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Thermal and Spectral Studies to Assess the Spontaneous Combustion Propensity of Coal Seams of Raniganj Coalfield, India

Pintu Kumar Mandal[1*], Manoj Kumar Mishra[1] and Niroj Kumar Mohalik[2]

[1]Department of Chemistry, BIT Sindri (Under Jharkhand University of Technology, Ranchi), Dhanbad Road, Sindri - 828123, Jharkhand, India; pintum.rs.che21@bitsindri.ac.in

[2]Mine Fire Ventilation Miners, Health Research Group, CSIR-CIMFR, Dhanbad - 826015, Jharkhand, India

Abstract

The extraction of coal from Raniganj Coalfield (RCF) plays a vital role in the country's industrial growth. The mechanisation of underground coal mines of RCF for excavation raises several issues in India, namely: multi-seam workings, upper seam worked-out areas, strata management, subsidence, ventilation, heat, humidity, spontaneous heating, and mine fires. This research is focused on studying the spontaneous combustion characteristics of ten RCF coals by investigating the CPT, TG_{ign} and FTIR, along with intrinsic properties of coal, viz. proximate analysis. The proximate analysis and spontaneous combustion study reveal that seven samples have optimum moisture (4-8 %), high volatile matter content (>30.0%), low CPT (<160°C) and TG_{ign} (<290°C) value. The remaining three samples have low moisture (<4.0%), low volatile matter content (<25.0%), high CPT (>180°C) and TG_{ign} (>320°C) values. FTIR study of these coals reveals that the same samples have quality organic functional groups (mainly including alcoholic group, carboxylic group, aldehyde group, ketonic group, alkene and alkyne group, etc.) which enhance the propensity of coal to spontaneous combustion. Based on all the results, it was observed that RCF coal has a critical issue of spontaneous combustion in many coal seams, which ultimately leads to coal mine fire in such cases.

Major Findings: *Coal samples of RCF with VM_d > 30%, M_{ad} range 4 to 8 %, CPT < 160 °C, TG_{ign} < 290 °C are more susceptible to spontaneous combustion. This finding is also confirmed by FTIR analysis of coal samples.*

Keywords: *Coal, CPT, FTIR, Spontaneous Combustion, TGA*

1.0 Introduction

Coal plays an important role in India's industrialisation and economic development because it is the second-largest coal consumer in the world[1]. The history of coal mining in Raniganj Coalfield is older than the 18th century, and after nationalisation, Indian coal industries are facing tremendous challenges for the safety of miners as well as mines[2]. The mounting energy and steel demand of the country, desire coal exploitation from the greater depths (>300 m), which constitutes a considerable proportion (31.89%) of the total coal reserve. Presently, India has a reserve of 324.65 billion metric tons (BT) (89.83% of non-coking coal), followed by 35.10 BT (9.71% of coking coal), spreading across two major coal-bearing regions, *i.e.*, Jharia Coalfield (JCF) and Raniganj Coalfield (RCF)[3]. India has a great coking coal reserve in the Raniganj coalfield (RCF), which plays an important role in industrialisation and energy security. The RCF covering 1900 km^2 contains several coal seams of Barakar formation (BR-I – BR-VII) and Raniganj formation (R-I – RXIII), having a trough-like semi-elliptical elongated

**Author for correspondence*

intracratonic rift basin of the half-graben shape[4]. There are 44 operative underground coal mines in Raniganj Coalfield, which are exploited through mechanisation with the help of technologies, *viz.* longwall and continuous miner[5]. However, the mechanisation of underground coal mines comes with several associated issues, namely: environmental pollution, multi-seam workings, upper seam worked-out areas, strata management, subsidence, ventilation, heat, humidity, spontaneous heating, and mine fire. This study addresses the issues of spontaneous heating and fire in underground coal mines in RCF, a significant coking coal reserve[6-8].

The spontaneous combustion of coal is influenced by a variety of intrinsic and extrinsic parameters, such as coal properties, geological factors, mining practices, and environmental conditions, due to the heterogeneous properties of coal[9-11]. Previous studies have investigated the impact of intrinsic properties of coal, including moisture, volatile matter, ash, carbon, and pyritic sulphur content, on spontaneous combustion[12,13]. Several researchers have also investigated the petrographic components, *i.e.*, vitrinite reflectance and macerals of coals for spontaneous combustion susceptibility[14,15]. Similarly, extrinsic geo-mining parameters, viz., geology of that area, faults, folds, seam thickness, fractures, methods of mining, multi-seam workings, mining of thick seams in slices, mine mechanisation, coal left in goaved-out areas, and complex ventilation networks of deep mines, also contribute to the spontaneous heating and mine fire[16]. Modern mining mechanisation has been greatly with several challenges, namely: excessive stress due to overlying strata, subsidence, ventilation anomalies, heat, humidity, multi-seam working, contiguous seams, upper seam goaved-out areas, and spontaneous heating[17]. The huge coal dust generated after mechanisation is primarily accountable for the spontaneous heating of coal because of its larger specific surface area[17-19]. The reaction between coal and oxygen is limited because the overall effective area of contact decreases intensely in coarser particles and vice versa. Smaller coal particles provide a larger surface area for oxygen diffusion, which in turn increases their spontaneous heating tendency and accelerates coal oxidation[20,21].

Researchers all over the world use a variety of large- and small-scale lab methods to study the spontaneous combustion of coals. These include the Crossing-Point Temperature (CPT) method, Differential Thermal Analysis (DTA), Thermo-Gravimetric Analysis (TGA), Differential Scanning Calorimetric (DSC), the Russian U index, the Olpinski index in Russia, and the R70 index[22-24]. However, researchers used the CPT and TG_{ign} methods to determine the spontaneous heating propensity of coals, due to their wide acceptability concerning accuracy[23-25]. Fourier Transform Infrared (FT-IR) spectroscopy is a powerful method of classifying coal by identifying the functional groups present for its characterisation. It can reveal the types and arrangements of hydrogenated carbon structure, heteroatom functional groups, and the knowledge of the cross-linked[26-28]. Therefore, this research is focused on the study of spontaneous combustion characteristics of coals in general, which is the country's primary source of prime coking coal. The present study aims to predict the best method for the spontaneous combustion propensity of RCF coal using CPT, TG_{ign} and FTIR, investigation[10,29-31].

2.0 Materials and Methods

2.1 Sample Collection and Preparation

The coal samples for this study were collected from ten different coal seams located in the RCF area. Details about these samples are provided in Table 1 and Figure 1. Out of the ten samples collected, seven are from four mines that have a documented history of spontaneous heating or fire, while the remaining three samples have not shown any signs of fire. Proximate analysis is a commonly used method for characterising coal, which measures four fundamental components: moisture, ash, volatile matter, and fixed carbon. This analysis was conducted according to the Indian Standard IS: 1350 (Part 1) – 1984[32-34]. The moisture, ash, and volatile matter contents were determined through experimental methods, whereas the fixed carbon content was calculated based on the other measured values.

2.2 Spontaneous Heating Experiments

2.2.1 Crossing Point Temperature (CPT)

The spontaneous combustion characteristics of coal were determined using CPT methods as per Director General Mine Safety (DGMS) rules and circulars[35-37]. The CPT experimental setup comprises a temperature controller, electric heater, glycerine bath, cylindrical combustion reactor tube, gas outlet tube, air motor,

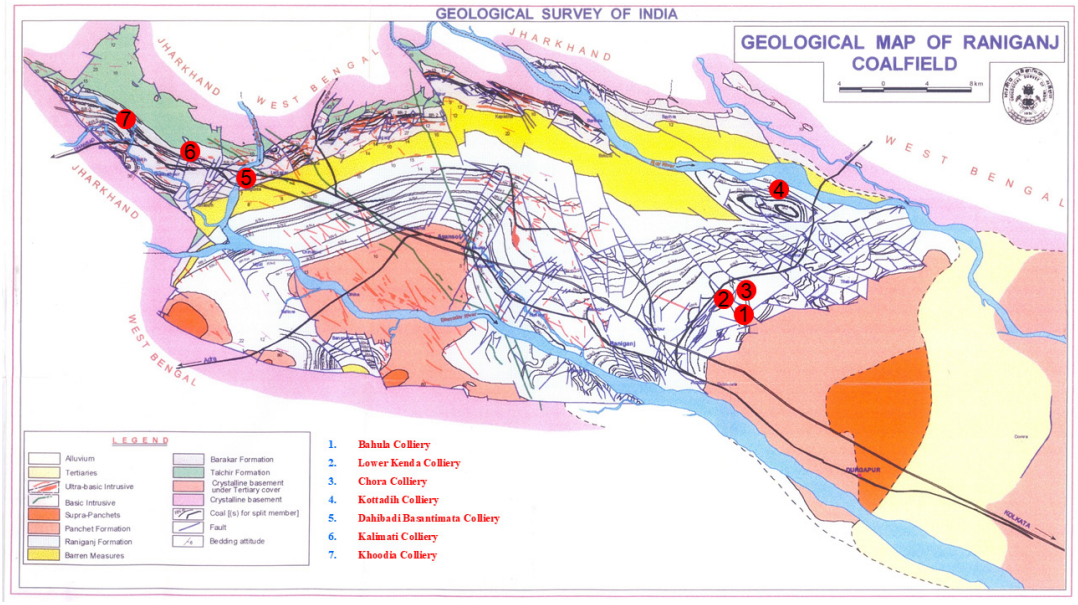


Figure 1. Sample collection locations from different mines of the Raniganj coalfield.

supply of oxygen gas, and computer-based software (Figure 2). Glass wool is placed at the bottom of the combustion tube to trap the dust and provide insulation. A 20-gram coal sample, with a particle size of (-) 212 microns, is carefully poured into the combustion tube, followed by covering the coal sample with another layer of glass wool. The coal sample is placed in a glycerine bath and heated the glycerine bath @1 °C min^{-1} with a flow rate of 80 cc/min^{-1} oxygen gas. Continuous stirring of the glycerine bath is ensured by passing air through a small air motor, maintaining a consistent temperature throughout the process. The temperatures of the glycerine bath and the coal are meticulously recorded using computer-based software equipment to determine the CPT of coal, as shown in Figure 3. The CPT values for the coal samples are determined for 10 coal samples and depicted in Table 1.

2.2.2 Thermo-gravimetric Analysis (TGA)

TGA is a well-known thermal method for studying the spontaneous heating behaviour of coal samples. The TGA experiments were performed in standard experimental conditions using a simultaneous thermal analyser (STA 443F3) instrument of M/s NETZSCH, Germany, concerning weight, temperature, and sensitivity calibration (Figure 4)[23,24]. A blank correction was implemented to reduce the buoyancy effect. The



Figure 2. Experimental set-up of a crossing point apparatus.

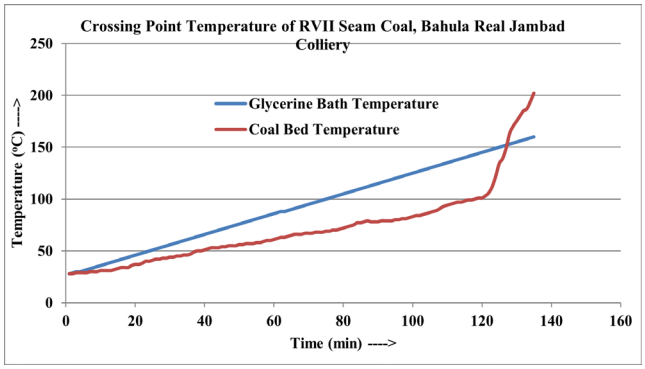


Figure 3. Crossing point temperature of Bahula Real Jambad coal sample.

combustion experiments of the coal sample were carried out keeping the mass of sample mass 10 ± 0.5 mg, particle size (–) 212 µm, heating rate @5°C.min^{-1}, in a



Figure 4. Experimental set-up of a simultaneous thermal analyser.

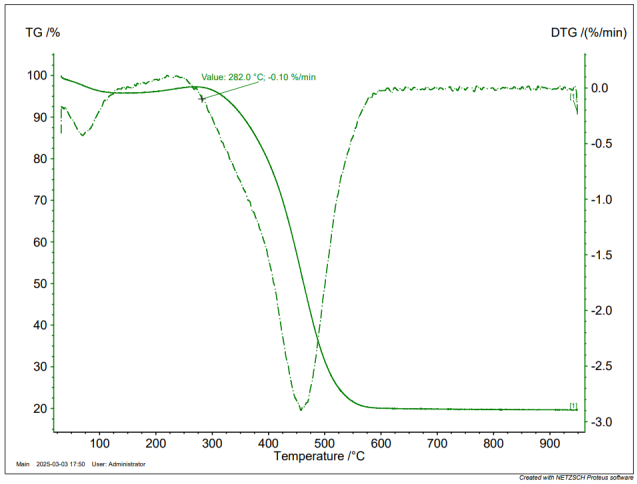


Figure 5. TGA/DSC curve of a coal sample of Bahula Real Jambad Colliery for TG_{ign} using a simultaneous thermal analyser.

normal air atmosphere at a flow rate of 100 ml/min^{-1}, temperature range 30-750 °C, for all samples as shown in Figure 5. The experiments were repeated two times for the repeatability of this study. A total of 20 experiments, including repeatability, were performed for the ten coal samples from the RCF to determine their spontaneous heating behaviour, and their results are presented in Table 1.

2.2.3 Fourier Transforms Infrared Spectroscopy (FT-IR)

FT-IR spectroscopy has been used to identify the functional groups present in the coal, as it is a heterogeneous compound. The IR portion of the electromagnetic spectrum lies between visible light and microwave range, which is divided into three regions: the near IR (13000-4000 cm^{-1}), mid IR (4000-400 cm^{-1}) and far IR (400-10 cm^{-1})[30,32,33]. As most organic functional groups (OH, NH_2, C=O, C-H, *etc.*) absorptions occur in the mid-IR range (4000 - 400 cm^{-1})[34-37]. The mid-infrared spectral signals of coal samples were collected using Frontier FT-IR equipment of M/s Perkin Elmer (Figure 6). Approximately 2 mg of the coal sample was taken in a sample holder and 20 scans through detectors in the wave number regions from 4000 to 400 cm^{-1} with a spectral resolution of 8 cm^{-1}. The absorption bands in the spectrum of each sample were identified with their matching functional groups with reference to the standard, as shown in Figure 7. The FT-IR spectra for the coal samples are determined for 10 coal samples and depicted in Table 2.

Table 1. Proximate analysis, CPT and TG_{ign} of all collected coal samples of RCF

S. No.	Sample Code	Seam Name	Colliery Name	$\%M_{ad}$	$\%VM_d$	$\%A_d$	$\%FC_d$	CPT (°C)	TG_{ign} (°C)
1	BR6	RVII	Bahula Colliery	4.24	30.40	20.48	49.12	152.0	282.0
2	LK5	R-V	Lower Kenda Colliery	3.26	37.01	17.83	45.16	153.0	282.5
3	TR6	R-VI	Chora Colliery 10 Pit	4.76	32.26	18.12	49.62	154.0	280.0
4	NR6	R-VI	Chora Colliery 7/9 Pit	4.32	36.60	21.16	42.23	157.0	283.5
5	KR4	R-IV	Kottadih Colliery	5.99	40.35	16.33	43.33	166.0	276.0
6	KR5	R-V	Kottadih Colliery	6.06	39.12	16.10	44.78	154.0	278.5
7	KR6	R-VI	Kottadih Colliery	7.06	40.92	16.71	42.37	161.0	277.0
8	DGP	GP	Dahibadi Basantimata Colliery	1.03	23.29	32.94	43.77	188.0	328.0
9	KKM	KM	Kalimati Colliery	2.28	23.39	22.62	53.99	185.0	332.0
10	KBP	BP	Khoodia Colliery	0.74	17.86	43.67	38.46	215.0	329.0

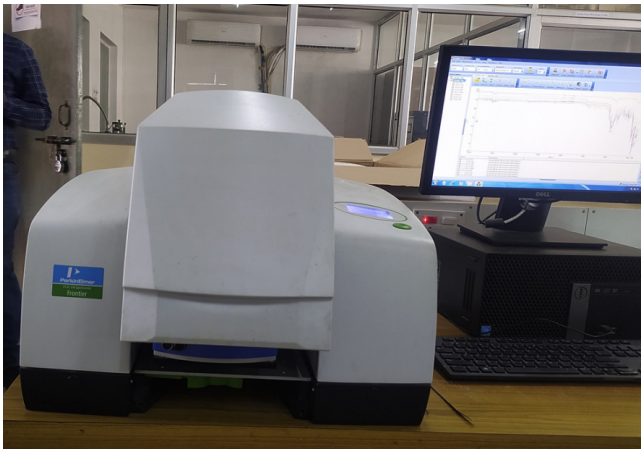


Figure 6. Experimental setup of FTIR equipment for functional group analysis.

3.0 Result and Discussion

The proximate analyses of different coal samples of RCF reveal that moisture (M_{ad}) varies between 1.60-7.06 %, VM_d content varies between 17.73-38.03 % and ash content varies between 15.12-43.35 % respectively. Proximate data reveal that the coal of RCF, which is rich in volatile matter and medium moisture, is easily combustible. Out of ten samples, seven samples M_{ad} varied from 4.0-8.0 % and VM_d content varied from 30.0-40.0 % which are prone to spontaneous heating, as compared to the other three samples. The CPT values vary across a range between 152°C to 215°C. The CPT values determined for four samples are low (<160 °C), which is categorised as moderately prone to spontaneous combustion, whereas three samples are in the range of 160 to 180°C, which is less susceptible. Out of ten samples remaining three samples have a high CPT value >180 °C, which denotes very low susceptibility to spontaneous combustion. The TG_{ign} values vary across a range between 276°C to 340°C. The TG_{ign} values determined for seven samples are low (<290 °C), which is categorised as highly prone to spontaneous combustion, whereas three samples have high TG_{ign} values>320 °C, which denotes poor susceptibility to spontaneous combustion. The correlation study was carried out between all parameters to determine the relationship between them (Table 3). It reveals that M_{ad}, VM_d has a strong linear relationship (r >0.75) between CPT and TG_{ign}. A 3D scatter plot was presented to know the relationship between M_{ad}, VM_d, CPT (Figure 8(a)) and Mad, VM_d, TG_{ign} (Figure 8(b)). Feature selection involves identifying and selecting the most relevant features (or predictors) from a dataset for model construction. The feature selection was performed with the six independent (M_{ad}, VM_d, A_d, FC_d) and two dependent (CPT and TG_{ign}) variables. The results of the best significant features (F>1) for both CPT and TG_{ign} are depicted in Figure 9, indicating the importance of the independent variables. Accordingly, a polynomial regression analysis was carried out between Mad with CPT, TG_{ign}, and observed that the polynomial fit has R^2 values of 0.8652 and 0.8827 with p<0.05(Figure 10(a)). Similarly, the correlation study among VM_d vs CPT,

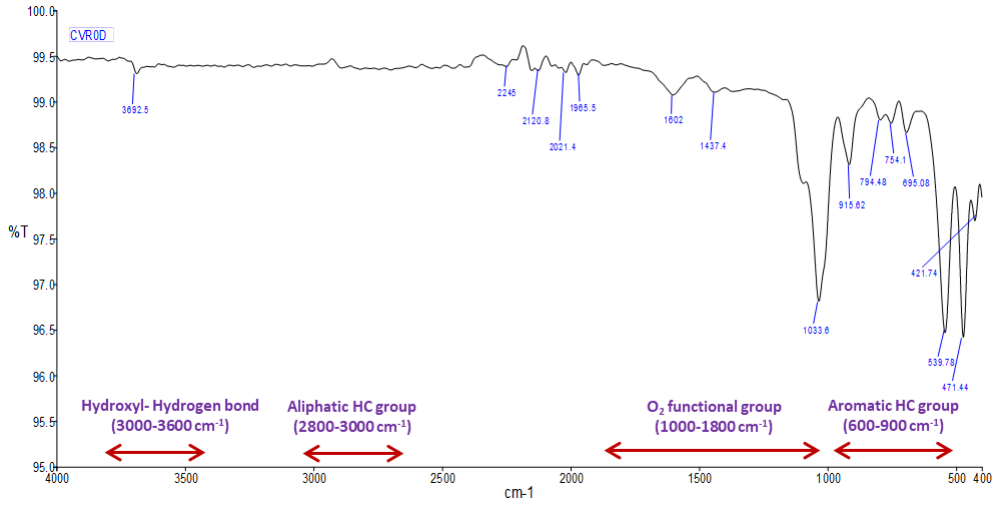


Figure 7. FTIR experiment results of a coal sample of Bahula Real Jambad Colliery.

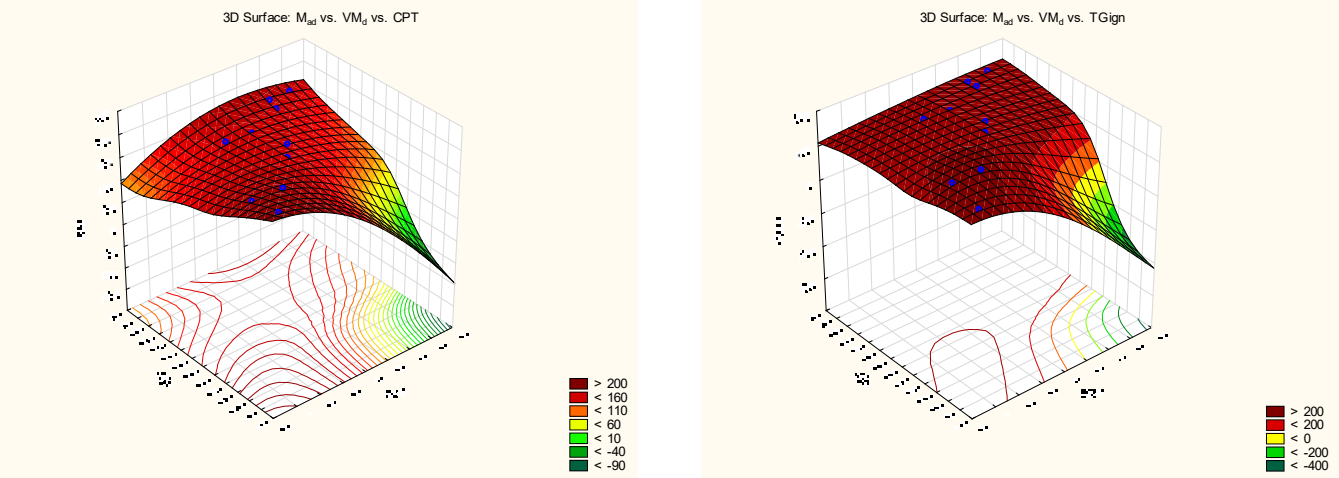
Table 2. FTIR experiment results (functional group analysis) of all studied coal samples

Bahula RVII		Lower Kenda RV		Chora 10 pit RVI		Chora 7-9 pit RVI		Kottadih RIV		Kottadih RV		Kottadih RVI		Dahibadi GP		Kalimati KM		Khoodia BP	
BR6		LK5		TR6		NR6		KR4		KR5		KR6		DGP		KKM		KBP	
WN	FGP	WN	FGP	WN	FGP	WN	FGP	WN	FGP	WN	FGP	WN	FGP	WN	FGP	WN	FGP	WN	FGP
3692.5	-OH	3689.4	-OH	3692.5	-OH	3695.6	-OH	3692.4	-OH	3692.5	-OH	3686.3	-OH	3692.5	-OH	3754.6	-OH	3683.2	-OH
3618	-OH	2922.2	MG	3618	-OH	3618	-OH	1595.9	-COOM	3618	-OH	3618	-OH	2245	-C≡C-	3686.3	-OH	3611.7	-OH
1599	-COOM	1735.6	-CHO	1592.8	-COOM	1595.9	-COOM	1437.4	-COOM	1741.8	-HC=O	1599	-COOM	2120.8	-C≡C-	1592.8	Aromatic	1729.4	-CHO
1437.4	-COOM	1595.9	-COOM	1443.7	-COOM	1437.4	-COOM	1095.8	Amine	1592.8	-COOM	1440.6	-COOM	2021.4	-CONH2	1437.4	MG	1595.9	-COOM
1372.2	-COOM	1440.6	-COOM	1362.9	-COOM	1102	C6H6/ Si-O-C	1024.3	1°-OH	1431.2	-COOM	1098.9	3°amine/1°-OH	1985.5	-CONH2	1092.7	Amine	1434.3	-COOM
1167.2	3°amine/1°OH	1372.2	-(CH3)2	1095.8	3°amine/ 1°OH	1027.4	C6H6/ Si-O-C	915.62	CH-C=C-	1366	-COOM	1027.4	F-/1° -OH	1602	AC	1030.5	1°-OH	1015	AC
1089.6	F-/1° -OH	1098.9	-C-OH	1030.5	F-/1° -OH	912.51	C6H6/Si-O-C	751	-OH	1210.7	3°-amine	918.72	CH-C≡C-	1437.4	MG	1008.8	1°-OH	909.4	AC
1031.97	F-/1° -OH	1033.6	-C-OH	1008.8	F-/1° -OH	797.58	-CH2-	667.13	-C≡C-	1095.8	Si-O-C	791.37	CH-C=C/Cl-	1033.6	1°-OH	909.4	CH-C=C-	794.48	-C-H
1005.7	F-/1° -OH	1012	-C-OH	912.51	-CH-C≡C-	688.87	MG	533.56	2°OH/Ar	1027.4	Si-O-C	751	Ar-H	915.62	CH-C=C-	865.95	CH-C=C-	747.89	-OH
913.54	CH-C≡C-	902.4	amine	797.58	-CH-C=C-	539.78	CS2	471.44	2°OH/Ar	999.48	Si-O-C	691.98	CH-C≡C-	794.48	-CH2-	794.48	-CH2-	682.66	-C≡C-
800.7	CH-C=C/Cl-	747.89	AC	747.89	AC	468.33	CS2			912.51	Si-O-C	542.88	2°OH, Ar-S	754.1	-C=C-	747.89	-C=C-	530.46	AC
747.89	AC	691.98	CH-C≡C-	688.87	-C≡C-	427.96	AC			747.89	-OH	468.33	2°OH, Ar-S	695.08	-C≡C-	691.98	-C≡C-	462.12	AC
691.98	H-C≡C-H	533.56	2°OH, Ar-S	657.8	-C≡C-					695.08	-C≡C-	431.06	2°OH, Ar-S	539.78	AC	530.46	AC		
664.02	H-C≡C-H	465.23	2°OH, Ar-S	533.56	AC					530.46	AC			471.44	AC	468.33	AC		
536.04	AC			465.23	AC					468.33	AC			421.74	AC	431.06	AC		
468.54	AC			421.74	AC					424.85	AC								
427.96	AC			400	AC														

AC-Aromatic compound, MG-Methyl group

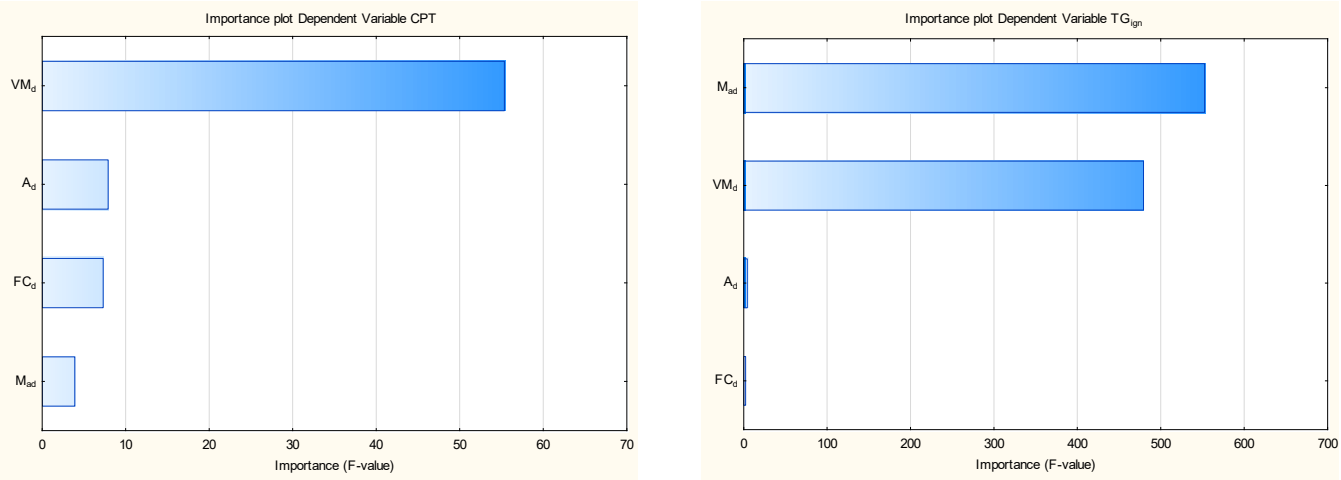
Table 3. Correlation colour map results of all studied coal samples among all parameters

Variable	Colour map of correlations (Spreadsheet1) N=10 r>= -1 -0.80 -0.60 -0.40 -0.20 0 0.20 0.40 0.60 0.80 1-1					
	M_{ad}	VM_d	A_d	FC_d	CPT	TG_{ign}
Mad	1.000000					
VMd	0.909715	1.000000				
Ad	-0.838562	-0.866685	1.000000			
FCd	0.014563	-0.093978	-0.415199	1.000000		
CPT	-0.751008	-0.825592	0.908091	-0.306516	1.000000	
TGign	-0.877184	-0.914547	0.804926	0.061482	0.882160	1.00000.



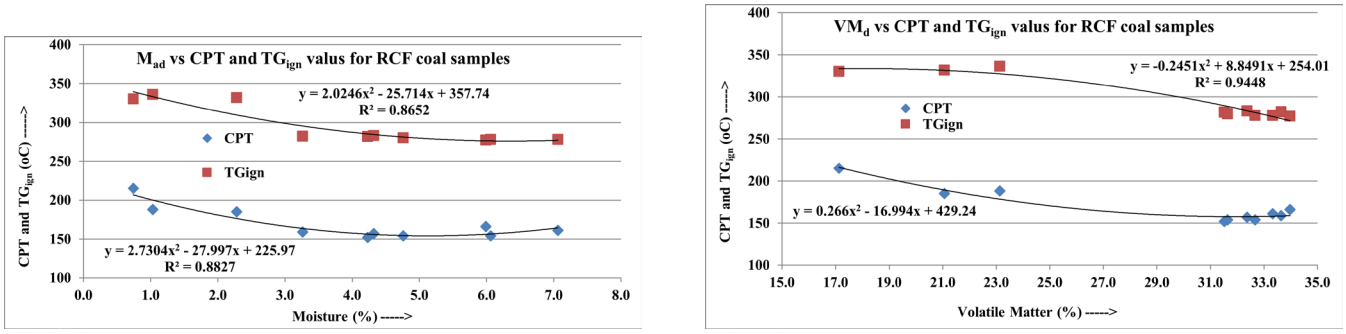
(a) **(b)**

Figure 8. 3D surface plot results of all coal samples **(a)** M_{ad} vs VM_d vs CPT, **(b)** M_{ad} vs VM_d vs TG_{ign}.



(a) **(b)**

Figure 9. Variable Importance plot results of all coal samples **(a)** CPT, **(b)** TG_{ign}.



(a) **(b)**

Figure 10. **(a)** Relationship between moisture (M_{ad}) vs CPT, TG_{ign}, **(b)** Relationship between volatile matter (VM_d) vs CPT, TG_{ign}.

and TG_{ign} was carried out to determine the relationship between them (Figure 10(b)). It reveals that there is a strong polynomial fit having R^2 values of 0.9448 and 0.9383 with $p<0.05$.

FTIR analysis of different coal samples of RCF by different wave numbers was measured to determine various functional groups, *i.e.*, hydroxyl-hydrogen bond, aliphatic hydrocarbon group, oxygen-containing functional group, aromatic hydrocarbon group present in coal samples. The FTIR spectrum depicts that the –OH group (3692 cm^{-1}), –C≡C– group (2445 cm^{-1}), amides (2021 cm^{-1}), aldehydes (1741 cm^{-1}), aromatic compounds (1602 cm^{-1}), metal acetates (1592 cm^{-1}), methyl group (1447 cm^{-1}), 3° amines (1210 cm^{-1}), 1° alcohol (1043 cm^{-1}), organic silicates is (999 cm^{-1}), conjugated system (915 cm^{-1}), alkenes (754 cm^{-1}), and several other functional groups also present. The FTIR spectroscopy results (functional groups) for all coal samples are shown in Table 2 and Figure 11. FTIR study reveals that the coal samples of RCF are rich with organic compounds having both aromatic hydrocarbons and oxygen-containing functional groups. The aromatic hydrocarbon groups also contain heteroatoms of sulphur, which is highly reactive at low temperatures, leading to the spontaneous combustion of coal. The oxygen-containing functional groups contain a hydroxyl group (OH) in the range 950-1300 cm^{-1} and carbonyl (COO) with aldehyde (CHO) group in the range of 1300-1800 cm^{-1}. Out of these, carbonyl (COO) and aldehyde (CHO) groups are highly unstable due to exothermic reactions, which may be responsible for spontaneous combustion at lower temperatures. Three forms of hydroxyl functional group found in FTIR spectra analysis were free –OH group, intermolecular associated –OH group and self-associated –OH group having infrared vibration peak in the range of 3692 - 3618 cm^{-1}.

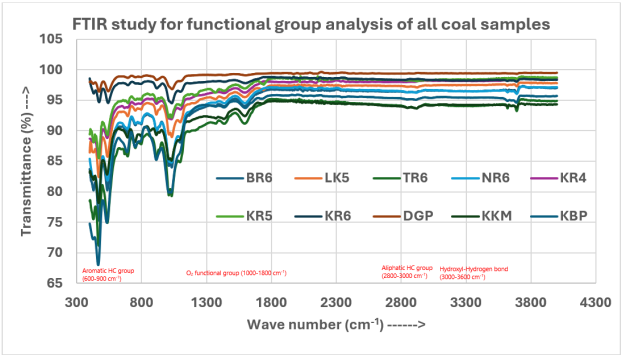


Figure 11. FTIR spectrum of 10 coal samples (Transmittance v/s Wave number).

4.0 Conclusion

The comprehensive study conducted on ten coal samples from the RCF reveals important insights into their spontaneous combustion behaviour. Based on proximate analysis, Crossing Point Temperature (CPT), Thermo-Gravimetric Analysis (TG_{ign}), and Fourier Transform Infrared Spectroscopy (FTIR), the key conclusions drawn are mentioned:

(i) *Susceptibility to Spontaneous Combustion*: The coal samples with higher volatile matter (VM_d) and moderate moisture content (M_{ad}) exhibited lower CPT<160°C and TG_{ign} values <290°C, indicating a greater propensity for spontaneous heating. Specifically, seven out of ten samples showed VM_d >30% and M_{ad} between 4-8%, correlating with higher spontaneous combustion risk.

(ii) *Thermal Behaviour*: The CPT values ranged from 152°C to 215°C, while TG_{ign} values ranged from 276°C to 340°C. Coal samples with CPT <160°C and TG_{ign} <290°C were identified as highly prone to spontaneous combustion, while samples with CPT >180 °C and TG_{ign} >320°C showed very low susceptibility.

(iii) *Statistical Correlation*: Correlation and regression analyses demonstrated strong negative correlations between CPT/TG_{ign} and VM_d/M_{ad} ($r>0.75$), and a positive correlation with ash content (A_d). Polynomial regression models between CPT/TG_{ign} and M_{ad}/VM_d yielded high coefficients of determination ($R^2>0.86$), reinforcing their predictive value in assessing combustion risk.

(iv) *Functional Group Analysis via FTIR*: FTIR spectra identified the presence of various reactive functional groups such as hydroxyl (–OH), carbonyl (COO), aldehydes (–CHO), and aromatic hydrocarbons. These groups, particularly the oxygen-containing and sulfur-substituted aromatic compounds, contribute significantly to exothermic reactions at low temperatures, thereby enhancing the risk of spontaneous combustion at relatively lower temperatures.

(v) *Key Chemical Indicators*: The presence of free and associated hydroxyl groups (3618–3692 cm^{-1}) and unstable carbonyl and aldehyde groups (1300–1800 cm^{-1}) is are critical chemical marker for predicting spontaneous combustion potential in RCF coals.

5.0 Acknowledgement

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6.0 Author Contributions

All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication.

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