mines Symposium, The AusIMM Illawarra Branch, 1981, pp. 5-1-5-19.
[4] A.G. Kim, Laboratory Studies on Spontaneous Heating of Coal: A Summary of Information in the Literature, US Bureau of Mines Information Circular IC 8756, 1977, 13 pp.
[5] W.E. Vance, X.D. Chen, S.C. Scott, Combustion Flame 106 (1996) 261.
[6] T.X. Ren, J.S. Edwards, D. Clarke, Fuel 78 (1999) 1611.
[7] S.C. Banerjee, Spontaneous Combustion of Coal and Mine Fires, Balkema, Rotterdam, 1985, 168 pp.
[8] L.D. Schmidt, J.L. Elder, Ind. Eng. Chem. 32 (2) (1940) 249.
[9] A.H. Clemens, T.W. Matheson, D.E. Rogers, Fuel 70 (1991) 215.
[10] J.M. Kuchta, V.R. Rowe, D.S. Burgess, Spontaneous Combustion Susceptibility of US Coals, US Bureau of Mines Report of Investigations RI 8474, 1980, 37 pp.
[11] T.A. Daly, C.F. Budge, Fuel 53 (1974) 8.
[12] D.J. Watson, V.R. Gray, N.Z. J. Sci. 23 (1980) 313.
[13] B.B. Beamish, K.J. Shaw, K.A. Rodgers, J. Newman, Fuel Proc. Tech. 53 (1998) 243.
[14] P.F. Miles, The Prevention and Detection of Spontaneous Combustion in Longwall Waste Areas with Respect to the Huntly East Mine, Undergraduate Project Report, The University of Otago, New Zealand, 1986.
[15] R. Moreby, Proc. 6th International Mine Ventilation Congress, The Society of Mining, Metallurgy and Exploration Inc., Littleton, USA, 1997, pp. 39–45.
[16] W.E. Vance, Proc. 5th New Zealand Coal Conference, Coal Research Limited, Wellington, New Zealand, 1993, pp. 94–100.
[17] M.A. Barakat, Spontaneous Combustion Investigation of Some New Zealand Coals Using an Adiabatic Oven, Postgraduate Project Report, Department of Civil and Resource Engineering, The University of Auckland, New Zealand, 1998.
[18] S.C. Scott, Drying and Moisture Effects on the Self Heating Potential of Huntly East Subbituminous Coal, Masters Thesis, Department of Mining Engineering, The University of Auckland, New Zealand, 1994.
[19] E.A.C Chamberlain, Colliery Guardian March (1974) 79.