

10.1016/j.jlp.2018.09.00

Authors Accepted Manuscript: Journal of Loss Prevention

September 2018

1

Development a Modified Crossing Point Temperature (CPT_{HR}) Method to Assess Spontaneous Combustion Propensity of Coal and its Chemo-metric Analysis

N. K. Mohalik ¹[0000-0003-1521-432X] **E. Lester**² and **I. S. Lowndes**²

1 Principal Scientist, Mine Ventilation Division, CSIR- CIMFR, Barwa Road, Dhanbad, India

2 Faculty of Engineering, University of Nottingham, Nottingham, NG7 2RD, UK

niroj.mohalik@gmail.com

Abstract. Spontaneous combustion of Indian coals was investigated using spontaneous combustion rig at University of Nottingham, UK to assess their susceptibility. In the present study authors have used eleven coal samples collected from the Jharia coalfield (JCF), India. Both thermal as well as gas profiles from spontaneous combustion rig were studied critically to develop a modified crossing point temperature to assess the spontaneous combustion propensity of coal. The product of combustion gases (CO , CO_2 , CH_4 , and H_2) emitted from spontaneous combustion rig within the temperature range between ambient and $300\text{ }^\circ\text{C}$ of these coal samples were studied. The initial product of combustion gas i.e. CO followed by H_2 indicates propensity towards oxidation of coal in laboratory condition for Jharia coalfield. The temperatures at which CO and H_2 releases in the level of 50ppm (T_{CO50} , T_{H250}), crossing point temperature of coal (CPT_{CT}) (temperature of coal and bath temperature is same) and modified crossing point temperature of coal (CPT_{HR}) (temperature where dT/dt is equal to $2.0\text{ }^\circ\text{Cmin}^{-1}$ because heating rate is double of programme temperature $1\text{ }^\circ\text{Cmin}^{-1}$) determined from spontaneous combustion rig categorises the coal as per their propensity to spontaneous combustion. The results of these methods have been compared with other standard method i.e. crossing point temperature method – India, which is widely adopted in Indian regulatory bodies to verify the suitability of this method.

Keywords: Spontaneous Combustion, Spontaneous combustion Rig, Crossing Point Temperature

1 Introduction

Thermal studies of coal are widely used all over the world. In thermal studies, different countries have adopted different methods to assess the propensity of coals to spontaneous combustion in the laboratory. Researchers have employed a range of different thermal methods, including - crossing point temperature (CPT) and ignition point temperature (IPT), puff temperature (PT), Basket heating test method, Chen's method, Critical air blast method, Olpinski index, adiabatic calorimetric (SHT - USA, R70 - Australia), isothermal calorimetric, differential thermal analysis (DTA), differential scanning calorimetric (DSC) and thermogravimetric analysis (TGA), to determine the susceptibility of coal towards spontaneous heating. Amongst these assessment techniques: the CPT method is widely used in India, South Africa, Poland, China, and Turkey; the isothermal and adiabatic calorimetric methods are used in UK, USA and Australia and the puff temperature and Olpinski index methods are widely used in Russia. Subsequently, a number of further modifications to CPT methods with respect to their experimental parameters and the apparatus design have been proposed (Bagchi, 1965; Banerjee, Banerjee, & Chakravorty, 1970; Chamberlain & Hall., 1973; X. Chen, Li, Wang, & Zhang, 2018; X. D. Chen & Chong, 1997; Feng, Chakravorty, & Cochrane, 1973; Ganguli & Banerjee, 1953; Gouws & Wade, 1989a, 1989b; Kreulen, 1948; Nimaje, Tripathy, & Nanda, 2013; D. C. Panigrahi & Sahu, 2004; D. C. Panigrahi, Saxena, V. K, Udaybhanu, G, , 2000; Parr & Coons, 1925; Sahu, Padhee, & Mahapatra, 2011; Tideswell & Wheeler, 1920). The higher the determined CPT value, the less will be the susceptibility of coal to spontaneous combustion. In India, CPT and moisture content of coal data is required by mine planner to design the mine, mine operators, and regulators for ensuring the safety of miners and machines. These two techniques are very simple, basic practices, user friendly and often time consuming process. The repeatability and reproducibility are sometimes uncertain for their reliability to a laboratory as well as field condition. However, it is conceded that the results of this laboratory analytical techniques may often be contradicted by actual mine conditions due to the influence of extraneous parameters such as mining,

geological and environmental parameters. The low temperature oxidation process can be divided into three stages, the first stage (30 °C critical temperature), the second stage (> 90 °C critical temperature dry cracking temperature), and the third stage (> 140 °C over the dry temperature)(G. Wang, Liu, et al., 2018). The potential emission of CO may endanger the health and safety of workers, especially in underground mines(G. Wang, Wang, et al., 2018). Struminski and Madeja-Struminska (2005) developed an approximate method of determining the temperature of the centre of the spontaneous fire on the basis of CO released from coal after spontaneous combustion/ fire. Similarly, the CMR 1957 of India, states that if the CO content of the ventilating air exiting a working face is greater than 50 ppm then the mining activities should cease and the workers should be withdrawn.

This paper summaries about the experimental studies to develop a modified crossing point temperature for 11 coal samples collected from mines within the Jharia Coalfield (JCF), India. It also discusses about the temperature at which the concentration of combustion gas release i.e. CO and H₂ having 50 ppm (T_{CO50}, and T_{H250}) to determine oxidation characteristics of the coal samples. Above study will help mine regulators and mine operators to predict spontaneous combustion/ fire risk for ensuring the safety of miners and machine in the era of sustainable growth.

2.0 Materials and Methods

About 2 kg of representative coal sample from different parts of the Jharia coalfield (JCF) containing both fiery and non-fiery coal seams were collected and different sizes were prepared as per requirement keeping aerial oxidation minimum. The locations of the eleven coal seams sampled are detailed in Table 1. Among these eleven samples, five samples (sample number: 1, 2, 3, 4 and 5) are having present and past history of fires & spontaneous heating and rest of these is not having any observation of fire. Proximate analysis, ultimate analysis and calorific value of all samples were carried out following the ASTM standard on a received basis. The rank analysis (BS-6127-5, 1995) of the prepared polished blocks was carried out using a microscope. The crossing point

temperature (Indian method - CPT_I) of coal samples were determined as per the Directorate General Mine Safety (DGMS) circular i.e. DGMS Cir.Tech.3/1975. The results of the proximate analysis, ultimate analysis, calorific value, rank and CPT_I for all the samples were presented in Table 1. Five different prepared samples of each coal type were tested, and the mean data of these tests are presented.

2.1 Experimental setup and procedure

A spontaneous combustion rig (spontaneous combustion rig) comprises of a vertical furnace, sample holder, thermocouples with their attachment and configuration, a controlled gas exhaust system to collect the product gases, and a multi gas analyser (Fig. 1). This rig was measuring thermal profile inside (both vertical & radial) and outside of heated coal sample holder. The detail of the sensor position (thirteen numbers of thermocouples - K type) inside the sample holder are indicated in Fig.1. The measurements were collected through a data interface to a personal computer. The coal sample holder is formed by a stainless steel cylinder (length-0.08 m and inner diameter - 0.05 m, wall thickness - 1mm), with a capacity to hold 100 g of coal of - 272 size micron. All of the heated coal samples were exposed to slow ramp rate to increases in temperature having following experimental parameters: 100g of sample, a heating rate of $1\text{ }^\circ\text{Cmin}^{-1}$, atmospheric air flow rate of 200 mlmin^{-1} , under an oxidative atmosphere from $20\text{ }^\circ\text{C}$ to $350\text{ }^\circ\text{C}$. The exhaust gases are passed through a multi gas analyser (MX6iBrid) continuously to determine the different gas species concentrations i.e. CO , H_2 , CO_2 , CH_4 , and O_2 . An analysis of the results of these experiments is subsequently conducted to identify coal that is prone to self-oxidation and less reactive coal at low temperatures.

3.0 Analysis of results

3.1 Basic coal characteristics

The proximate analyses of coal samples reveals that M, VM and A content of the samples varies across a range from 0.61% (sample 5) to 1.30 % (sample 4); 20.43 % (sample 9) to 29.55 % (sample 3); 7.78 % (sample 3) to 20.94 % (sample 5) respectively. Similarly the ultimate analysis results reveals that C, H, N, S & O of the

tested coal samples varies from 63.78 % (sample 5) to 72.72 % (sample 2); 3.89 % (sample 9) to 4.81 % (sample 3); 1.27 % (sample 5) to 1.64 % (sample 11); 0.0 % (sample 9) to 0.44% (sample 6) respectively. The gross calorific values (GCV) vary across a range from 25.59 MJkg⁻¹ (sample 5) to 29.64 MJkg⁻¹ (sample 1). The CPT_I vary across a range from 136 °C with sample 3 to 171 °C with sample 6. The CPT_I values determined for samples 3 is low (<140 °C) which is categorized as highly prone to spontaneous combustion, whereas sample number 1, 2, 4, 7 and 8 are in the range of 140 to 160 °C which are moderately susceptible and sample number 5, 6, 9, 10 and 11 are high (>160 °C), which denotes low susceptibility to spontaneous combustion.

3.2 Thermal Analysis

The result of thermal profile concludes that initially rise in coal bed temperature was less than of the air in the oven for all samples. One of the thermocouples located at the centre top (CT) of the sample holder, achieves a maximum temperature very close to 550°C during experiment from ambient to 350°C. Other thermocouples achieve maximum temperatures of up to 300 °C for all samples. The stage at which both the coal bed and bath temperature (furnace programme temperature [FPT]) are equal is known as crossing point temperature. Following the achievement of the CPT, the coal sample temperatures are observed to increase at a much faster rate until the ignition temperature of the coal is reached. The rate of the temperature rise then slows down once the coal reaches its ignition point temperature. The thermal profile of the different thermocouples varying from the centre of the sample holder to wall, depends upon the thermal conductivity of coal, the packing density of sample in sample holder, the distance between outer wall of sample holder and furnace wall. The evolution of the temperature measured at the centre top with respect to the FPT is shown in Fig. 2. An analysis of the data on these figures reveals there is no ignition after the hot spot in the centre middle and bottom sensor because of lack of oxygen except at centre top (presence of oxygen at the top). So, the centre bottom and middle temperature cannot reach crossing point temperature because it did not trigger the ignition of coal. This temperature difference recorded along the central vertical axis thermocouples (centre)

occur due to the reactions that take place at the top of sample holder and not in the middle (4.5 mm vertical distance from the top of sample holder) or bottom (5.5 mm vertical distance from the top of sample holder) of the sample. Consequently, crossing point temperature of each coal sample was determined with respect to centre top i.e. CPT_{CT} . The computed values of the CPT_{CT} are shown in Table 2. An analysis of the thermal profile data trends concludes that the CPT_{CT} values vary across a range between 196 °C (sample 8) to 231°C (sample 5). The CPT_{CT} values follow a similar trend mainly due to the availability of oxygen at the top of the sample holder. The present study investigated that the recorded heating rate (dT/dt) of coal samples of centre top thermocouple could be employed to classify the coal samples. An attempt is made to simplify and find out the trigger points of reaction where the reaction starts exponentially and when the point comes, the rate of reaction slows down (Fig.3). The temperature recorded by the centre top thermocouple is the most crucial to determine the spontaneous mechanism of coal, whereas two other thermocouple measurements (centre bottom and centre middle) play no role (Fig. 3). This proves the concept of fire tetrahedral i.e. coal [fuel], oxygen, heat and chemical reaction. The trigger point of the reaction may be considered as modified crossing point temperature i.e. CPT_{HR} (temperature where dT/dt is equal to $2.0\text{ }^{\circ}\text{Cmin}^{-1}$ because the heating rate is double of programmed temperature $1\text{ }^{\circ}\text{Cmin}^{-1}$). The computed values of the CPT_{HR} are shown in Table 2.

3.3 Gas Compositional Analysis

The MX6IBrid multi gas analyser was used to continuously measure and record the concentration of the product of combustion gases i.e. CO , H_2 , CO_2 , CH_4 , and O_2 . During the execution of the experiment, the product gases liberated on the heating of the coal samples were measured every 30 seconds and all this data matched against the thermal data record to determine the product of combustion (POC) gases released at a different temperature. The results of CO and H_2 gases for all samples with respect to time are given in Fig. 4 & 5.

An analysis of the gas emission profile data measured from the heated samples reveals that the CO and H₂ released are at low temperatures (<130 °C) (Marinov (a); Marinov (b), 1977). The production of CO and H₂ is more than 1500 ppm and 2000 ppm for most of the samples except sample number 4, 5 and 9. A study from above figure reveals that in initial phase there was a slow increase of combustion gases followed by immediate increases of combustion gases (Liu & Qin, 2017; Trenczek, 2008; H. Wang, Dlugogorski, & Kennedy, 2003; Xie & Pan, 2001). All of the coal samples heated under laboratory conditions initially released CO and H₂ gases, which may indicate the onset of spontaneous heating confirmed by previous studies (Mohalik, Singh, Pandey, & Singh, 2006; Struminski & Madeja-Struminska, 2005). The thermal profiles (temperature of top centre thermocouple) were determined to maintain the gas emissions (CO and H₂)h at 50 ppm which are known as T_{CO50} and T_{H250} (Fig. 6). The thermal profile results for above two are given in Table 2.

3.4 Discussion

An analysis of the thermal profile data trends concludes the following:

- The CPT_{CT} values vary across a range between 196 °C (sample 8) to 231°C (sample 5) and CPT_{HR} varies across a range between 151 °C (sample 3) to 207 °C (sample 5). The CPT_{CT} and CPT_{HR} values follow a similar trend, mainly due to the availability of oxygen at the top of the sample holder. With the exception of sample number 5, the CPT_{HR} values of all of the other samples are follow a similar trend to the CPT (Indian method) values.
- The samples 1, 2 and 3 exhibit low CPT_{HR} values (< 170 °C) which means they are prone to spontaneous heating susceptibility, whereas samples number 5, 6, 9 and 10 have values in the range of 170 °C to 200 °C which means they are moderately susceptible. The sample number 5 has high CPT_{HR} value (>200 °C) which is less prone to spontaneous heating. The samples 1, 2 and 3 are more prone to spontaneous heating as compared to other samples as CPT_{HR} may be

shown to be inversely proportional to the proneness of coal to spontaneous heating.

- An analysis of the thermal profile data of the heated coal samples presented in table 2 concludes that the samples 1, 2 and 3 have low thermal values. The sample number 5, 6, 9 and 10 have high thermal values for CO at 50 ppm, and H₂ at 50 ppm. The sample 1, 2 and 3 are more prone to spontaneous heating which verifies CPT Indian method and thermal profiles from the spontaneous combustion rig.

4.0 Chemo-metric Analysis

4.1 Correlation Analysis

The proximate, ultimate analysis, GCV data obtained from the above studies were subsequently compared statistically using correlation analysis, multivariate analysis (i.e. principal component and classification analysis (PCCA), hierarchal clustering techniques – joining tree and fixed nonlinear regression models (FNRM)). Statistica 7.1 statistical package was used (Rencher, 2002; STATISTICA-7.0, 2004) to perform correlation studies to identify potential relationships between the different spontaneous combustion susceptibility indices (CPT, CPT_{CT}, CPT_{HR}, T_{CO50}, and T_{H250}) and the coal characteristic data provided by the proximate, ultimate and GCV, analyses of coal samples. The values of the correlation coefficients determined ($p < 0.05$ confidence interval) for the above studies are presented in Table 3. A study of the data presented in Table 3 reveals that CPT_I and CPT_{HR}, possess the highest significance with the ash content ($r = 0.86$, $r = 0.92$) and GCV ($r = -0.81$, $r = -0.93$). The positive correlation coefficients reveal that it has a positive correlation, whereas negative correlation indicates a weak relationship.

4.2 Principal component analysis (PCA)

The PCA technique is widely applied to analyse highly complex datasets. The method seeks to reduce the dimensionality of the data set and to identify relationships between variables. The PCA analyses performed considered the relationships between the

following **ten** determined coal characteristic variables (moisture [M], ash [A], volatile matter [VM_{daf}], fixed carbon [FC_{daf}], carbon [C_{daf}] hydrogen [H_{daf}], nitrogen [N_{daf}], sulphur [S_{daf}] oxygen [O_{daf}] and calorific value [GCV]) with the **five** susceptibility indices determined for each coal sample (CPT_I, CPT_{CT}, CPT_{HR}, T_{CO50}, and T_{H250}). For this study, the principal components (PCs) with Eigen values greater than 1.0 were considered. However, the total variance for the given data sets, is observed to vary by 89.15% for the first three PCs, and found very small values for the remaining seven PCs (Table 4). The eigenvalues of these three PCs, modify the magnitude of the corresponding eigenvectors significantly (Table 4). The eigenvectors with the largest eigenvalues identify the parameters with the strongest correlation in the data set. Similarly, the scree plot finds the factorial loadings where the observed decrease in eigenvalues appears to level off to the right of the plot. As a result, the first three PCs were selected for the principal component matrix. The factorial loadings and their projections of the variables on the factor plane (1x2) and (1x3) are depicted in Table 5 and Fig.7. Factorial loadings close to 1 indicate stronger correlations (Table 5). The projection of the first two factorial loading plots indicates whether the parameters are correlated or not. If the plotted variables are close to the centre, it means that some information may be carried over to other axes. The projection of the variables on the factor plane 1x2 and factorial plane 1x3 shows that first group i.e. GCV, M, VM, and O are far from center but close to each other. Similarly second group (CPT_I, CPT_{CT}, CPT_{HR}, and T_{CO50}) are on the opposite side of the centre as well as to the first group. An analysis of a plot of the weighted parameter will indicate a significant correlation where these parameters are spatially grouped together. As first the group is on opposite sides of the second group so, they are negatively correlated.

4.3 Hierarchical Clustering

An attempt has been made to classify the coal tested by the application of hierarchical clustering using euclidian distance method to measure the distance function and an average linkage method as similarity measures. The classification of coal seams with independent variable (parameters of proximate analysis; elemental analysis; calorific

value) and one of the susceptibility indices (CPT Indian method, CPTs from spontaneous combustion rig) at a time are considered together (Khare, Baruah, & Rao, 2011; D. C. Panigrahi & Sahu, 2004). The hierarchical clustering (joining tree) was performed using STATISTICA 7 statistical software. The dendrograms results were derived using the independent variables (defined by the parameter variables determined by the proximate, elemental, and calorific value) and the dependent variables parameters (CPT_i) is given in Fig.8. The above procedure was repeated to obtain dendrograms for other five spontaneous heating susceptibility experiments i.e. crossing point temperature–spontaneous combustion rig experiments. The dendrograms obtained for the above five susceptibility indices (crossing point temperature –Indian method, and spontaneous combustion rig experiments) are as presented in Fig. 8 to 12. An analysis of the five different dendrograms (Fig. 8 to 12) reveals the following:

- The number of clusters obtained from the dendrograms for these five cases is 3, each having an equal linkage distance of 15 except Fig. 12. This indicates that the identified clusters are natural. All the samples are forced to one cluster at a linkage distance of approximately 40.
- If the number of clusters remains the same (i.e. 3) then the linkage distance could be achieved as a linkage distance of 15, 12, 15, 12 and 17 respectively. It may be concluded that in all cases three clusters are chosen for the classification of coals seams. The details of the clusters identified from the dendrograms for coal samples tested are displayed in Table 6.
- The eleven coal samples studied were divided into three categories as per their susceptibility to self-heating i.e. low (first cluster: coal samples 1, 2 &4), medium (second and third cluster: coal samples 3, 7, 8, 9 & 10) and high (fourth cluster: coal samples 5, 6 &11). The samples 1, 2, 3 and 4 have been identified as being more prone to spontaneous heating from the experimental investigation which is further confirmed by the cluster analysis. The sample number 1, 2, 3 and 4 exhibit actual experimentally measurable characteristics

and confirmed the occurrence of the fire at operating or closed mines (Table 1).

With the exception of sample number 5, the combined evidence provided by an examination of all of the experimental results and the subsequent statistical analysis of this data and field observations corroborate the same conclusions.

It is proposed that CPT_{HR} may be used for the determination of spontaneous heating characteristics of coal.

4.4 Fixed Nonlinear Regression Models (FNRM)

Fixed nonlinear multiple regression is to learn more about the relationship between several independent variables and a dependent variable. In multiple regression, the regression coefficient R can assume a value between 0 and 1. This study employs the following set of standard functions (including X^2 , X^3 , X^4 , X^5 , \sqrt{X} , $\ln X$, $\log X$, and $1/X$) to specify nonlinear transformations. The above analyses data sets (M, A, VM) were considered as the independent parameters and the susceptibility indices (CPT_I , CPT_{HR}) were taken as dependent parameters, where the dependence of these variables was considered sequentially. The fitness of each model equation is determined by an analysis of the computed regression coefficients, level of significance and standard error. Accordingly, one model equations were tested for each of the susceptibility indices. A summary of the fitness of each of these models to each of the susceptibility indices in terms of the computed R-Squared, Adjusted R-squared and standard error of mean are presented in Table 7. 'R-Squared' value of these model equations obtained from non-linear regression analysis ranges from 0.64 & 0.81; and the standard error estimate of 7.07 and 6.61 respectively. To develop each model equation it has been observed that model was significant in the range 0.015 to 0.001. Therefore by using multiple fixed nonlinear regression analysis of the experimental data, it may be concluded that the susceptibility index i.e. CPT_{HR} may be used to categorize/classify the coal seam, which also correlates with a standard method like CPT_I .

5.0 Validation of Results with CPT Indian Method (CPT_I)

Correlation studies were carried out to classify and validate the results from spontaneous combustion rig study with established potential methods of spontaneous heating susceptibility indices. The correlation coefficients derived for a $p < 0.05$ confidence interval for all the spontaneous heating susceptibility indices were given in Table 8. The results of Table 8 reveals that the CPT_I show the highest significance with the CPT_{CT} ($R^2=0.93$), CPT_{HR} ($R^2=0.87$) and T_{CO50} ($R^2=0.73$) and has poor correlation with the T_{H250}. The CPT_{CT} has better correlation with CPT_{HR} ($R^2=0.84$) and poor correlation with both the indices T_{CO50} and T_{H250}. The CPT_{HR} has better correlation with T_{CO50} ($R^2=0.84$) and poor correlation with both the indices T_{H250}. Similarly, the T_{CO50} has poor correlation with T_{H250}. The modified crossing point temperature study of all the coal samples is correlated with crossing point temperature Indian method (Fig. 13). It has been observed that CPT_{HR} results are well corroborated with CPT_I.

6.0 Conclusions

The present study has the results of a series of analytical investigations to characterize the characteristic properties viz. proximate, ultimate, GCV and spontaneous combustion susceptibility indices (CPT_I, CPT_{CT}, CPT_{HR}, T_{CO50}, and T_{H250}) for eleven coal samples across the Jharia Coalfield, India. The product of combustion gases (CO, CO₂, CH₄, and H₂) emitted from spontaneous combustion rig within the temperature range between ambient and 300 °C of these coal samples were studied. It has been observed that in this study temperature of coal samples reached 550 °C. The signature of gases released from heating reveals that the CO is released in low temperature range i.e. 60-120 °C whereas H₂ in the temperature range 80-140 °C. The initial product of combustion gas i.e. CO followed by H₂ indicates spontaneous combustion of coal in laboratory condition for Jharia coalfield.

A chemo metric analysis of the intrinsic properties of the coal samples i.e. moisture, ash and volatile matter on daf basis confirms that these parameters exhibit a positive correlation to the spontaneous combustion susceptibility indices. Multivariate analysis i.e. PCA, HC and FMNRA concludes that CPT_{HR} gives a better indicator for the study of

spontaneous combustion of coal as compared to CPT_I which further corroborates the experiments. The modified crossing point temperature of coal (CPT_{HR}) determined from spontaneous combustion rig categorizes the coal as per their propensity to spontaneous combustion. The results of these methods have been compared with other standard method i.e. crossing point temperature method, which is widely adopted in India to verify the suitability of this method.

Acknowledgement

Authors are grateful to Commonwealth Scholarship Commission, UK and University of Nottingham for their financial support (Commonwealth Scholarship and Fellowship Plan –2010, INCS-2010-192). The authors are obliged to Ministry of Human Resources and Development, Government of India and Council of Scientific and Industrial Research (CSIR) for their kind permission to avail the above fellowship. Authors acknowledge thanks to Director, CSIR-CIMFR, all staffs of Mine Fire, Ventilation and Miner's Safety Research Group, CSIR-CIMFR for necessary help for the sample collection.

References

1. Bagchi, S. (1965). An investigation on some factors affecting the determination of crossing point of coals. *J Mine Metal & Fuels*, 13(8), 243-247.
2. Banerjee, S. C., Banerjee, B. D., & Chakravorty, R. N. (1970). Rate studies of aerial oxidation of coal at low temperatures (30–170 °C). *Fuel*, 49(3), 324-331. doi: 10.1016/0016-2361(70)90024-4
3. BS-6127-5. (1995). Petrographic analysis of bituminous coal and anthracite – Part 5:method of determining microscopically the reflectance of vitrinite. *British standards institute*.
4. Chamberlain, E. A. C., & Hall., D. A. (1973). The Practical Early Detection of Spontaneous Combustion. *Colliery Guardian*, 221(5), 190-194.
5. Chen, X., Li, H., Wang, Q., & Zhang, Y. (2018). Experimental investigation on the macroscopic characteristic parameters of coal spontaneous combustion under adiabatic oxidation conditions with a mini combustion furnace. *Combustion Science and Technology*, 190(6), 1075-1095. doi: 10.1080/00102202.2018.1428570
6. Chen, X. D., & Chong, L. V. (1997). Several important issues related to the crossing-point-temperature (CPT) method for measuring self-ignition kinetics of combustible solids *Transactions of IChemE Part B: Process Safety and Environmental Protection* 76(B)(90-93).
7. Feng, K. K., Chakravorty, R. N., & Cochrane, T. S. (1973). Spontaneous combustion - A coal mining hazard. *Can Min Metall Bull*, 66(738), 75-84.
8. Ganguli, M. K., & Banerjee, N. G. (1953). Critical oxidation and ignition temperature of coal. *Indian Mining Metallurgy Association(IMMA) Review*, 2 1.
9. Gouws, M., & Wade, L. (1989a). The self-heating liability of coal: Predictions based on composite indices *Mining Science and Technology*, 9 81-85.
10. Gouws, M., & Wade, L. (1989b). The self-heating liability of coal: Predictions based on simple indices. *Mining Science and Technology*, 9 75-80.
11. Khare, P., Baruah, B. P., & Rao, P. G. (2011). Application of chemometrics to study the kinetics of coal pyrolysis: A novel approach. *Fuel*, 90(11), 3299-3305. doi: <http://dx.doi.org/10.1016/j.fuel.2011.05.017>
12. Kreulen, D. J. W. (1948). Elements of coal chemistry. *Nijgh and Van Ditmer N.V., Rotterdam*.

13. Liu, W., & Qin, Y. (2017). A quantitative approach to evaluate risks of spontaneous combustion in longwall gobs based on CO emissions at upper corner. *Fuel*, 210, 359-370. doi: <https://doi.org/10.1016/j.fuel.2017.08.083>
14. Marinov (a), V. N. (1977). Self-ignition and mechanisms of interaction of coal with oxygen at low temperatures. 1. Changes in the composition of coal heated at constant rate to 250 °C in air. *Fuel*, 56(2), 153-157. doi: [http://dx.doi.org/10.1016/0016-2361\(77\)90136-3](http://dx.doi.org/10.1016/0016-2361(77)90136-3)
15. Marinov (b), V. N. (1977). Self-ignition and mechanisms of interaction of coal with oxygen at low temperatures. 2. Changes in weight and thermal effects on gradual heating of coal in air in the range 20–300 °C. *Fuel*, 56(2), 158-164. doi: [http://dx.doi.org/10.1016/0016-2361\(77\)90137-5](http://dx.doi.org/10.1016/0016-2361(77)90137-5)
16. Mohalik, N. K., Singh, V. K., Pandey, J., & Singh, R. V. K. (2006). Proper sampling of mine gases, analysis and interpretation – a pre requisite for assessment of sealed off fire area. *Journal of Mines Metals & Fuels*, , 54(10 & 11), 210-217.
17. Nimaje, D. S., Tripathy, D. P., & Nanda, S. K. (2013). Development of regression models for assessing fire risk of some Indian coals. *I.J. Intelligent Systems and Applications, Published Online January 2013 in MECS*, 2, 52-58. doi: <http://www.mecs-press.org/>
18. Panigrahi, D. C., & Sahu, H. B. (2004). Classification of coal seams with respect to their spontaneous heating susceptibility—a neural network approach. *Geotechnical & Geological Engineering*, 22(4), 457-476. doi: 10.1023/b:gege.0000047040.70764.90
19. Panigrahi, D. C., Saxena, V. K., Udaybhanu, G, . (2000). A study of susceptibility of Indian coals to spontaneous combustion and its correlation with their their intrinsic properties. *Proceedings, 1st International Conference on Mine Environment and Ventilation, Dhanbad, India December*, 347-353.
20. Parr, S. W., & Coons, C. C. (1925). Carbon Dioxide as an Index of the Critical Oxidation Temperature for Coal in Storage. *Industrial & Engineering Chemistry*, 17(2), 118-120. doi: 10.1021/ie50182a006
21. Rencher, A. C. (2002). *Methods of Multivariate Analysis. A JOHN WILEY & SONS, INC. PUBLICATION.*
22. Sahu, H. B., Padhee, S., & Mahapatra, S. S. (2011). Prediction of spontaneous heating susceptibility of Indian coals using fuzzy logic and artificial neural network models. *Expert Systems with Applications*, 38(3), 2271-2282. doi: 10.1016/j.eswa.2010.08.015
23. STATISTICA-7.0. (2004). *Electronic manual statistica 7.*
24. Struminski, A., & Madeja-Struminska, B. (2005). Evaluation of arising spontaneous fire centre temperature and time of coal self-Ignition. *Eighth International Mine Ventilation Congress, Brisbane Queensland, 6 - 8 July*, 511-515.
25. Tideswell, F. V., & Wheeler, R. V. (1920). LXXXVI.-The oxidation of the ingredients of banded bituminous coal. Studies in the composition of coal. *Journal of the Chemical Society, Transactions*, 117(0), 794-801.
26. Trenczek, S. (2008). Levels of possible self-heating of coal against current research. *Archives of Mining Sciences*, 53(2), 293-317.
27. Wang, G., Liu, Q., Sun, L., Song, X., Du, W., Yan, D., & Wang, Y. (2018). Secondary Spontaneous Combustion Characteristics of Coal Based on Programed Temperature Experiments. *Journal of Energy Resources Technology*, 140(8), 082204-082204-082208. doi: 10.1115/1.4039659
28. Wang, G., Wang, Y., Sun, L., Song, X., Liu, Q., Xu, H., & Du, W. (2018). Study on the Low-Temperature Oxidation Law in the Co-Mining Face of Coal and Oil Shale in a Goaf—A Case Study in the Liangjia Coal Mine, China. *Energies*, 11(1), 174.
29. Wang, H., Dlugogorski, B. Z., & Kennedy, E. M. (2003). Coal oxidation at low temperatures: oxygen consumption, oxidation products, reaction mechanism and kinetic modelling. *Progress in Energy and Combustion Science*, 29(6), 487-513. doi: 10.1016/s0360-1285(03)00042-x
30. Xie, W., & Pan, W. P. (2001). Thermal characterization of materials using evolved gas analysis. *Journal of Thermal Analysis and Calorimetry*, 65(3), 669-685. doi: 10.1023/a:1011946707342

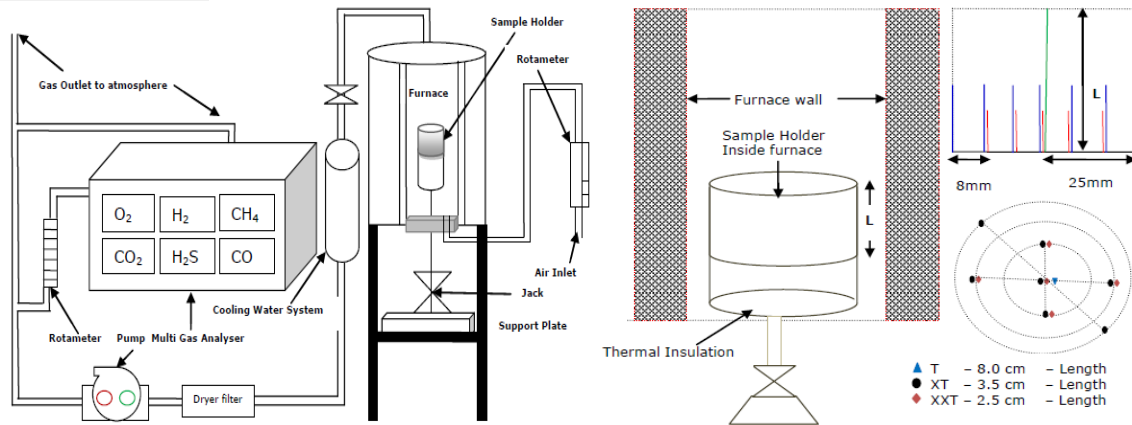


Fig. 1. Schematic of the UoN spontaneous combustion rig, sample holder and their sensor locations (not to scale)

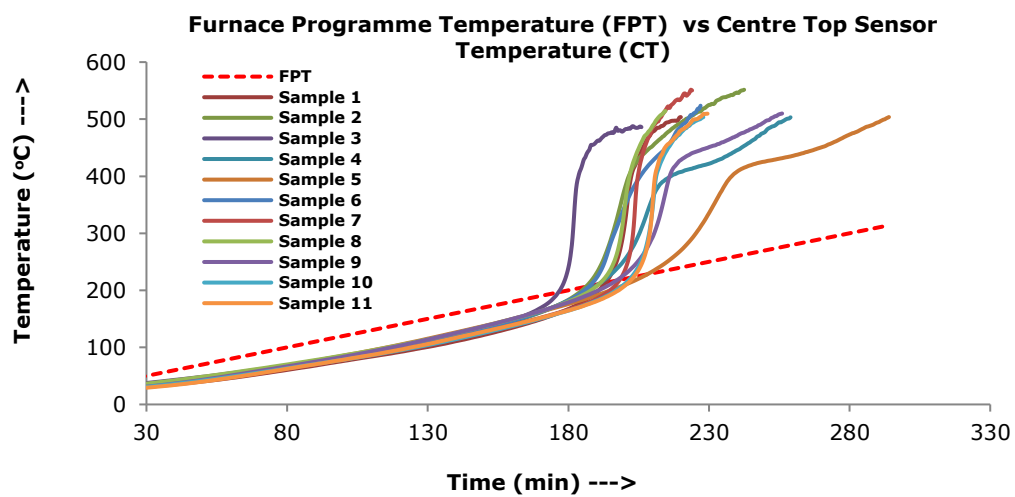


Fig. 2. CPT of coal samples (FPT-Centre Top)

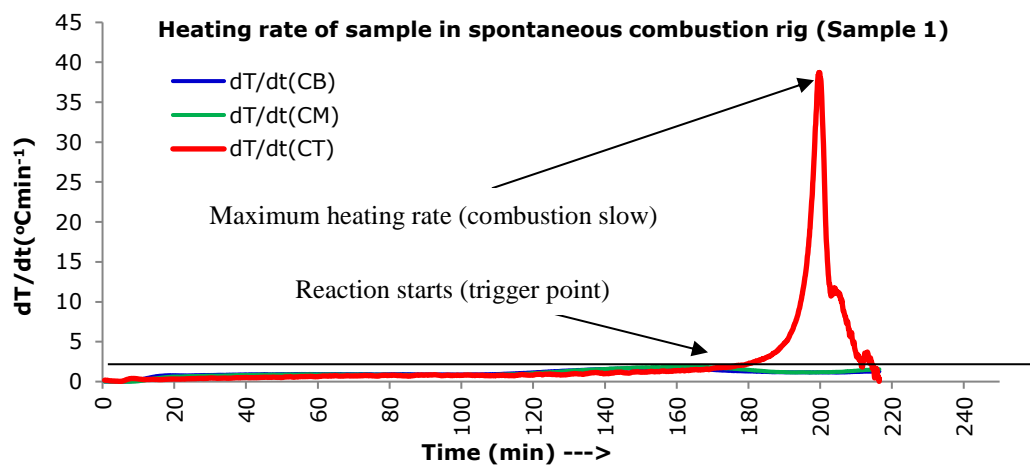


Fig. 3. CPT_{HR} of coal samples (dT/dt=2.0)

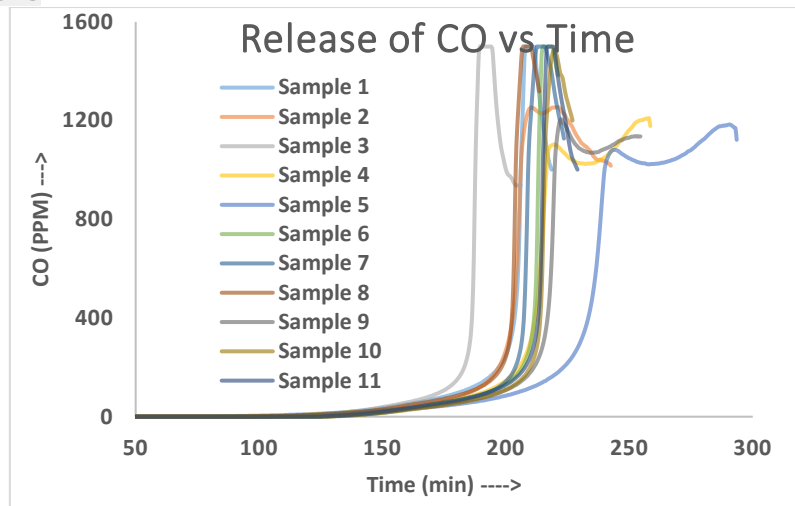


Fig. 4. Release of CO vs time

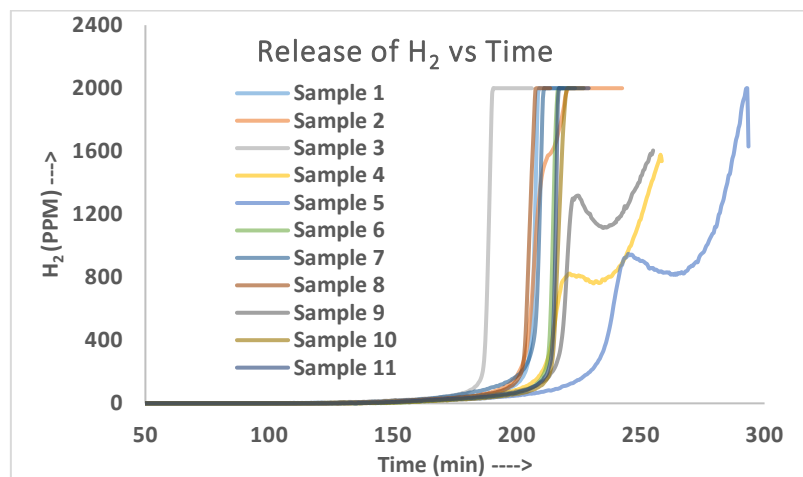


Fig. 5. Release of H₂ vs time

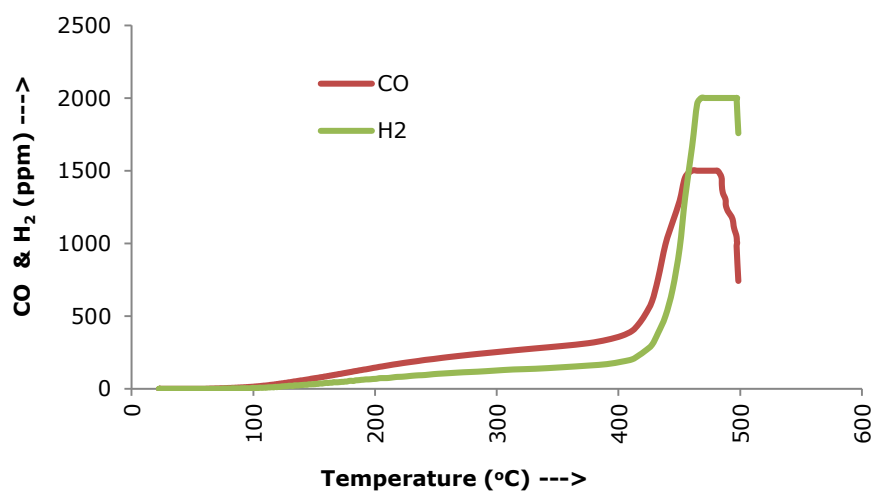


Fig. 6. Gas analysis result for all coal samples at CPT

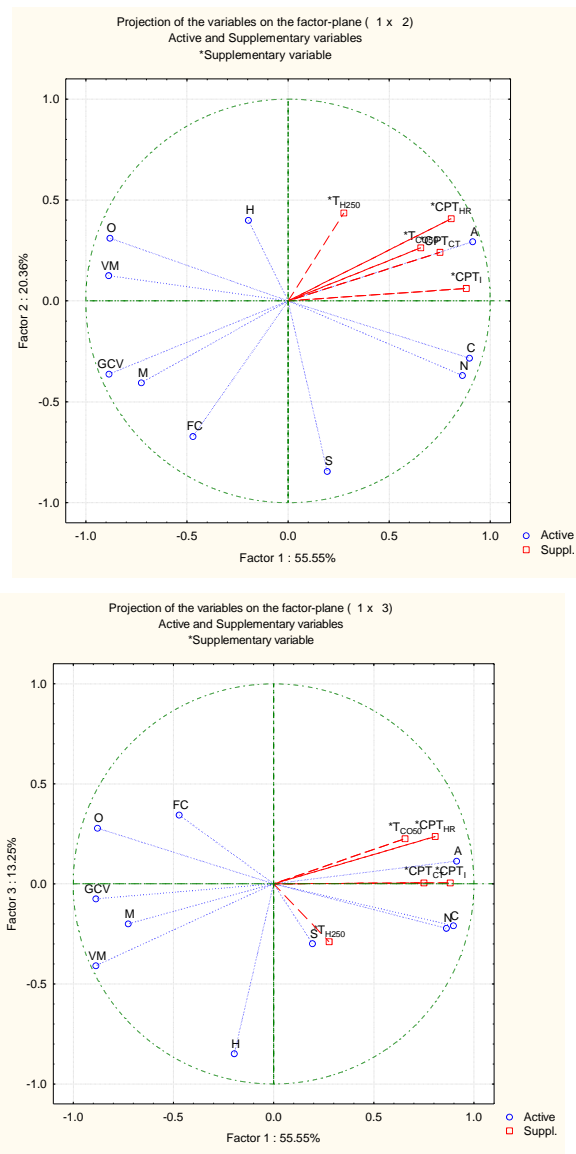


Fig. 7. Projection of variables on the factor plane (1x2) and factor plane (1x3).

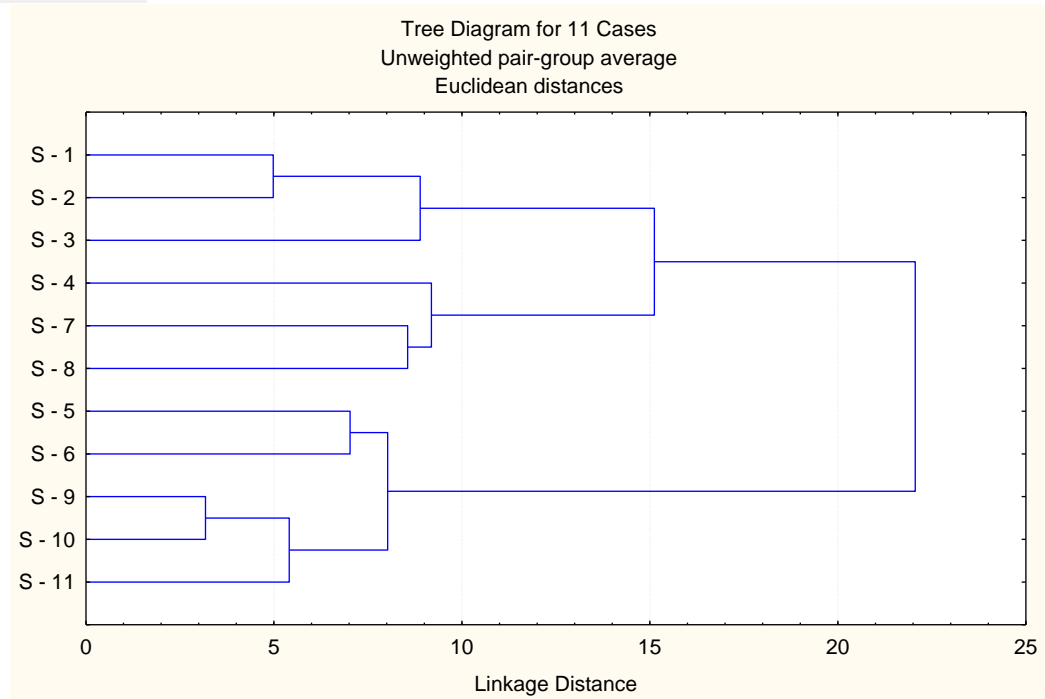


Fig. 8. Dendrograms of CPT_I (dependent variables) vs independent variables (M, VM_{daf}, FC_{daf}, C_{daf}, H_{daf}, N_{daf}, S_{daf}, O_{daf}, GCV)

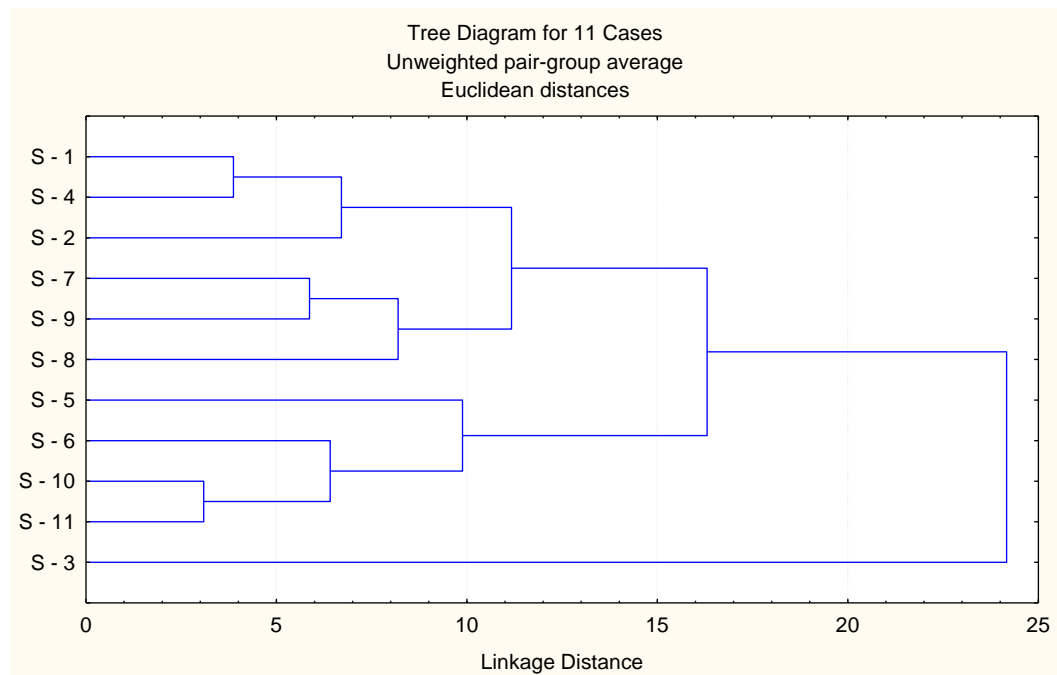


Fig. 9. Dendrograms of CPT_{CT} (dependent variables) vs independent variables (M, VM_{daf}, FC_{daf}, C_{daf}, H_{daf}, N_{daf}, S_{daf}, O_{daf}, GCV)

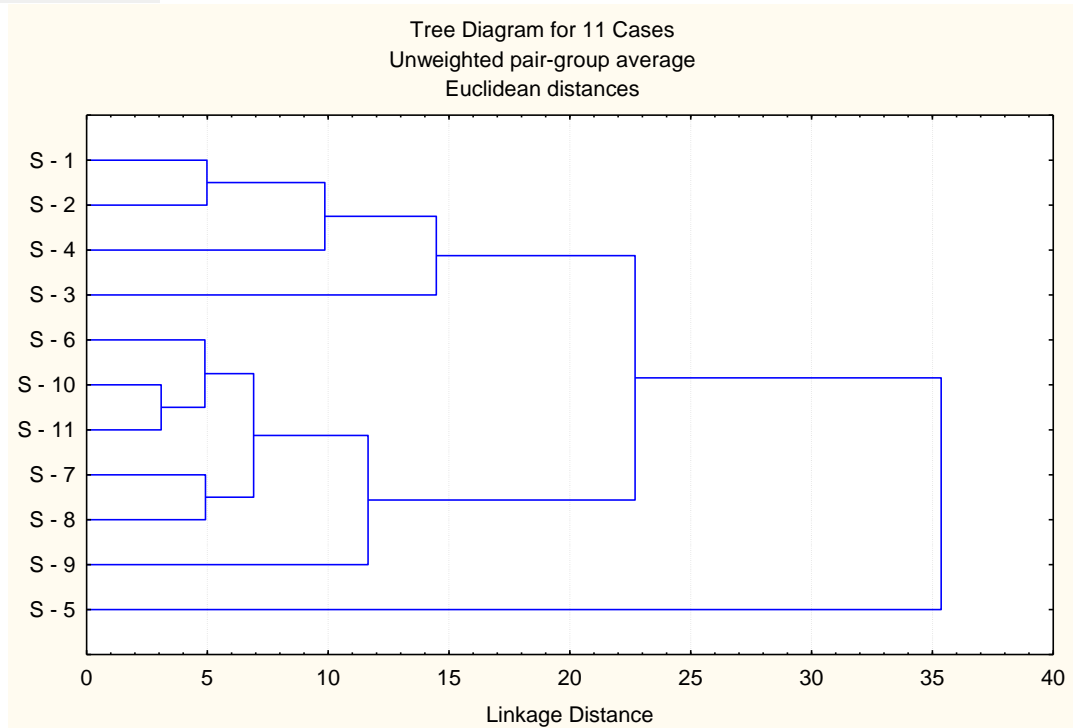


Fig. 10. Dendrograms of CPT_{HR} (dependent variables) vs independent variables (M , VM_{daf} , FC_{daf} , C_{daf} , H_{daf} , N_{daf} , S_{daf} , O_{daf} , GCV)

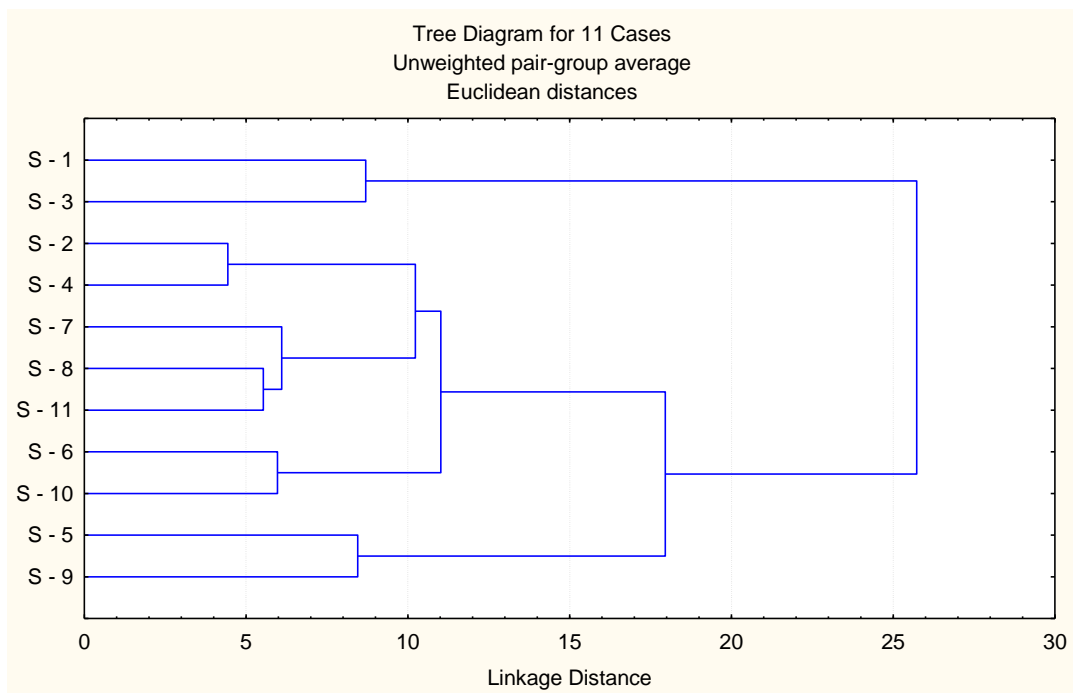


Fig. 11. Dendrograms of TCO_{50} (dependent variables) vs independent variables (M , VM_{daf} , FC_{daf} , C_{daf} , H_{daf} , N_{daf} , S_{daf} , O_{daf} , GCV)

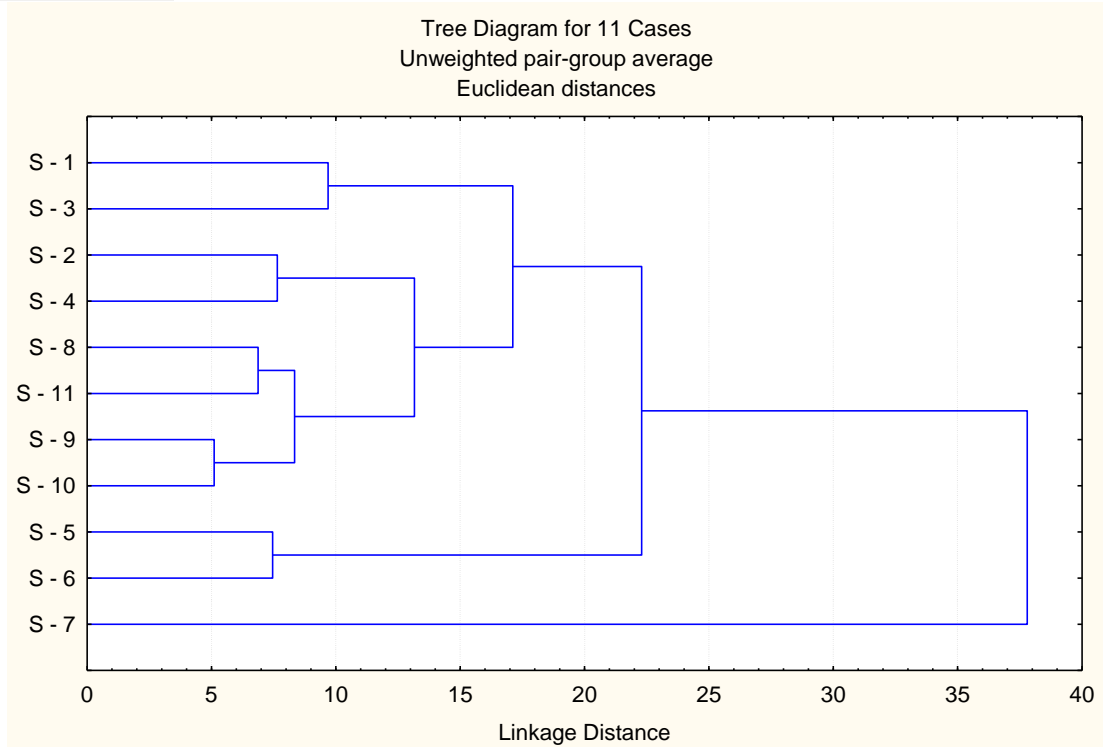


Fig. 12. Dendrograms of T_{H250} (dependent variables) vs independent variables (M , VM_{daf} , FC_{daf} , C_{daf} , H_{daf} , N_{daf} , S_{daf} , O_{daf} , GCV)

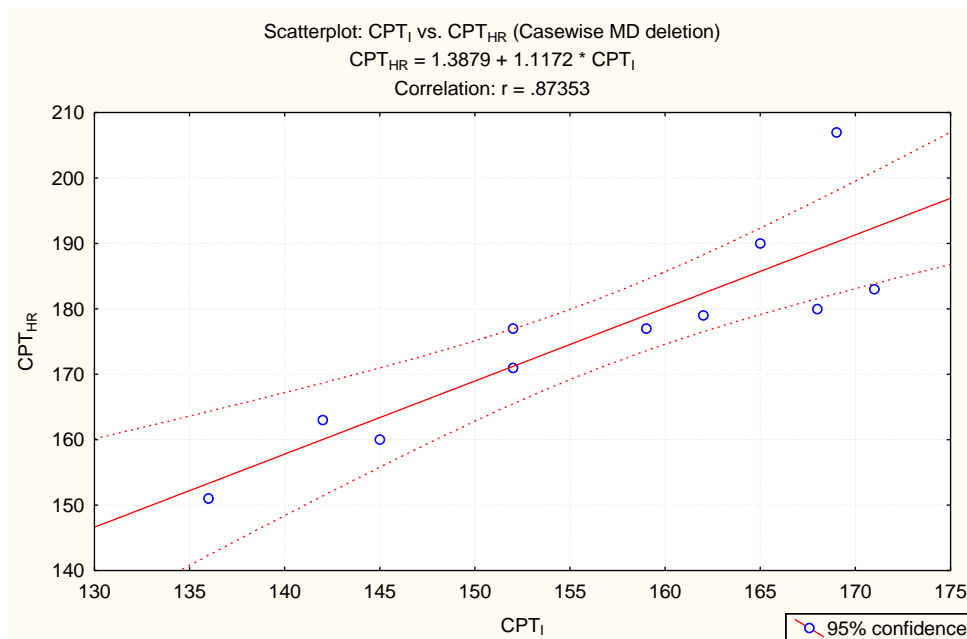


Fig. 13. Scatter plot Relationship of CPT_{HR} from sponcomg rig experiments with crossing point temperature Indian method (CPT_I).

Table 1**Proximate, ultimate, GCV and CPT analysis of the eleven coal samples**

Sample	Seam Name	Mine Name	Fire Status	Moisture (%wt)	Ash (%wt)	Volatile Matter (%wt dafb)	Fixed Carbon (%wt dafb)	C (%wt dafb)	H (%wt dafb)	N (%wt dafb)	S (%wt dafb)	O (%wt dafb)	Calorific Value (MJ/kg)	CPT _i (°C)	Vitrinite Reflectance (VR _m)
1	S-14	Chasnala	Yes	1.14	10.31	30.69	61.37	77.16	4.92	1.52	0.28	16.10	29.64	145	0.74
2	S-14	Jitpur	Yes	1.18	10.84	29.54	61.99	79.77	4.81	1.58	0.27	13.57	28.86	142	0.76
3	S-13	Chasnala	Yes	1.16	7.78	32.44	61.52	79.85	5.28	1.57	0.31	12.98	29.29	136	0.87
4	S-16	Jitpur	Yes	1.30	9.34	30.06	62.50	79.12	4.91	1.43	0.32	14.21	28.68	152	0.97
5	S-11	Enna	Yes	0.61	20.94	26.95	57.31	81.31	5.02	1.62	0.00	12.06	25.59	169	1.00
6	S-11	Bhalgora	Yes	1.22	16.71	28.26	58.88	83.96	5.06	1.88	0.54	8.57	27.42	171	1.02
7	S-11	Simlabahal	No	1.25	13.40	27.22	62.11	82.10	4.69	1.73	0.41	11.07	28.34	159	1.15
8	S-12	Simlabahal	No	1.13	16.74	26.42	60.44	79.99	4.81	1.64	0.50	13.05	27.24	152	1.08
9	S-10	Bhalgora	No	0.64	17.19	24.86	61.74	83.94	4.73	1.90	0.35	9.08	26.72	165	1.04
10	S-10	Simlabahal	No	0.63	16.75	25.08	61.90	84.14	4.95	1.90	0.39	8.63	27.46	168	1.05
11	S-09	Simlabahal	No	0.92	16.88	27.02	59.99	83.53	4.94	2.00	0.50	9.04	26.77	162	1.09

M- moisture, A –Ash, VM- Volatile matter, FC – Fixed carbon, C- Carbon, H- Hydrogen, N-Nitrogen, S- Sulphur, O- Oxygen, CV- Calorific value, CPT- Crossing point temperature of coal, V_m – Vitrinite, L_m – Liptinite, SF_m - Semi-Fusinite, F_m – Fusinite, VR_m - Vitrinite Reflectance

Table 2**Crossing point temperature from Spontaneous combustion rig experiments**

Sample No.	CPT _{CT}	CPT _{HR}	T _{CO50}	T _{H250}
1	213	160	134	176.8
2	206	163	157	194.7
3	196	151	141	185.0
4	211	171	161	202.1
5	231	207	175	208.5
6	228	183	164	211.7
7	218	177	155	157.2
8	211	177	152	190.1
9	216	190	171	191.8
10	222	180	160	196.8
11	223	179	151	185.9

Table 3

Results of the correlation study performed between the proximate, ultimate and susceptibility indices determined for the coal samples

	CPT_I	CPT_{CT}	CPT_{HR}	T_{CO50}	T_{H250}
Moisture (%wt)	-0.57	-0.48	-0.66	-0.52	-0.29
Ash (%wt)	0.86	0.83	0.92	0.67	0.36
Volatile Matter (%wt dafb)	-0.79	-0.64	-0.76	-0.62	-0.10
Fixed Carbon (%wt dafb)	-0.51	-0.66	-0.65	-0.38	-0.50
C (%wt dafb)	0.80	0.59	0.58	0.59	0.21
H (%wt dafb)	-0.19	-0.15	-0.25	-0.24	0.39
N (%wt dafb)	0.69	0.55	0.43	0.30	0.04
S (%wt dafb)	0.12	-0.02	-0.24	-0.25	-0.19
O (%wt dafb)	-0.77	-0.57	-0.53	-0.53	-0.20
Calorific Value (MJ/kg)	-0.81	-0.74	-0.93	-0.76	-0.44

Table 4

The eigenvalues of the correlation matrix derived by the PCA method, and the variance in correlation computed for the four spontaneous combustion susceptibility indices

PC	Eigenvalue	% Total variance	Cumulative %
1.	5.554690	55.54690	55.5469
2.	2.036433	20.36433	75.9112
3.	1.324608	13.24608	89.1573
4.	0.713565	7.13565	96.2930
5.	0.198211	1.98211	98.2751
6.	0.123812	1.23812	99.5132
7.	0.045121	0.45121	99.9644
8.	0.003514	0.03514	99.9995
9.	0.000047	0.00047	100.0000
10.	0.000000	0.00000	100.0000

Table 5

The computed PCCA factor loadings of the variables in the principal component matrix for the three principal components

Variables	PC1	PC2	PC3
M	-0.725	-0.406	-0.199
A	0.913	0.294	0.114
VM	-0.888	0.124	-0.409
FC	-0.471	-0.671	0.344
C	0.897	-0.284	-0.210
H	-0.197	0.401	-0.849
N	0.863	-0.370	-0.222
S	0.194	-0.844	-0.298
O	-0.880	0.311	0.279
GCV	-0.887	-0.363	-0.074
*CPTI	0.882	0.062	0.006
*CPTCT	0.751	0.240	0.006
*CPTHR	0.806	0.407	0.236
*TCO50	0.656	0.263	0.225
*TH250	0.276	0.436	-0.290

Table 6

Results of the clustering of the coal sample data sets from the dendrograms

No. of clusters	CPT _I	CPT _{CT}	CPT _{HR}	T _{CO50}	T _{H250}
Cluster 1	1,2,3	1,4,2,7,9,8	1,2,3,4	1,3	1,3,2,4,8,11,9,10
Cluster 2	4,7,8	5,6,10,11	6,10,11,7,8,9	2,4,7,8,11,6,10	5,6
Cluster 3	5,6,9,10,11,3	3	5	5,9	7

Table 7

Measure of fit of experimental data to fixed multiple nonlinear regression models

Sl. No.	Equation	R ²	Adjusted R ²	P level	Standard error of estimate
1	Proximate Analysis (M, A, VM _{daf})				
	$CPT_I = 162.09 + 1.959 * A - 1.155 * VM - \frac{1.102}{M}$	0.751	0.645	0.015	7.070

	$CPT_{HR} = 161.665 - 0.408 * VM + \frac{1.603}{M} + 0.110 * A^2$	0.867	0.81	0.001	6.618
--	-------------------------------------------------------------------	-------	------	-------	-------

Table 8**Correlation matrix of different spontaneous heating indices**

	CPT	CPT_{CT}	CPT_{HR}	T_{CO50}	T_{H250}
CPT	1.00				
CPT_{CT}	0.93	1.00			
CPT_{HR}	0.87	0.84	1.00		
T_{CO50}	0.73	0.59	0.84	1.00	
T_{H250}	0.37	0.34	0.41	0.58	1.00