

A SPONTANEOUS COMBUSTION LIABILITY INDEX

KENDİLİĞİNDEN YANMA YATKINLIĞI İNDEKSİ/

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ÖZET

Kömürün kendiliğinden yanması Güney Afrika kömür madenlerindeki yeraltı yangınlarının en önemli nedenidir. Johannesburg'daki Witwatersrand Üniversitesi Maden Mühendisliği Bölümü kömürün kendiliğinden yanmaya yatkınlığının tayinini belirleyen bir indeks geliştirmiştir. Bu indeks WITS-EHAC İndeksi olarak adlandırılmış olup kesişim noktası sıcaklık deneylerine ve diferansiyel termal analiz deneylerine (tutuşma sıcaklığı deneyleri) dayanmaktadır. Bu indeksin son beş yıldır başarılı bir şekilde kullanılmasına karşın ampirik bir değer olması dezavantaj teşkil etmektedir.

ABSTRACT

Spontaneous combustion is the major cause of underground fires in South African coal mines. The Department of Mining Engineering of the University of the Witwatersrand, Johannesburg, has developed a spontaneous combustion liability index to identify coals that have a tendency to self-heat. This index has been called the WITS-EHAC liability index and is based on crossing-point temperature tests and differential thermal analysis (ignition temperature tests). While this index has been used successfully for the past five years, it is suffered from the disadvantage of being empirical in nature.

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INTRODUCTION

The spontaneous combustion of coal is responsible for the majority of underground fires in South African collieries (Fig. 1). Incidents of spontaneous combustion are also prevalent in opencast collieries and in stockpiles, discard dumps, trains, trucks and ships.

The magnitude of the problem prompted industry to fund a research programme at the Department of Mining Engineering of the University of the Witwatersrand, to investigate a means of predicting the liability of a coal to self-heat. An ignition temperature apparatus was designed and crossing-point temperature tests and differential thermal analysis (DTA) tests were conducted on over one hundred different coal samples.

This programme of research led to the adoption of a liability index for South African coals. The WITS-EHAC liability index was proposed in 1987 and was based on characteristics of a given coal, as identified on a DTA thermogram². The thermogram (Fig. 2) is obtained by heating a coal sample and a sample of a thermally inert reference material in identical sample holders in an oil bath. The thermogram is obtained by plotting the temperature of the coal minus that of the inert material against the temperature of the inert material. The WITS-EHAC liability index is given by:

$$\text{WITS-EHAC INDEX} = \left(\frac{\text{STAGE II SLOPE}}{\text{CROSSING-POINT TEMPERATURE}} \right) * 500.$$

WITS-EHAC liability index values lie between three and six, with a higher value indicating a coal more likely to self-heat.

The liability index proved to be successful, and was validated by an investigation of apparent anomalies³, and a research project in which the liability of a seam to self-heat was contoured⁴. The latter project identified high risk areas at a particular colliery, which agreed with the history of incidents at that colliery⁵. The index, however, suffers from the drawback of not being mathematically consistent, and the ignition temperature apparatus requires constant supervision and continued maintenance.

THE CALORIMETER

In order to overcome the problems associated with the ignition temperature apparatus, and to simplify the test procedure, it was decided to construct an

alternate test apparatus. The experimental technique that was chosen for this phase of the research programme was adiabatic calorimetry. Descriptions of the apparatus have been published elsewhere^{6,7}.

The calorimeter was designed to test the self-heating of coal using one of the following three test options :

- * incubation test,
- * minimum self-heating temperature test, and
- * crossing-point temperature test.

The incubation test can be regarded as the classic adiabatic test. Using the calorimeter in this mode entails heating the sample and the apparatus to a predetermined starting temperature and, after a chosen stabilizing period, percolating air through the sample. The oven and input gas are then heated to follow the temperature of the sample while oxidization of the sample takes place. Factors such as the initial rate of heating of the coal sample and the total temperature rise have been used to predict the propensity of a coal to self-heat.

The minimum self-heating temperature test is a variation of the incubation test. In this experiment the incubation test is started at a specific temperature and if no self-heating takes place the apparatus is heated to a higher starting temperature. The incubation test is then repeated. This continues until the coal self-heats. The starting temperature at which this occurs is known as the minimum self-heating temperature of the sample. A coal with a low minimum self-heating temperature is more likely to self-heat than a coal with a higher minimum self-heating temperature.

The third test option that the calorimeter was designed to perform is the crossing-point temperature test. This experiment was intended to simulate the tests conducted using the ignition temperature apparatus, and entails heating the coal sample at a linear rate and observing its thermal behaviour with respect to some datum temperature.

THE SELECTED EXPERIMENTAL PROCEDURE

The crossing-point temperature option of the calorimeter was selected in order to simulate the ignition temperature tests. It was decided to heat the

sample at the same rate as was used during the ignition temperature tests but it was found that this heating rate could be achieved by the oven, **but** not the sample. Once the calorimeter oven reached a temperature of 200°C the experiment was shut down by the computer in order to protect the thermometers. The sample temperature lagged far behind that of the oven and the coal temperatures attained in the ignition temperature tests could not be reached in this experiment.

In order to overcome these difficulties it was decided to heat the sample with a band-heater controlled by a variac. The variac was switched on at the beginning of the experiment by means of a relay connected in parallel with a relay controlling the airflow solenoid. The variac was set so as to result in the heating of the inert material in the sample holder at the required rate. The incubation mode of the calorimeter was used and the oven matched the rise in temperature of the sample. The experiment was then repeated using a coal sample. The data pertaining to the heating of the inert material was used as a datum, against which the heating characteristics of the coal could be examined.

This experimental technique is known as the rising temperature technique (or the ramping temperature technique) and is similar to the differential thermal analysis tests described earlier, because both tests measure the heating rate of the coal with respect to the rate at which an inert substance is heated in identical circumstances.

RESULTS

Spontaneous combustion is primarily a first order oxidation reaction where heat loss can be assumed to be solely due to conduction. The reaction is temperature dependant and can be expressed as follows :

$$w C_p \frac{dT}{dt} = w Q Z e^{-E/RT} + \text{conduction term}$$

where :

- w = density
- C_p = specific heat of coal at constant pressure
- T = temperature (Kelvin)
- t = time
- Q = heat of reaction
- e = exponential (natural log)
- E = activation energy
- R = molar gas constant

This is known as the Arrhenius Equation and, under near adiabatic conditions where the conductivity term is negligible, it can be simplified to :

$$C_p \frac{dT}{dt} = QZ e^{-E/RT}$$

or

$$\frac{dT}{dt} = A e^{-E/RT}$$

where :

$$A = QZ/C_p = \text{the rate coefficient}$$

This can be simplified further, as follows :

$$\ln \frac{dT}{dt} = -E/RT + \ln A$$

which is of the form :

$$y = m x + c.$$

The log of the rate of temperature rise ($\ln \frac{dT}{dt}$) can be plotted against the inverse of the absolute temperature of the coal (V_T), as in Figure 3. The slope of the straight line during the latter part of the experiment is equal to $-E/R$. The activation energy (E) of the coal can be calculated from this slope. The vertical intercept is the rate coefficient (also known as the pre-exponential coefficient or frequency factor) for the reaction, which is believed to be greater for coals more likely to self-heat⁸.

An examination of the kinetic plot (Fig. 3) shows that the temperature of the coal increases at a reasonably constant rate at the start of the experiment (the horizontal section of the curve) and then increases at an increasing rate until the experiment is terminated. Besides the characteristics already mentioned, another characteristic of the coal curve is immediately apparent. This is the temperature of the coal at which the coal curve cuts the $\log \frac{dT}{dt}=0$ line.

In order to investigate the characteristics of the Arrhenius plot the differential thermal analysis thermogram (from the ignition temperature tests) can be modified to show the inverse absolute temperature of the coal on the horizontal axis. The modified D.T.A. thermogram for Sample No. 104 is shown in Figure 4.

After comparing Figures 3 and 4 it appears that the point at which the coal curve cuts the $\ln \frac{dT}{dt}=0$ line is the crossing-point for the coal.

The crossing-point temperature of the coal shown in Figure 3 is seen to be 154 °C ($^{000}/T=2.34$). This is the same as the value found for the crossing-point temperature in Figure 4. It was found that the first ten crossing-point temperatures determined using the calorimeter were in good agreement with those determined using the ignition temperature apparatus. A linear regression performed on the crossing-point temperatures of the first ten coals tested yielded a correlation coefficient (R) of 96 % and a coefficient of determination (R^2) of 92 %.

The eleventh sample tested in the calorimeter proved to be a major disappointment in that a crossing-point temperature of 140 °C was observed when a temperature of 128,2 °C was expected. It was decided to re-examine all the coal curves in the light of this new development. It was discovered that the sample in question had a different initial rate of heating (the horizontal portion of the Arrhenius curve) from that of the other coals, which was as a direct result of difficulties in combining the coal and inert data files. Once the coal curve was moved to a position where the initial rate of heating matched that of the average of the other samples tested, a crossing-point of 129°C could be read off the Arrhenius curve. It was, however, clear that the difficulties in finding the crossing-point temperature of a sample deserved further attention.

After further comparing Figures 3 and 4, it seems that there should be some relationship between the slope of the coal curve during the latter part of the experiment and the stage II slope of the D.T.A. thermogram. The slopes of coals tested in the calorimeter were compared with the stage II slopes of the same samples and a linear regression showed a correlation coefficient (R) of 94 % and a coefficient of determination (R^2) of 89 %.

Since the calculation of the WITS-EHAC index value of a coal sample requires only the crossing-point temperature and the stage II slope, it can be calculated from the calorimeter data that has been shown to correlate well with ignition temperature data. The WITS-EHAC index values of samples tested during the initial stages of the research programme are compared in Figure 5.

A linear regression performed on the WITS-EHAC index values listed in Figure 5 shows a correlation coefficient (R) of 88 % and a coefficient of determination (R^2) of 77 %. The good correlation between the index values obtained by using different apparatus suggests that the calorimeter can be used to determine the relative liability of coals to self-heat.

It has been shown that the WITS-EHAC index can be calculated from the crossing-point temperature (XPT) and the slope of the coal curve as determined with the calorimeter. It has also been pointed out that the slope is equal to E/R . The index can therefore be rewritten as :

$$\text{WITS-EHAC} = E_{/XPT} \times \text{constant}$$

The determination of the crossing-point temperature has been identified in this paper as being a source of potential error when using the calorimeter. An examination of the Arrhenius plot, however, shows a point which is more easily identified than the crossing-point. This is the point at which the almost horizontal curve begins to slope sharply upwards. From an examination of the D.T.A. curve for this coal it is found that this point occurs between the point of maximum negative temperature differential ($^{1000}/T=2,9$) and the crossing-point. It is at this point that the rate of temperature rise exceeds that of an inert sample at the same temperature. This point is known as the threshold point⁷.

The fact that a point has been identified which could be more relevant to the onset of the exothermic reaction, and is easier to determine than the crossing-point temperature, suggests that the crossing-point temperature in the WITS-EHAC index can be replaced by the threshold temperature (Ttemp) to yield the equation :

$$\text{WITS-EHAC} = E_{/Ttemp} \times \text{constant}$$

but E corresponds to $\log(A) \times Ttemp$, therefore :

$$\text{WITS-EHAC} \sim \log(A)$$

A linear correlation of the liability index values from the calorimeter against those from the ignition temperature apparatus yielded a correlation coefficient of 92,0 % for the initial sample set. These initial findings have been confirmed by testing an additional 25 coal samples⁵. The samples were tested in both the ignition temperature apparatus and the calorimeter, and the WITS-EHAC index values of the two different experiments yielded a linear correlation coefficient of 87% (Fig 6).

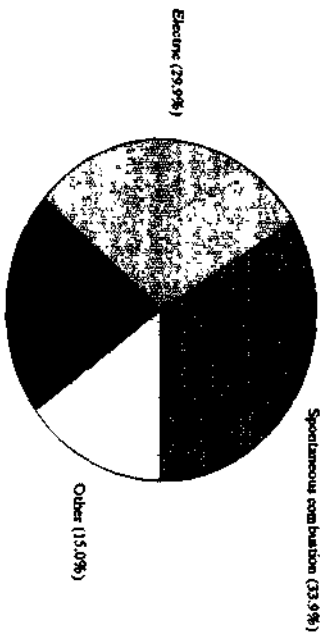


Figure 1. Spontaneous combustion, the major cause of the 254 fires in colleries reported during the period 1970 to 1990(1)

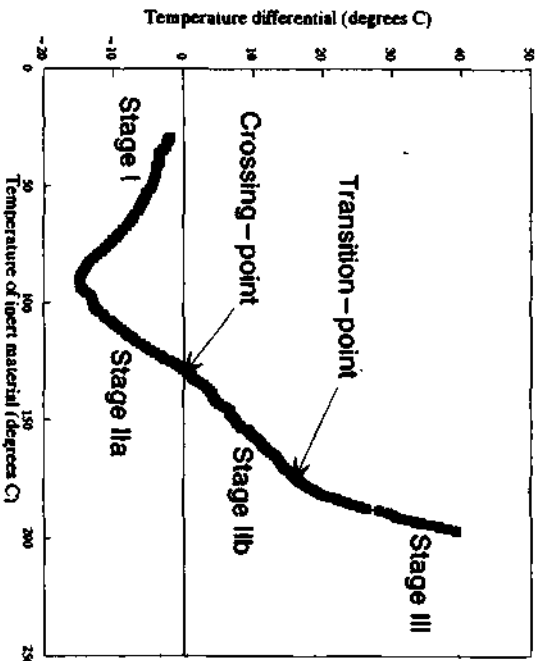


Figure 2. Typical differential analysis thermogram.

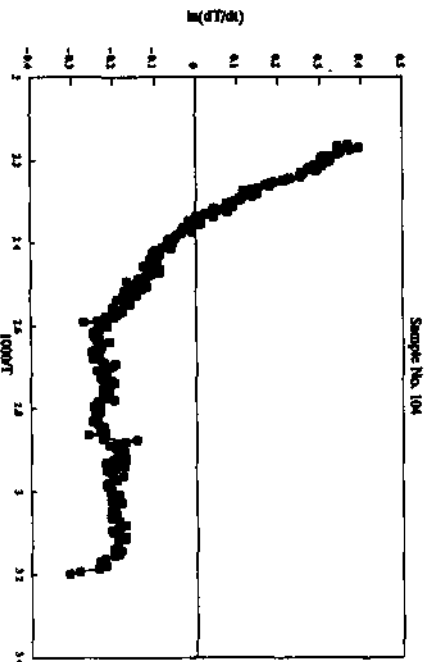


Figure 3. Typical Arrhenius plot.

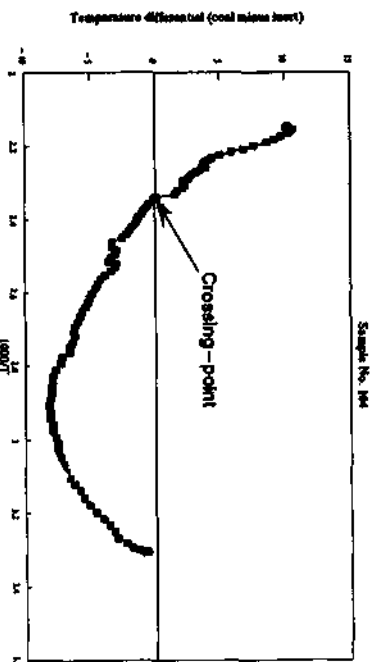


Figure 4. Modified D.T.A. curve obtained using ignition temperature tests.

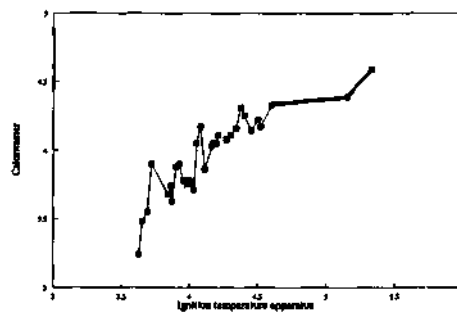
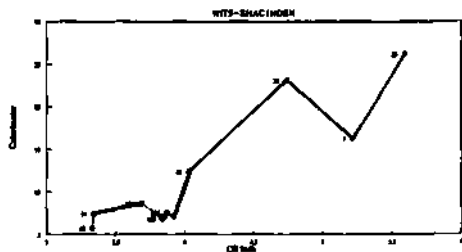


Figure 5. Comparison of HITHS- Figure 6. The relationship between EHAC index values obtained using the WITS-KHAC index values determined the ignition temperature apparatus in the two sets of equipment, and the calorimeter.

CONCLUSION

An adiabatic calorimeter has been designed and constructed to enable the self_heating liability of coal to be investigated using several testing techniques. An experimental technique was developed in which the calorimeter could be used to simulate the ignition temperature tests conducted earlier in this research programme. The results of tests conducted using the calorimeter were shown to be consistent with those of the ignition temperature tests and it was shown that the WITS-EHAC index is, in fact, directly proportional to the frequency factor.

The relationship between the WITS-EHAC index and the frequency factor is particularly satisfying in that the frequency factor ($\log A$) has often been reported in the literature as being a characteristic that is indicative of the liability of a coal to self-heat⁸.

The calorimeter was designed to replace the ignition temperature apparatus and simplify the test procedure. This has been achieved, with the

particular advantage of the adiabatic test being of similar duration to the ignition temperature test. The adiabatic test also has the advantage of requiring very little preparation time and is fully automatic, requiring no supervision at all.

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