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Assessment of spontaneous heating susceptibility of coal seams by experimental techniques - a comparative study

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ABSTRACT: Mine fires due to spontaneous heating of coal is a common, but major problem in the coal mining industry. They not only endanger the lives of men in mines, but also cause considerable economic losses to the organization. Since all coals are not susceptible to spontaneous heating to the same extent, it is essential to assess their degree of proneness in order to plan advance precautionary measures. This paper describes the determination of spontaneous heating susceptibility of coal samples by three different experimental methods viz., crossing point temperature (CPT), differential thermal analysis (DTA) and differential scanning calorimetry (DSC). The above tests were carried on 30 coal samples collected from different Indian Coalfields and a comparative analysis has been presented.

1 INTRODUCTION

Coal mine fire is a major concern for the Indian coal mining industry. Studies carried out by different researchers on genesis of these fires reveal that the main cause is the spontaneous heating of coal. Apart from causing accidents, resulting in loss of lives and property, they also cause considerable economic losses to the organization. Development of spontaneous heating in any section/part of a mine creates problems for the workings of other areas. Some of the other problems associated with spontaneous heating are diminution of heating values and coking properties, and in some cases serious environmental pollution. A careful analysis into occurrence of these fires reveals that most of these fires could have been averted if suitable preventive measures have been taken. It is therefore essential to first assess their degree of proneness in order to plan advance precautionary measures against the occurrence of these fires.

In the past, a number of researchers have attempted to assess the spontaneous heating tendency of coals by carrying out different experiments in the laboratory and different coal producing countries of the world follow different methods for this purpose. These are crossing point temperature (CPT) in India, Russian U-index in Russia, Olpinski index in Poland, adiabatic calorimetry in USA etc. Some of the other methods attempted by different researchers include differential thermal analysis (Banerjee and Chakravarty 1967, Gouws and Wade 1989), wet oxidation method (Singh et al. 1985, Tarafdar and Guha1989, Panigrahi et al. 1996) etc. However, there is no unanimity among researchers for the adoption of a particular method for the assessment of spontaneous heating susceptibility of coals. In fact some of the researchers have suggested that a number of methods may be attempted to determine fairly accurately the degree of proneness of a particular coal to spontaneous heating.

In the present work, an extensive study has been carried out by collecting a large number of coal samples from different Indian Coalfields and by determining their spontaneous heating susceptibility using three different experimental techniques, viz. crossing point temperature (CPT), differential thermal analysis (DTA) and differential scanning calorimetry(DSC). The intrinsic properties of these coal samples have been determined by proximate and ultimate analysis and a comparative study has been presented.

2 COAL SAMPLES COLLECTED FOR THE STUDY

Coal samples were collected from different Indian Coalfields belonging to both fiery and non-fiery seams of varying ranks spread over 7 different mining companies. Table 1 presents the details of samples collected, i.e. name of the seam, colliery and the subsidiary company. The coal samples were collected from different seams following channel sampling procedure(Peter 1978). It may be noted that some of the coal samples collected like Jhingurda seam (sample no. 30), Samla seam (sample no. 24), Chirimiri-III seam (sample no. 28) Kampti-IVB seam (sample no. 29) are highly susceptible coal seams where as seam-0 of Bastacola colliery (sample no. 21) takes a very long time to catch fire in the mine as well as in coal stacks and is known to be the least susceptible to spontaneous heating.

Table 1. Details of the coal samples collected for the study

Sample no.	Coal Seam	Colliery
1	Seam - IX	Hingula
2	Seam - VII	Lingaraj
3	Seam - V	Lingraj
4	Seam - IV	Lingaraj
5	Seam - III	Ananta
6	Seam - III	Bharatpur
7	Seam - II	Ananta
8	Seam - II	Bharatpur
9	Seam - II	Jagannath
10	Seam - II	Lingaraj
11	Seam - I	Deulbera
12	Seam - I	Nandira
13	Lajkura - IV	Orient-3
14	Lajkura - III	Orient-3
15	Lajkura - II	Orient-2
16	Lajkura - I	Orient-3
17	Rampur - IV	Hingir-Rampur
18	Rampur - B	Hirakhand Bundia
19	Rampur - A	Hirakhand Bundia
20	Ib	Belpahar
21	Seam - 0	Bastacola
22	Mahuda bottom	Murlidih
23	Laikdih	West Victoria
24	Samla	Samla
25	Burra Dhemo	Methani
26	Dakra	Dakra Bukbuka
27	Hatidhari	Saunda
28	Seam - III	Chirimiri
29	Seam - IVB	Kampti
30	Jhingurda	Jhingurda

Sample nos. 1-20 : Mahanadi Coalfields Ltd.,
Sample nos. 21-23: Bharat Coking Coals Ltd.
Sample nos. 24-25 : Eastern Coalfields Ltd.,
Sample nos. 26-27 : Central Coalfields Ltd.
Sample no. 28 : South Eastern Coalfields Ltd.,
Sample no. 29 : Western Coalfields Ltd.
Sample no. 30 : Northern Coalfields Ltd.,

3 DETERMINATION OF INTRINSIC PROPERTIES OF COAL SAMPLES

In order to make a comparative study, the intrinsic properties of these coal samples were determined by proximate and ultimate analysis.

3.1 Proximate Analysis

The moisture(M), Volatile matter(VM) and ash content(A) of coal samples were determined by proximate analysis following the method specified by IS(Indian Standard) 1350, Part - I, and the results are presented in Table 2.

3.2 *Ultimate Analysis*

The carbon (C) and hydrogen (H) contents of coal samples were determined using the Central Fuel Research Institute (CFRI) method as per IS: 1350, Part IV/Sec (1974). The nitrogen content of coal was determined by Kjeldahl method as per IS: 1350 (1975), the total sulphur present was determined as per IS: 1350 (1969). Oxygen content in coal was computed by subtracting the sum of percentage of hydrogen, carbon, nitrogen, sulphur, ash and moisture from 100. Nitrogen has no effect on the spontaneous heating tendency of coal. The total sulphur content of all the coal samples are less than 1.20% and a part of this will be pyretic sulphur. Munzer (1975) and Banerjee (2000) have inferred that pyrite might have an appreciable effect, if its concentration in finely dispersed form exceeds 5 to 10%. If the pyretic sulphur is less than 5%, its effect would not be of much importance. Therefore, the results of carbon (C), hydrogen (H) and oxygen (O) have been presented in Table 2.

Table 2. Intrinsic properties of coal samples

4 DETERMINATION OF SPONTANEOUS HEATING SUSCEPTIBILITY OF COAL SAMPLES

Susceptibility indices of coal samples were determined by three different experimental methods, viz. crossing point temperature (CPT), differential thermal analysis (DTA) and differential scanning calorimetry.

4.1 *Crossing Point Temperature(CPT) of Coal Samples*

The crossing point temperature (CPT) of the coal samples were determined following the procedure and experimental set up described by Panigrahi et al. (1999). Glycerine bath was used as the heating medium. The rate of rise of temperature was

S. No.	Proximate analysis			Ultimate Analysis		
	M%	VM%	A%	C%	H%	O%
1	11.13	25.19	38.46	66.38	8.77	21.22
2	14.29	31.25	16.27	72.62	6.63	17.44
3	7.25	23.9	40.50	71.48	8.71	17.23
4	9.59	28.93	7.68	80.61	5.69	11.23
5	8.97	29.49	22.88	75.78	7.00	14.50
6	10.02	26.06	31.57	71.91	7.76	17.67
7	14.5	31.97	12.35	73.85	6.33	17.83
8	11.32	29.81	21.80	73.67	6.98	16.93
9	6.67	27.9	36.40	77.72	8.43	15.74
10	6.65	30.31	22.53	77.94	6.96	12.88
11	9.12	33.51	8.37	80.20	6.51	10.22
12	8.72	25.14	13.75	80.83	5.86	10.47
13	6.78	26.82	33.09	74.79	7.87	14.68
14	6.94	28.48	28.22	76.11	7.42	14.27
15	11.34	23.48	30.56	72.01	7.44	18.24
16	6.74	26.14	34.89	69.11	8.05	20.04
17	7.64	22.13	37.02	73.85	8.00	15.67
18	5.00	24.01	32.40	78.47	7.46	11.55
19	4.88	24.17	38.87	75.61	8.32	12.91
20	6.31	28.39	32.42	75.76	7.92	13.69
21	1.00	17.36	15.73	88.7	4.71	3.48
22	1.90	33.08	16.00	87.86	4.12	5.24
23	0.60	22.32	10.73	86.75	5.33	5.58
24	8.43	24.43	9.60	80.85	5.73	10.48
25	1.80	36.13	14.27	80.6	5.99	10.36
26	10.00	32.27	18.00	77.71	5.26	14.83
27	10.52	29.47	11.91	80.44	5.01	11.28
28	7.67	29.83	18.88	79.80	4.97	13.31
29	14.39	29.31	12.76	79.40	3.73	14.75
30	9.68	29.80	18.12	76.83	6.58	13.93

maintained at 1^0C per minute and oxygen was supplied at 80cc per minute, while the glycerine bath was continuously stirred with the help of an air tube and stirrer. The CPT of all the coal samples is presented in Table 3.

4.2 *Differential Thermal Analysis (DTA)*

Differential thermal analysis was carried out by a Differential Thermal Analyser having proportional integral derivative control. The standardized parameters suggested by Banerjee and Chakravorty (1967) were followed while performing the experiments. DTA thermograms were obtained until 350^0C at a heating rate of 5^0C per minute. It has been shown by Banerjee and Chakravorty (1967) that a thermogram of coal can be divided into three segments or stages. These stages have also been identified in the thermograms generated from the results of the experiments as indicated in Figure 1.

In the initial stage of heating (stage I), the endothermic reaction predominates, probably due to the release of inherent moisture in coal. In the second stage (stage II), the exothermic reaction becomes significant, but the rate of heat release is not steady all through, as it changes with temperature. A steep rise in heat evolution is observed in the third stage (stage III).

The rate of temperature rise in stage II is cited by different researchers, viz. Banerjee and Chakravorty (1967), Gouws and Wade (1989); as being less for coals with less susceptibility to spontaneous heating. The exothermicity in stage III is not regarded as a reliable indicator of the self heating risk, because it may be equally high for low rank coals. However, the temperature of transition or characteristic temperature or onset temperature is considered to be significant. It is considered that lower is this temperature, more susceptible is the coal towards spontaneous heating. Therefore, all the thermograms were analysed for the following details.

(i) Characteristics temperature (T_c)
(ii) Average slope of stage IIA
(iii) Average slope of stage IIB
(iv) Overall slope of stage II

4.2.1 *Onset temperature or characteristic temperature*

The onset temperature or characteristic temperature was determined by the following procedure:

- A tangent was drawn at the inflexion point of the endothermic region and another tangent was drawn at the rising portion of the curve of stage III.
- The intersection between the two tangents gives the characteristic temperature.

Determination of characteristic temperature or onset temperature for sample number 29 has been demonstrated in Figure 1.

4.2.2 Slopes of stage II of the thermogram

In the thermograms obtained from the experiments, a linear stage II exothermicity was not observed. In view of the disjointed nature of stage II slopes it was further divided into two different regions, viz. stage IIA and stage IIB. The following three parameters of stage II were determined for further analysis.

a) average slope of stage IIA.
b) average slope of stage IIB.
c) overall slope of stage II.

The determination of the above three slopes for sample no. 29 has also been demonstrated in Figure 2 as an example.

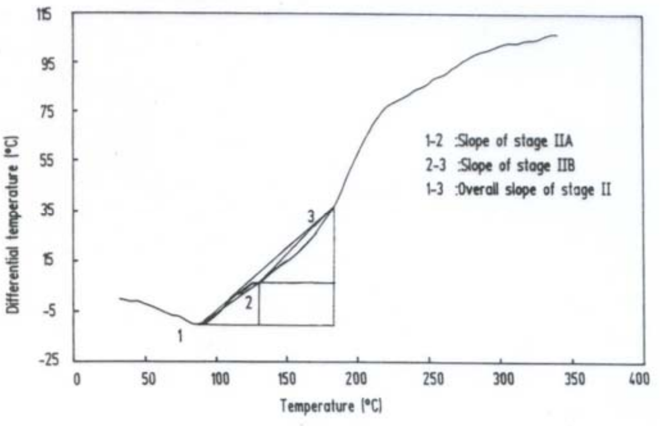

Figure 2. Slopes of stage II, IIA and IIB presented on thermogram of DTA for sample no. 29

4.3 *Differential Scanning Calorimetry*

Differential Scanning Calorimetry (DSC) is a technique in which the difference in energy inputs into a substance and a reference material is measured as a function of temperature while the substance and reference materials are subjected to a controlled temperature program. In this technique the ordinate value of an output curve at any given temperature is directly proportional to the differential heat flow between a sample and reference material and in which the area under the measured curve is directly proportional to the total differential calorific input. By this technique, coal samples can be studied under experimental conditions that simulate spontaneous heating process of materials (Mahajan et al., 1976).

In the present investigation a DSC-7 differential scanning calorimeter manufactured by Perkin-Elmer, Germany was used. The complete experimental set up comprises of the differential scanning calorimeter, a thermal analysis controller, purge gas supply arrangement, a computer with softwares and a graphic plotter. The photographic view of the experimental set up is presented in Figure 3.

(1) Differential scanning calorimeter
(2) Thermal analysis controller
(3) Personal computer
(4) Graphics plotter

Figure3. Photographic view showing experimental setup of Differential Scanning Calorimeter

4.3.1 Differential Scanning Calorimeter

The Perkin-Elmer DSC-7 differential scanning calorimeter is of power compensated type. It is based on “thermal null balance” principle in which energy absorbed or evolved by the sample is compensated by adding or subtracting an equivalent amount of electrical energy to a heater located in the sample holder throughout the controlled temperature program. Platinum resistance heaters and thermometers are used in the DSC-7 calorimeter to accomplish the temperature and energy measurements in this design. Any energy difference in the independent supplies to the sample and the reference is then recorded against the program temperature. Thermal events in the sample thus appear as deviations from the DSC baseline, in either endothermic or exothermic direction, depending upon whether more or less energy has to be supplied to the sample relative to the reference material. In DSC therefore, endothermic responses are usually represented as being positive, i.e. above the baseline, corresponding to an increased transfer of heat to the samples compared to the reference.

4.3.2 Experimental Procedure Adopted for DSC Study

The cover and the silver lid were removed from the DSC cell. The instrument was calibrated using standard zinc and indium samples. About 10mg coal sample of -212µ was taken in the perforated aluminium sample pans and weighed accurately using a Mettler AE-240 single pan electronic balance (0.01mg sensitivity). After placing cover on the pan it was crimped using a crimper press. The crimped container with the sample was put on the sample furnace while the reference furnace was kept empty.The DSC thermogram was now obtained upto 550^0C at a heating rate of 30^0C per minute with oxygen as purge gas at a rate of 20cc per minute.

The above procedure was followed to obtain the DSC thermograms for all the 30 coal samples. Four representative thermograms for sample nos. 1, 20, 21, 30 are presented in Figures 4(a), (b), (c) and (d) respectively.

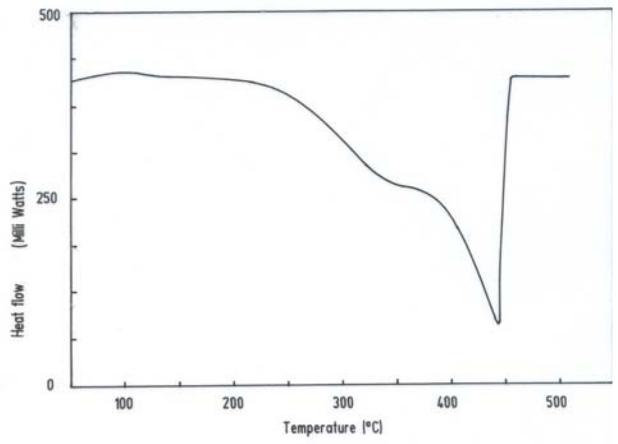

Figure 4(a). DSC thermogram for sample no. 1

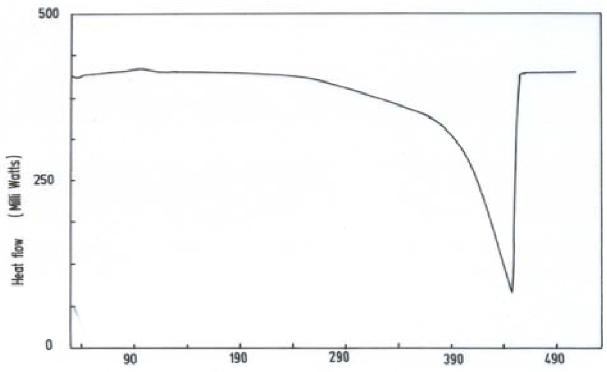

Figure 4(b). DSC thermogram for sample no. 20

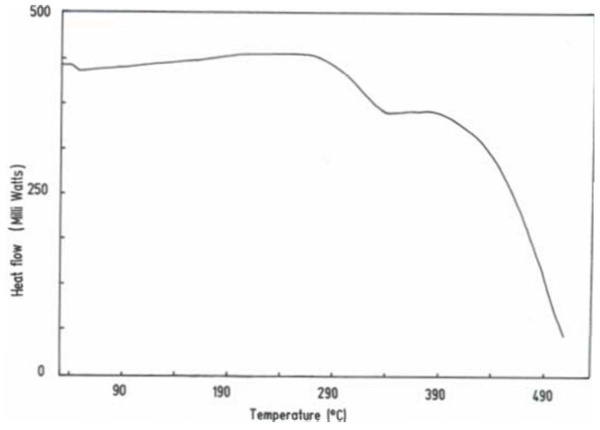

Figure 4(c). DSC thermogram for sample no. 21

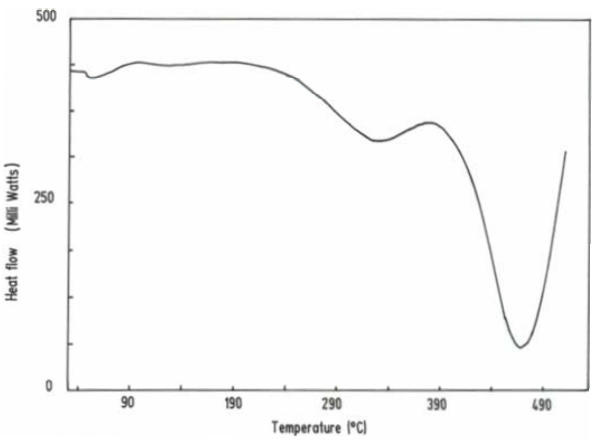

Figure 4(d). DSC thermogram for sample no. 30

4.3.3 Analysis of Results from DSC thermograms

The DSC thermograms represent the various reactions occurring during coal oxidation. It can be observed from all the thermograms that initially the endothermic reaction dominates followed by the exothermic reactions. The temperature of initiation of the exothermic reaction can be considered as an indicator of spontaneous heating susceptibility of coal samples, which is known as the onset temperature. The lower is this temperature, higher is the spontaneous heating susceptibility.

The procedure of determination of onset temperature or characteristics temperature (T_o) of exothermic reaction is as given below:

- A tangent is drawn at the inflexion point of the pre-transition.
- A second tangent is drawn at the greatest slope of the first exothermic reaction.
- The intersection of the two tangents gives the characteristics or onset temperature.

Determination of onset temperature for sample no. 24 is presented in Figure 5 for ready reference.

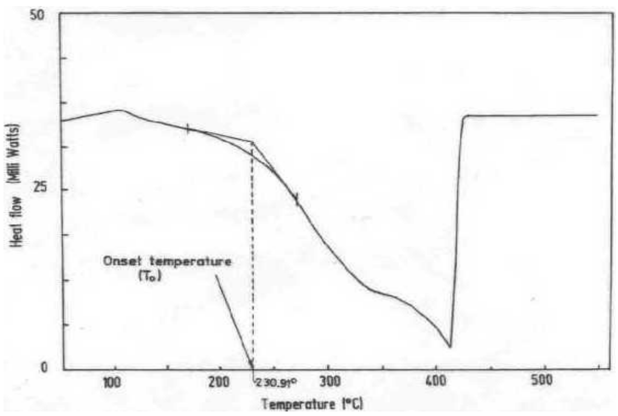

Figure 5. Determination of onset temperature for sample no. 24

In DSC-7 calorimeter the onset temperature is determined by selecting the point (temperature) at which the tangents are to be drawn. The onset temperature is read out (displayed) directly from the intersection of the tangents.

The onset temperature of all the 30 coal samples are determined following the above procedure and the results are also presented in Table 3.

Table 3. Results of different susceptibility indices of coal samples

Sample no.	CPT (^{0}C)	DTA				DSC
		T_c (^{0}C)	IIA Slope	IIB Slope	II Slope	T_0 (^{0}C)
1	149	171.87	0.10	0.20	0.11	249.37
2	149	136.47	0.45	0.19	0.28	235.21
3	146	137.33	0.41	0.27	0.29	240.76
4	148	124.82	0.30	0.23	0.23	240.02
5	147	153.34	0.26	0.09	0.13	234.30
6	148	157.89	0.22	0.12	0.14	236.39
7	155	158.46	0.24	0.10	0.13	225.40
8	150	127.69	0.45	0.31	0.33	233.22
9	148	160.00	0.03	0.19	0.08	236.96
10	147	146.25	0.20	0.22	0.18	229.17
11	144	145.88	0.57	0.13	0.21	229.89
12	144	148.33	0.10	0.23	0.12	226.66
13	150	135.88	0.16	0.58	0.47	240.79
14	151	140.67	0.20	0.48	0.43	242.75
15	142	155.71	0.13	0.40	0.21	233.89
16	150	165.71	0.04	0.15	0.07	237.77
17	148	178.46	0.04	0.23	0.09	245.63
18	141	168.33	0.05	0.20	0.08	245.84
19	143	146.25	0.16	0.37	0.30	248.90
20	154	152.94	0.17	0.23	0.21	245.75
21	180	188.98	0.07	0.36	0.17	286.02
22	155	128.38	0.18	0.32	0.20	235.20
23	160	169.66	0.10	0.38	0.15	259.46
24	152	162.30	0.16	0.18	0.17	230.91
25	150	129.33	0.11	0.25	0.19	217.61
26	144	132.94	0.47	0.41	0.41	225.94
27	152.5	136.87	0.27	0.35	0.30	216.28
28	155	145.33	0.16	0.29	0.21	224.14
29	150	128.00	0.45	0.44	0.42	222.06
30	138	122.67	0.35	0.32	0.32	235.32

5 CORRELATION ANALYSIS BETWEEN SUSCEPTIBILITY INDICES AND INTRINSIC PROPERTIES

The susceptibility of coal to spontaneous combustion depends on its intrinsic properties. Therefore, the correlation studies have been carried out between the different susceptibility indices and the coal characteristics as obtained from proximate and ultimate. The susceptibility indices are taken as dependent variables and each constituent obtained from the proximate and ultimate analyses as an independent variable. The correlation coefficients obtained in all cases are presented in Table 4.

Table 4. Correlation coefficients between different susceptibility indices and results of proximate and ultimate

Sl. no.	Susc. Indices → Intrinsic properties ↓	CPT (^{0}C)	DTA				T_0 (^{0}C)
			T_c (^{0}C)	IIA slope	IIB slope	II slope	
1.	M	0.76	0.33	0.77	0.32	0.49	0.69
2.	VM	0.65	0.72	0.49	0.41	0.39	0.74
3.	A	0.50	0.39	0.71	0.25	0.44	0.43
4.	C	0.47	0.51	0.31	0.28	0.30	0.56
5.	H	0.45	0.31	0.57	0.45	0.49	0.06
6.	O	0.79	0.38	0.60	0.17	0.29	0.63

It can be observed from the above table that in case of DTA, the constituents of proximate analysis shows comparatively better correlation with the average slope of stage IIA of DTA thermogram than the other parameters (T_c, IIB slope and II slope) and this parameter may be taken as a measure of the spontaneous heating susceptibility of coals. In general, the correlation between the different susceptibility indices and the constituents of proximate analysis is better than the correlation between the susceptibility indices and the constituents obtained from ultimate analysis. Therefore, an attempt has been made to study the combined influence of moisture, volatile matter and ash on different susceptibility indices by multivariable analysis and these are presented in Table 5.

A study of Table 5 indicates that the correlation coefficients between these three susceptibility indices (CPT, average slope of stage IIA of DTA thermogram and onset temperature from DSC thermogram) and the constituents of proximate analysis taken together has improved. Thus, it may be considered that moisture, volatile matter and ash contents of coal jointly influence the spontaneous heating susceptibility of coals.

Table 5. Correlation between the constituents of proximate analysis and different susceptibility indices

Sl. no.	Dependent variable	Empirical relation	Correlation coefficient
1	CPT	$146.7262M^{-0.027} + 7417.875 VM^{-2.056} + 353.6539A^{-1.925}$	0.76
2	IIA	$0.016M^{1.052} - 3.744VM^{-0.705} + 0.698A^{-0.151}$	0.83
5	T_o	$103M^{-0.037} + 609.6461VM^{0.443} + 1638.623A^{-2.996}$	0.85

6 CONCLUSIONS

The characteristics temperature (T_c) obtained from the thermogram of DTA is taken as a measure of spontaneous heating susceptibility of coals (Gouws and Wade 1989), but in the present work its correlation with the constituents of proximate and ultimate analyses were found to be less than 0.51 (Table 4) and therefore is not suitable as a measure of the spontaneous heating tendency of the coals. However, the stage IIA slope obtained from these thermograms shows a better correlation with the constituents of proximate analysis (Tables 4 and 5). Therefore this parameter (IIA slope) may be considered as the measure of the spontaneous heating susceptibility of coals.

It may be observed from Table 3 that seams like Samla(sample no. 24), Burra Dhemo (sample no. 25), Chirimiri-III (sample no. 28), and Kampti-IVB (sample no. 29) are showing CPT values higher than 150^0C, indicating that the coals are less susceptible, where as in actual field condition they are highly susceptible. The onset temperatures (T_0) of DSC thermograms are lower for the aforementioned samples indicating that these coal samples are highly susceptible to spontaneous heating. The IIA slope obtained from DTA for Samla(sample no. 24), Burra Dhemo (sample no. 25) and Chirimiri-III (sample no. 28) are less (below 0.16), which implies that they are less prone to spontaneous heating which is contradictory to the field results. However, the IIA slope of DTA for Kampti-IVB (sample no. 29) and Jhingurda seam (sample no. 30) are higher, implying that they are highly susceptible and is comparable with the field observation

The CPT value for Bastacola – 0 seam (sample no. 21) is 180^0C and is the highest showing that it is least susceptible to spontaneous heating. The IIA slope of DTA is 0.07 and is the lowest and T_o of DSC thermogram is 286.02^0C, which is highest among all the samples (Table 3). These results are in agreement with the results obtained from CPT and actual field observations.

It could be summarised from the above observations that differential scanning calorimetry predicts the spontaneous heating susceptibility more accurately than crossing point temperature and differential thermal analysis, for the coal samples studied in this investigation.

It takes approximately 3 hours to determine the CPT of a coal sample and failure of power supply for a few minutes during experiments, especially when the temperature approaches the crossing point, gives erroneous value of CPT and the whole experiment has to be repeated. In case of DTA, it is difficult to maintain an oxidising atmosphere for the experiments and it takes about 1.5 hour for the completion of an experiment. The onset temperature, T_0 from DSC thermograms can be obtained within 30 minutes and thus much of the time and energy can be saved.

The acceptability of a method for determining spontaneous heating characteristics of coal mainly depends upon how closely it predicts the spontaneous heating behaviour in the field conditions. Considering this, it may be concluded that the onset temperature obtained from differential scanning calorimetry may be a better method than crossing point temperature.

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