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Geochemical investigation of hydrocarbon generation potential of coal from Raniganj Basin, India

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Abstract

Methane content in a coal seam is a necessary parameter for evaluating coal bed gas, and it poses an environmental risk to underground coal mining activities. Keeping in pace with comprehensive studies of coal bed gas, 12 coal samples were selected from the Sitarampur block of Raniganj Coalfield for analysis. The Petrographic examination illustrated that significant values of reactive macerals present in samples demonstrate that organic matter is dominated by the prominent source of aromatic hydrocarbons with a minor proportion of aliphatic hydrocarbon, which falls in the region of (Type III) kerogen, confirms the suitability for the potential of hydrocarbon generation. "A" factor (aliphatic/aromatic bands) and "C" factor (carbonyl/carboxyl bands) value concluded that the sample has the lowest aromaticity and the highest hydrocarbon-generating potential, which was also validated by the Van Krevelen diagram. The Van Krevelen diagram plots between the H/C and O/C ratio indicate that coal samples lie in the type III kerogen, and bituminous coal (gas prone zone) is present in the block, which is confirmed by the cross-plot between desorbed and total gas (cc/g). The in situ gas content values are high enough to produce methane from coal beds. The overall study concludes that the Sitarampur block from Raniganj Coalfield is suitable for hydrocarbon generation.

Keywords Hydrocarbon generation potential \cdot Petrographic study \cdot *In situ* gas content \cdot Oil yield \cdot Kerogen type \cdot Fourier Transform Infrared spectrometry analysis

Introduction

Natural gas discoveries have been made in many basins around the world, and governments are paying close attention and focusing on exploration (Abudeif et al. 2016; Attia et al. 2015; Radwan et al. 2021). Natural gas has become a promising substitute for coal and plays a significant role in

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power generation and other alternative sources of energy in different applications in India (CEA 2019). The demand for natural gas in India is steadily increasing over the past few decades from 49.10 to 59.70 billion cubic feet (BP 2020). In India, natural gas consumption grew continuously, with an annual rate of 6.2% between 2000 and 2019 (MPNG 2020). In the last few decades, natural gas production from coal beds has experienced rapid development in many countries such as the USA, Australia, China, India, and Canada (Panwar et al. 2016a, 2016b, 2020). When the methane is recovered simultaneously with coal during underground mining of coal, it is known as coal mine methane (CMM). Coal mine methane mining is done to reduce methane concentration in the mines and working areas around the mines for its effective utilization of coal and CMM. Methane is the principal gas in CMM with varying concentrations from 25-60% (Karacan 2009) and must be released or vented for safety reasons. So, CMM is an unconventional form of natural gas found in association with coal seams, and it is a clean burning fuel with a good heating value > 8500 kcal/ Kg. (Ojha et al. 2013; Panwar et al. 2017a, 2017b, 2017c).



The literature reveals that coal is a sedimentary rock, with complex pore and fracture structure (Gao et al. 2014). The macro-micro pores in coal provide extremely massive surface areas for methane, and it accumulates as a free, adsorbed, and dissolved state (Kumar et al. 2018; Wang et al. 2020). The different ranks of coal, mineral matter and maceral composition significantly influence the methane sorption capacity (Goraya et al. 2019; Moore et al. 2014). The literature reveals that Fourier transform infrared spectroscopy (FTIR) is extensively employed to characterize various source rocks to determine hydrocarbon generation potential (Ganz and Kalkreuth 1991; Mishra et al. 2018; Varma et al. 2018). The FE-SEM and EDX analysis were significantly used for organic matter association with mineral matter and coal surface structure.

In the current investigation, the author's tried to analyze the petrographic composition, geochemical characteristics, organic richness, oil generation potential, thermal maturity, liquefaction behavior, and hydrocarbon generation potential for Gondwana coals belonging to Barakar formations in Raniganj Basin, which is the primarily targeted basin for hydrocarbon productions (CBM) in India. This investigation may be serving as a guide for future deep hydrocarbon resource evaluation.



)	Age	Group	Formation							
	Recent, Sub Recent & Quaternary		Alluvia Soil, Clays, loose sand etc.							
		Unconforr	nity							
	Jurassic	Intrusives (Dykes& Sills)	Dolerite, Mica-peridotite & Lamprophyres							
	Middle/Upper Triassic	Supra-Panchet	Coarse grained quartzose sandstone with bands of dark red silty shale							
	Unconformity									
	Lower Triassic	Panchet	Coarse grained, grey sandstone, brownish to reddish clay and khaki green fissile shale							
	Upper Permian	Raniganj	Fine to coarse grained, micaceous sandstones, shale and coal seams Dark grey to brown, laminated fissile shales, shale and sandstone alternation and clays with sideritic nodules and bands Coarse grained felspatic sandstone, shales, carbonaceous shales and coal seams							
	Middle Permian	Barren Measures								
	Lower Permian	Barakars								
	Upper carbonaceous	Talchirs	Greenish sandstone and needle shales							
		Unconfor	mity							
	Pre- Cambrian	Archaeans	Gneisses and schists with quartz veins							

Fig. 1 Sitarampur block location in map

Methods of experiment and analysis

A total of 12 coal core samples were collected from one borehole during the exploratory drilling in different five coal seams present in the Sitarampur block. The Sitarampur Coal Block is situated in Raniganj coalfield in Burdwan District, West Bengal, India, having coordinates with Latitude N 23° 43′ 25′′ to N 23° 45′ 28.11′′ and Longitude E 86° 51′ 23′′ to E 86° 53′ 28.16′′ as shown in Fig. 1 along with geological setting such as age, group and formation. However, the methodology adopted and equipment used for the characterization of the sample are shown in Table 1.

Proximate analysis

Proximate analysis was carried out in the IIT (ISM), Dhanbad, by following the standard procedure of the Bureau of Indian Standard [BIS Standard: 1350, Part-1, 2003] were used. For analysis, -212μ size of coal was prepared, and 1 g of coal was taken for each analysis, and it was used for the determination of ash content, moisture, and volatile matter of coals.
 Table 1
 Tests adopted for coal sample characterization

Name of experiment	Equipment	Size of sample	Institute
Proximate analysis	Oven for Moisture and Muffle furnace for ash and volatile content measurement	-72 BSS mesh	IIT (ISM) Dhanbad
Ultimate analysis	Vario Macro Cube CHNS analyzer	-72 BSS mesh	IIT (ISM) Dhanbad
SEM-EDX	FE-SEM Supra 55 (Carl Zeiss, Germany)	-72 BSS mesh	IIT (ISM) Dhanbad
FTIR spectroscopy	Perkin Elmer Spectra 2, USA	-72 BSS mesh	IIT (ISM) Dhanbad
Petrographic analysis	Petrographic microscope	_	IIT (ISM) Dhanbad

Ultimate analysis

The elemental analysis of coal core samples collected from different seams of the Sitarampur block of Raniganj coalfield was analyzed by the elemental analyzer (Elementar Vario EL III-CHNS analyzer) in the IIT (ISM), Dhanbad. For analysis, -212μ size of coal was prepared, and 1 g of coal was taken for the analysis of C, H, N, and S.

Petrographic analysis

Coal petrography for collected coal samples were carried out at the Department of Applied Geology, IIT (ISM), Dhanbad. Maceral groups were identified. Subsequently, estimation of vitrinite reflectance (Goraya et al.) has been done in oil medium on polished coal surface in reflectance with white light and fluorescence illumination under an advanced research polarizing microscope using Leica DMRXP-HC setup. Besides this, the Bureau of Indian Standards was followed to prepare the sample [BIS No 9127 part 2 for preparation, grinding and polishing of pellet, part 3 for maceral and mineral identification and part 5 for VRo measurement].

Fourier Transform Infrared spectrometry (FTIR) analysis

For FTIR spectroscopy, samples were prepared in the form of pellets, following the procedures outlined by Painter et al. (Painter et al. 1981). Dried pellets were used to reduce the contribution of water over the spectrum. FTIR of the coal samples was carried out in a wavelength frequency range of 4000 cm^{-1} to 200 cm^{-1} in absorbance mode. A 3000 Hyperion Microscope with Vertex 80 made by Bruker optics, Germany, was used for FTIR analysis.

Measurement of coal seam gas content

Direct method

There are mainly three steps involved in measuring the in situ gas content of coal core samples retrieved from exploratory boreholes. Lost gas denoted by Q_1 is the amount of gas lost

by the core sample since its extraction from undisturbed coal seam to its confinement in the desorption canister. The cumulative value obtained by desorbed gas volume measurement gave desorbed gas (Q_2) for the coal samples. From the measured volume of gas obtained after crushing a portion of the samples, the residual gas (Q_3) was calculated for the total weight of the samples. The total volume of gas obtained by the addition of Q_1 , Q_2 , and Q_3 is divided by the total weight of the sample.

$$\mathbf{Q} = \mathbf{Q}_1 + \mathbf{Q}_2 + \mathbf{Q}_3$$

Result and discussion

Hydrocarbon generation potential

The petrographic study is predominantly used for the determination of kerogen type and its suitability for hydrocarbon generation. In previous studies, many researchers reveal that Vitrinite and liptinite macerals are generally used as an indicator for the determination of hydrocarbon generation potential (Akanksha et al. 2020; Panwar et al. 2020; Tissot and Welte 1984). The hydrocarbon generation potential of the Sitarampur block was assessed using petrographic data. The samples have significant values of reactive macerals and liptinite content in the range of 67.57 to 81.10 vol.% and 5.24 to 10.05 vol.%, respectively, making them suitable for hydrocarbon generation (Akanksha et al. 2017; Kotarba et al. 2002; Varma et al. 2019). The Petrographic examination illustrated that type III kerogen present in studied samples, as shown in Fig. 2. The ternary plot of petrographic constituents indicates that samples are vitrinite ample and mature. It was also deduced from the study that there is also a possibility of excellent hydrocarbon (methane) generation potential in coal, as shown in Fig. 3.

Empirical equations are given by Guyot (Guyot 1978; Jin and Shi 1997) used for assessment of the conversion of coal into oil and oil yield.

$$RF = \frac{1000R_{max}}{RM}$$
(1)

Conversion(%) = 0.2RM + 76.6

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(2)



Fig. 2 Ternary diagram of maceral composition indicating kerogen types of samples

$$Oilyield(\%) = 0.22RM + 44.8$$
 (3)

where R_{max} indicates huminite maximum reflectance, RM indicate reactive macerals.

The coal samples demonstrate prominent values of conversion (%) and oil yield (%) in the range of 90.11 to 92.82 vol.% and 59.67 to 62.64 vol.%, respectively. It was also depicted from the study that emphasis correlations were



Fig. 3 Hydrocarbon potential based on petrographic components for representative samples

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90.0 90.5 91.0 91.5 92.0 92.5 93.0 Conversion (%)

Fig. 4 Correlation between conversion (%) and vitrinite (%) for studied samples

found in conversion (%) with vitrinite (%) and oil yield (%), as illustrated in Figs. 4 and 5.

Petrographic investigations

76

74

72 70

Vitrinite (%)

 $R^2 = 0.86$

The petrographic analysis indicates the quantity of organic content, maturity, gas generation, and storage capacity in coal (Goraya et al. 2019; Kumar et al. 2018). It was also significantly utilized to determine coal formation and diagenesis (Gao et al. 2014; Moore 2012). The macroscopic petrography of the coal seams is mainly bright coal with black color, obvious black streaks (Gao et al. 2014; Moore 2012). The studied samples are mainly bright, vitreous luster and black in color, amorphous in nature, and fine-grained. The quantitative distribution of different macerals like vitrinite, liptinite, and inertinite is shown in Table 2. The microphotographs of various macerals and their associated subgroups are illustrated in Fig. 6.

A ternary facies of various macerals and mineral matter content was plotted. The studied samples mostly fall in the E field. The ternary diagram also exemplifies that they were deposited in alternating oxic (dry) moor with sudden high flooding during peat accumulation, as shown in Fig. 7 (Esen et al. 2020; Kumar et al. 2018). The vitrinite groups of macerals are dominantly present in samples, primarily derived from woody plant tissue, and its value varies from 59.47 to 73.39 vol.% (averaging 66.43%) on a dry ash-free basis (Varma et al. 2015a; Varma et al. 2015b). The liptinite is generally derived from waxy or resinous plant parts, and its value varies in samples from 5.24 to 10.05 vol.% (averaging 7.64%) on a dry ash-free basis. The carbon-rich inertinite groups are found in moderate



Fig. 5 Correlation between conversion and oil yield for studied samples

quantity, and their values ranging from 16.18 to 35.31 vol.% (averaging 25.74%) on a dry ash-free basis. The moderate value of inertinite content indicates that studied samples were formed under an exposed peat swamps environment (Fu et al. 2016; Wang et al. 2020). It was observed in several studies that the depth, organic matter, and rank enhance the gas potential of coal seams. The vitrinite reflectance value of studied samples varies from 1.08–1.38% and indicates the existence of bituminous with a significant quantity of methane in the block (Hou et al. 2016; Li et al. 2020).

Assessment of gas content

Methane is found in coal bed as absorbed and adsorbed from the latter constitutes 90% of the total amount of gas (Evans et al. 2020; Yee et al. 1993). The quantity of gas present in coal is known as gas content, and it depends on coal geology, seam thickness, depth of burial, igneous intrusions, and bounding strata type (Crosdale et al., 1998; Kiani et al., 2018; Yan et al., 2020). Methane evolution in coal is a two-step process biogenic (microbial activity) and thermogenic (cracking of organic matter). Gases generated in the succeeding thermogenic stage could migrate due to high-pressure regimes and remain stored in the coal (Altowilib et al. 2020). The pore and fracture systems were developed in coal during maturation. The micro-pore systems significantly control the gas's storage and flow in the coal matrix (Moore et al., 2014; Sharma et al., 2019). Methane is trapped in pores and fractures in the free, adsorbed, and dissolved states. Some of the microscopic images of the pore and fracture system are shown in Fig. 8. Modified USBM Direct Method was used for the measurement of in situ gas content. The total gas content of samples ranged from 2.09 to 11.64 (cc/g) on a dry ash-free basis, whereas residual gas content varies from 0.17 to 1.07 (cc/g) at standard condition (STP). The lost gas of the studied sample varied from 0.07 to 0.52 (cc/g) on a dry ash-free basis (Table.2). The desorbed gas content varies from 1.37 to 10.18 (cc/g) on a dry basis. The study also depicted that emphasis correlations were found between desorbed gas and total gas, as illustrated in Fig. 9. The study also revealed that total gas content value increases with desorbed gas, which may be due to the high desorption rate of studied samples in the field.

 Table 2
 Maceral analysis and In situ gas of studied coal samples

Sample No	Vitrinite (%)	Liptinite (%)	Inertinite (%)	Ro (%)	Wt (g)	Q_1 (cc/g)	Q_2 (cc/g)	$Q_3 (cc/g)$	Q (cc/g)	
1	63.29	6.53	20.16	1.28	1210	0.46	2.99	0.21	3.67	
2	65.99	7.77	35.25	1.23	1090	0.37	2.76	0.21	3.34	
3	69.97	6.29	20.26	1.38	1050	0.07	2.65	0.17	2.89	
4	70.40	7.34	35.31	1.08	1440	0.21	6.85	0.51	7.58	
5	59.47	8.79	27.40	1.20	1640	0.25	1.37	0.47	2.09	
6	68.26	10.05	21.69	1.23	990	0.52	4.08	0.33	4.93	
7	63.75	8.20	21.18	1.21	1390	0.26	7.04	0.22	7.52	
8	66.96	9.09	23.50	1.19	1160	0.37	7.82	0.43	8.62	
9	71.90	9.20	18.28	1.25	1130	0.44	7.84	0.40	8.68	
10	73.39	6.46	19.38	1.13	1170	0.41	8.32	0.33	9.06	
11	66.71	5.24	16.18	1.09	1170	0.51	9.46	0.96	10.92	
12	63.04	4.54	20.63	1.31	1110	0.39	10.18	1.07	11.64	

Ro- Vitrinite Reflectance; Wt- Weight of the sample (gram); Q_1 - Lost Gas (cc/g); Q_2 - Desorbed Gas (cc/g); Q_3 -Residual Gas (cc/g); Q- Total gas content (cc/g)



Fig. 6 Description: Representative photomicrographs of macerals of Raniganj Bituminous coal. Figure (A) shows bed of vitrinite [Vt] and inertinite[It] macerals; frame (B) shows sporenite[Sp] and cutinite[Cu] macerals of liptinite group, under fluorescence light; (C) frame of telenite[TI] and (D) semifusinite[Sf] macerals; (E) groundmass of telovitrinite[Tv] with inertodetrinite[Id]; (F) bed of fusinite[Fu] macerals





Fig. 7 Ternary facies diagram involving maceral and visible mineral matter contents after Kumar et al. 2018



Coal quality analysis

The gross calorific value, ultimate and proximate analyses were performed, and their results are summarized in Table 3. Due to the existence of significant quantity combustible component in coal samples, calorific value high is in all samples, and it varies from 5160 to 6840 cal/gm. It can be illustrated from the various analyses sample encompasses lower moisture content, and it varies from 0.80 to 1.90%. Samples also contain a moderate to high amount of ash percentage, and it varies from 16.90 to 32.60%. Similarly, organic contents like volatile matter and fixed carbon vary from 7.20 to 22.40% and 51.70–70.50% (dry basis).

Samples have a significant quantity of carbon and hydrogen content; value varies from 78.59 to 92.16%, 3.40–5.36% on dry ash-free basis, respectively. The sulfur and nitrogen content of samples is below 1.16% and 2.71%. The Van Krevelen diagram (Van Krevelen 1993) chart represents







Fig.9 Correlation between desorbed gas and total gas content for studied samples

Table 3 Proximate and ultimate

analysis of samples

samples' maturity, kerogen type, and nature. The cross-plot between H/C and O/C shown in Fig. 10 demonstrates that the samples fall in bituminous rank and lie in the category of kerogen types III.

Fourier transform infrared spectroscopy studies

FTIR analysis not only provides information about functional groups, but it is also significant for the determination of hydrocarbon generation potential (Hossain et al. 2019; Kumar et al. 2018). "A" factor (aliphatic/aromatic bands) can evaluate the hydrocarbon-generating potential of coal, and the "C" factor primarily the ratio of carbonyl/carboxyl groups to the aromatic group and illustrate the maturity of coal. It has been seen in many studies that as the "C" factor value increases, the maturity level of coal decreases (Ganz and Kalkreuth 1987, 1991; Samad et al. 2020).

S. No	Proximate analysis, w (%)			Ultimate analysis, w (%) daf					Atomic ratio		G.C.V	
	M	А	VM	FC	C	Н	Ν	S	0	O/C	H/C	(cal/g)
1	1.30	32.30	7.20	59.20	90.33	3.40	2.66	0.81	2.80	0.021	0.451	5160
2	1.80	18.70	9.00	70.50	91.76	3.82	2.29	0.74	1.38	0.010	0.500	6230
3	1.90	20.00	7.80	70.30	92.16	3.86	1.46	0.74	1.77	0.013	0.503	6150
4	1.70	32.60	8.50	57.20	87.52	5.36	2.71	1.08	3.33	0.025	0.735	5200
5	0.90	20.10	21.50	57.50	85.53	4.85	2.16	1.16	6.29	0.049	0.681	6820
6	1.20	19.10	21.20	58.50	81.83	4.84	2.11	0.82	10.40	0.085	0.710	6700
7	0.80	18.60	21.80	58.80	78.59	4.83	2.04	0.79	13.75	0.117	0.738	6750
8	1.30	17.30	22.40	59.00	80.57	4.86	2.05	0.74	11.78	0.098	0.724	6840
9	0.80	20.10	20.60	58.50	83.42	4.75	2.18	0.54	9.11	0.073	0.683	6700
10	1.00	21.60	22.30	55.10	80.32	4.99	2.11	0.53	12.05	0.100	0.746	6300
11	1.10	25.20	22.00	51.70	86.15	4.18	2.25	0.45	6.98	0.054	0.582	6280
12	1.00	16.90	20.40	61.70	83.58	4.73	2.02	0.68	8.99	0.072	0.679	6840

M Moisture; A Ash; VM Volatile matter, FC Fixed Carbon; ad-air dry basis; daf- dry ash-free basis

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Fig.10 Cross-plot between H/C and O/C atomic ratio for studied samples

The "A" factor and "C" factor can be easily determined by the following equation as given as follows (Varma et al., 2018):

$$A factor = \frac{2930 \text{ cm}^{-1} + 2860 \text{ cm}^{-1}}{1630 \text{ cm}^{-1} + 2930 \text{ cm}^{-1} + 2860 \text{ cm}^{-1}}$$
(4)

$$C factor = \frac{1710 \text{ cm}^{-1}}{1710 \text{ cm}^{-1} + 1630 \text{ cm}^{-1}}$$
(5)

It was described in different studies that as the A factor value increases, the maturity decreases and the aliphatic nature increases in coal (Durand and Espitalié 1976; Esen et al. 2020; Lis et al. 2005). Identically, the C factor value decreases with maturity, which reveals a decrease in oxygenated components in coal. A factor (> 0.664) and C factor (> 0.501) value of studied samples demonstrates that samples have moderate aromatic and aliphatic hydrocarbon (Chen et al. 2012; Yao et al. 2011). A factor and C factor indicate that types II and III kerogen are present in investigated samples (Misra et al. 2020; Varma et al. 2015b, 2018). A similar result found from the plot between atomic H/C and O/C of the coal samples demonstrates that the samples lie in the kerogen types II and III consecutively (Akanksha et al. 2020, 2017).

Conclusion

Based on the current examination, the following conclusions are drawn:

- 1. The hydrocarbon generation potential of the Sitarampur block was assessed using petrographic data. The petrographic examination illustrated that significant values of reactive macerals are present in samples, and its value varies from 67.57 to 81.10 vol. % respectively. Similarly, the petrographic analysis also demonstrates that organic matter is dominated by the prominent source of aromatic hydrocarbon with a minor proportion of aliphatic hydrocarbon, making it suitable for hydrocarbon generation potential.
- 2. The petrographic analysis confirms the appearance of textinite and attrinite **as a** primary contributor to vitrinite macerals groups. Sporenite, cutinite, and resinite macerals were dominantly present in liptinite macerals groups. Similarly, semifusinite is the only visible maceral present in samples from the inertinite group.
- Some positive correlations were also found between conversion, reactive macerals, and oil yield, which integrates the participation of various macerals during oil and gas generation in studied samples.
- 4. Fourier transforms infrared (FTIR) spectroscopy was significantly utilized in our study to determine various functional groups. "A" factor (aliphatic/aromatic bands) and "C" factor (carbonyl/carboxyl bands) value concluded that the sample has the lowest aromaticity and the highest hydrocarbon-generating potential, which was also validated by the Van Krevelen diagram.
- 5. The plots between H/C and O/C ratio in the Van Krevelen diagram indicate that the coal samples were lied in the type III kerogen and bituminous coal (gas prone zone). Which is also confirmed by the cross-plot between desorbed and total gas (cc/g).
- 6. The SEM analysis reveals the occurrence of several size pores and fracture and distribution in studied samples.
- 7. The studied samples of Sitarampur block were characterized by low moisture, high ash, and moderate volatile matter content in nature, which is suitable for methane production.

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Declarations

Conflict of interest No potential conflict of interest was reported by the authors.



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