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Hydrocarbon Source Potential of Coal-bearing Tikak-Parbat Formation of Barail Group in a Part of the Belt of Schuppen, India

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ABSTRACT

The Oligocene succession of the Tikak Parbat Formation has prominent coal bearing horizons. The formation comprises of medium to coarse grained light coloured sandstone, clay and carbonaceous shale with four workable coal seams. Core samples from two wells has been studied with regards to organic matter content and type. Relatively high total organic carbon (TOC) contents are present (average 72.103 wt %) in coal; the hydrogen index (HI) values reach a maximum of 410 mg HC/g TOC, indicating presence of type III-II kerogen, and the organic matter is thermally immature (T_{max} 428°C) in coal. The genetic potential have maximum yield of 246.53 mg/g, so the coal deposits could act as an excellent source rock for hydrocarbons if the burial depth is sufficient. Carbonaceous shale samples are at early mature stage (T_{max} 438°C). Kerogen at this maturity level is within oil window and is capable of generating oil and thermogenic gas upon thermal cracking. The study highlights dense accumulation of reactive macerals (vitrinite + liptinite) and low concentration of inertinite. The empirically derived values for coal reveal a high conversion (>90%) and oil yield (>60%).

INTRODUCTION

All sedimentary formations contain organic matter which is the source of hydrocarbons generated and accumulated elsewhere within the formation. So the organic matter characterization is important for organic geochemistry and widely used in the evaluation of hydrocarbon generation prospect. The rock-eval pyrolysis technique is the fundamental geochemical analysis technique of organic matter characterization. Published literature reveal that Barker (1974); Peters and Simoneit (1982); Singh et al., (1993); Sykes and Snowdon (2002); Sebag et al., (2006); David L. LePain and Russell A. Kirkham (2015), mention a few, have used rock-eval pyrolysis technique to generate data on organic matter, type, maturity and interpret source potential.

The upper Assam basin is in the north-eastern extremity of the Indian sub-continent. The basin is demarcated by Himalayas in the north east and by Naga-Patkai hill ranges in the south-east. Mishmi and Mikir massif form the north-eastern and south-western boundaries of the basin. The coal deposits in the Barail Group of the basin have been exploited since 1882 A.D. In the Barail Group, Tikak Parbat Formation has prominent coal bearing horizons. The rock comprises of medium to coarse grained light coloured quartzose sandstone, clay and carbonaceous shale with four workable coal seams in the basal part.

The present study is based on coal samples collected from borewells drilled within upper Assam CBM block and an attempt is made first to understand source potential of coal bearing Tikak-Parbat Formation of Barail Group with reference to pyrolysis data and second to establish relationship between pyrolysis and proximate analyses parameter.

GEOLOGICAL SETTING

Structurally the coal fields of upper Assam and its adjoining area fall within the belt of Schuppen of Assam (Evans, 1964). This is a part of the Assam-Arakan sedimentary basin, a composite shelf-slope-basinal system. The shelf part spreads over the Brahmaputra valley and the Dhansiri valley between the Mikir hills and the Naga foothills. The shelf-to-basinal slope, the hinge zone lies below the Naga schuppen belt. The basinal (geosynclinals) part is occupied by the Cachar, Tripura, Mizoram and Manipur fold belts.

In generalised stratigraphic column of the basin, the Eocene Disang Formation is at the base. This is overlain by the Oligocene Barail Group, Tipam, Girujan and Pliocene Dihing formations, in ascending order (Sarmah, 2013). The rocks of Oligocene Barail Group are sub-divided into lower Naogaon Formation, a middle Baragolai Formation and an upper Tikak Parbat Formation. Naogaon Formation comprises massive sandstones with minor shales. This formation is succeeded by mudstones, sandstones, shale and minor coal bands of Baragolai Formation while Tikak Parbat Formation contains sand stones, mud stones and several coal seams (Bezbaruah and Muzamil, 2013). The four coal seams of Tikak Parbat Formation are in Supra thrust at a depth range of 723 m to 1,067 m.

METHODOLOGY

In the present study, ten samples from different coal seams of two bore holes were collected during exploration drilling. The samples were subjected to pyrolysis to measure TOC, its characteristics and petroleum generation potential using rock-eval pyrolysis 6 instrument. Rock-eval 6 allows acquisition of data for quantification of both organic and mineral carbon, a better precision for temperature measurements allow acquisition of data for kinetics and overall, higher temperature range allow better characterization of TOC and T_{max} specially for coal and organic matter. The proximate analysis is performed following BIS Standard 1350 (Part1).

ANALYTICAL RESULTS AND DISCUSSION

Rock-eval Pyrolysis and TOC

Total organic carbon (TOC) measurement of rock samples using Rock-Eval Pyrolysis 6 instrument is important since hydrocarbon production is related to its carbon content (Mc Carthy et al., 2011). The sensitivity of the ratio of volatile hydrocarbons (S1) to generated hydrocarbons (S2) and TOC provide the primary indications for the zone of interest. The TOC content of 10 rock samples from two boreholes have been carried out and results are reported in Table 1.

The study revealed that all samples have excellent hydrocarbon

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Table 1. Rockeval pyrolysis, TOC analysis, proximate analysis of the rock samples

Sample ID	S1 (mg HC/ g Rock)	S2 (mg HC/ g Rock)	T max (°C)	HI mg HC/ (gTOC)	OI (mg CO_2/ gTOC)	TOC (% Wt.)	VM (daf) %	FC (daf) %
PRA 1C1	0.58	53.08	438	170	5	31.19	84.27	15.73
PRA 1C2	1.59	195.16	428	308	13	63.45	53.02	46.98
PRA 1C3	1.39	244.94	428	410	11	59.76	95.27	4.73
PRA 1C4	1.42	192.18	427	315	13	60.97	52.87	47.13
PRA 1C5	0.44	38.24	436	392	12	9.76	57.32	42.68
PRA 3C1	2.39	215.05	418	288	11	74.77	45.5	54.5
PRA 3C2	5.66	213.89	422	248	10	86.12	43.41	56.59
PRA 3C3	2.2	215.42	424	261	12	82.52	52.72	47.28
PRA 3C4	1.6	198.21	425	253	12	78.33	43.64	56.36
PRA 3C5	1.08	169.94	427	240	8	70.91	50.58	49.42

Explanation: S1: free hydrocarbons present in the rock (mg HC/g of rock); S2: remaining generation potential (mg HC/g of rock); T_{max}: maturity parameter based on the temperature at which the maximum amount of pyrolyzate (S2) is generated from the Kerogen in a rock sample; TOC (wt %): total organic carbon; HI: hydrogen index, [(S2/TOC) x 100 mg HC/g TOC]; OI: oxygen index [(S3/TOC) x 100 mg CO2/g TOC]; VM(daf): volatile matter present percentage in dry ash free basis; FC(daf): fixed carbon present percentage in dry ash free basis.

generation potential due to their high TOC content; maximum stands at 86.12 % TOC, mean TOC value is 61.778 wt%. Coal typically has a TOC value equal to or greater than 50 weight percent and carbonaceous shale have less than 50 weight percent TOC (Potter *et al*., 2005). For all the samples, the TOC results are consistent with the lithology assigned to the sample when it was collected from wellsite.

S2, the hydrocarbon generated by pyrolytic degradation of kerogen by heating above 300°C through light on source potentiality and results indicate that significant volume of organic carbon were converted to hydrocarbons by pyrolysis. The S2 yield ranges from 195.16 mg/g to 215.42 mg/g for coal (mean S2 = 205.598 mg/g), from 38.24 mg/g to 53.08 mg/g for carbonaceous shale. Based on criteria outlined in Peters and Cassa, 1994, all samples have excellent hydrocarbon potential.

Figure 1 is a plot of S2 versus TOC, used for assessment of hydrocarbon generation potential (HGP) and all samples plot in "excellent" zone.

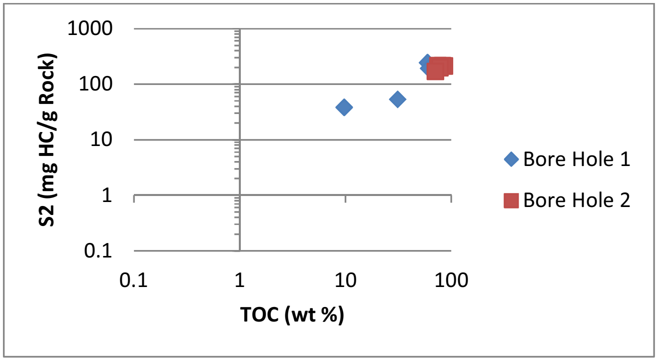

Fig.1. Cross-plot of S2 vs. TOC (%)

S1, the amount of hydrocarbon volatilized from the sample by heating to 300°C, have very low values and ranges from 1.08 mg/g to 5.66 mg/g for coal and from 0.44 mg/g to 0.58 mg/g for carbonaceous shale. Low S1 values indicate the thermal immaturity since few to no hydrocarbons were present in the samples at the start of analysis.

The plot of Hydrogen Index (HI) and TOC (wt %) shows good source rock quality because all the plots fall in the range of "fair oil source" and "gas and/or oil source" (Skret and Fabianska, 2009).

The genetic potential (GP = S1+S2; mg HC/g Rock) of studied samples from bore hole 1 have maximum yield of 246.53 mg/g and minimum yield of 38.68 mg/g while GP of bore hole 2 samples varies between 171.02 mg/g to 219.55 mg/g. This supports the fact that the coal deposits/ formation could act as an excellent source rock for hydrocarbons if the burial depth is sufficient (Fig.2)

Rock-eval Pyrolysis and Maturity

The Rock-eval temperature (T_{max}) is the temperature that represents the top of S2 peak at which the maximum amount of hydrocarbon generation takes place during pyrolysis. T_{max} provides an understanding on thermal maturity and also the burial depth attained by the source rock during evolution of the sedimentary basin. T_{max} value increases with the increase in maturation level of organic matter.

All the borehole coal samples fall on immature to late immature stage indicated by the T_{max} values of rock eval pyrolysis ranging between 418 to 428°C.

Carbonaceous shale samples of bore hole1 have 436° and 438°C T_{max} and it fall on early mature stage. Kerogen at this maturity level is within oil window and capable of generating oil and thermogenic gas upon thermal cracking (Gentzis, 2013).

The immaturity of organic matter in coal is also supported by the

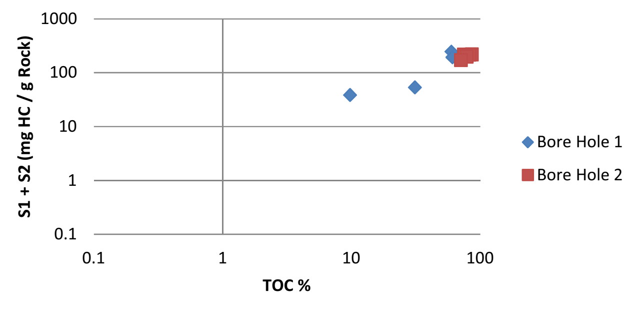

Fig.2. Plots of S1+S2 vs. TOC (%) (after Gurgey and Bati, 2018)

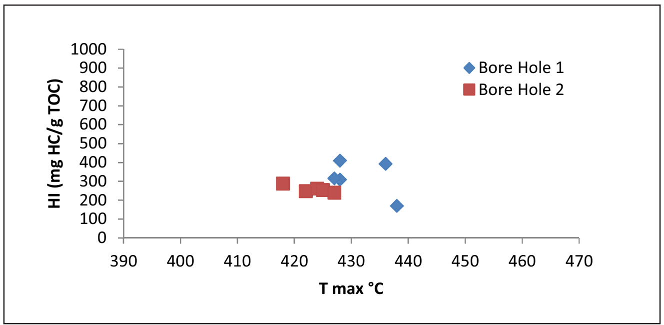

Fig.3. Plot of HI vs. Pyrolysis T_{max} (°C) (after Espitalie et al., 1986)

HI versus T_{max} plot (Fig.3).

No obvious relationship is found between HI and T_{max} and this can be explained by wide range of HI values prevail over oil window and it converges towards lower value when attain considerably high maturity.

Low production index (PI) values, the ratio of generated hydrocarbons to potential hydrocarbons, (PI = S1/(S1 + S2); table 1) are consistent with immature coals.

Rock-eval pyrolysis and Organic Matter Type

When combined with TOC measurements, S2 and S3 are used to calculate the hydrogen and oxygen indices, respectively (HI = S2/TOC and OI = S3/TOC) (David et al., 2015). The HI and OI are plotted on a modified van Krevelen diagram and used to indicate the type of kerogen present in a sample. It can be classified as Type I – highly oil prone, Type 2 – oil-prone, Type 3 – gas prone and Type 4 – Inert (Peters, 1986).

ype 1 commonly posses HI values more than 600 mg HC/g TOC while Type 2 are known to posses values greater than 300 but lower than 600 mg HC/ g TOC (Peters and Cassa, 1994; Nicholas et al., 2004; Hakimi et al., 2012 a,b).

Samples with HI values between 200 and 300 mg HC/g TOC correspond to a mixture of Type 2 and 3 and expected to expel a mixture of oil and gas at peak maturity (Peters and Cassa, 1994). Types 3 mostly composed of woody materials, posses HI values between 50 and 200 mg HC/ g TOC and expel mainly gas at peak maturity.

Kerogen consist of inert materials having HI value lower than 50 mg HC/ g TOC are known as Type 4 (Varma et al., 2015).

The HI of coal samples ranges from 240 to 410 mg HC/g TOC, while the OI ranges from 8 to 13 mg CO_2/g TOC. Important to mention here that the organic matter with HI value more than 200 mg HC/g TOC have ability to generate liquid hydrocarbon under favourable condition (Hunt, 1991).

Figure 4 is a plot of HI and OI of analysed samples in the Modified Van Krevelen diagram (values are in Table 1). Composition of organic matters of coals mostly falls in the evolutionary path of mixed Type 2 & Type 3. One Carbonaceous shale sample suite Type 3 and the other one Type-2 on Fig.4.

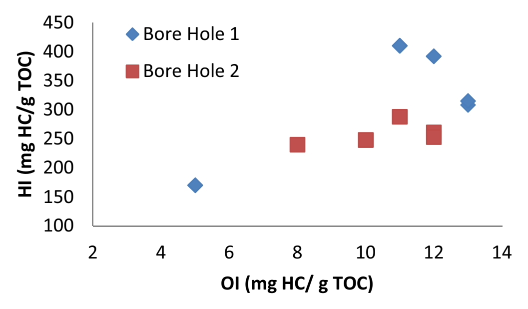

Fig.4. Plots of HI vs. OI (after Nady and Hammad, 2015)

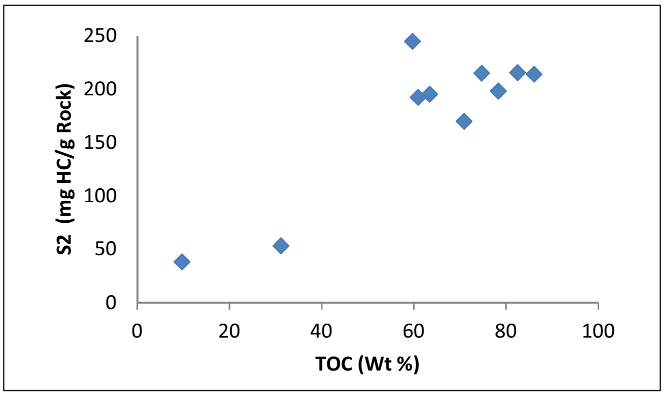

Fig.5. Plot of S2 vs. TOC (%) (after Rezaie et al., 2015)

A graphical plot of S2 versus TOC (Fig5) also confirms the same. The slope of regression line in the S2 vs. TOC diagram is low, apparently due to the presence of Type 3 materials (Langford and Blanc-Valleron, 1990).

Liquefaction of coal is influenced by its chemical constituents and maceral composition (Fisher et al. 1942). Petrographic constituent analysis of two samples have revealed reactive maceral concentration at higher side (96.4% & 90.5%) and vitrinite reflectance values stand at 0.55 & 0.53% (Table 2). Chemical constituent study (Table 1; Banik, Prasun, 2020) also fulfils the liquefaction criteria (Cudmore, 1977). Similar studies have been carried out by Singh et al. (1994) on coal deposits of the Rajmahal basin, Bihar and on lignite deposits of Saurashtra basin, Gujarat by Singh, M.P. et al. (2017), Singh, V.P. et al. (2017).

The empirically derived equations of Guyot (1978) and Jin and Shi (1997) have been used to calculate the 'conversion' of coal to oil and 'oil yield' of Barail formation. The conversion percentages of studied samples are in between 94.08 – 95.88% while oil yields (64.71 & 66.01%) (Table 2) are significant (Singh, P. K., et al. 2016, 2017; Pandey et al., 2018). The rock-eval analysis values validate the empirically derived result. Thus, petrographic, chemical and rock-eval studies of selected samples indicate that they may be potentially utilized through liquefaction. However, pilot scale study is essential and must for any kind of strategic decision.

Mean Vitrinite Reflectance and Maximum Burial Temperature

All sedimentary rocks basically contain organic matter. Hydrocarbons are produced by thermal alteration of this organic matter at temperatures between 50°C and 175°C in sub-surface under anaerobic condition through long period of time. In laboratory the same natural process has been substituted by subjecting/exposing the organic matter in unusually a very high temperature for very short duration of time known as pyrolysis method. The results are comparable to the natural catagenesis and helpful in interpretation of sub-surface activities happened.

Mean vitrinite reflectance (R_0 mean) is a measure of thermal maturation of sedimentary organic matter and is a function of maximum burial temperature (T_{bmax} in °C) of sediments (Barker and Pawlewicz, 1986). The relation is mentioned below:

$$\ln (R_0 \text{ mean}) = 0.0096\, T_{bmax} - 1.4 \qquad (1)$$

The onset of petroleum generation in source rocks is generally started at a vitrinite reflectance of 0.6% and termination of liquid petroleum generation corresponds to a vitrinite reflectance range of 1.5 to 2.0 percent. Using above formula (1), maximum burial temperature T_{bmax} will be 92.6°C for commencement of generation of liquid petroleum. So considering geothermal gradient of 25°C/km and 30°C/km, the onset of liquid petroleum generation would occur at depths of 2864 m (9397 ft) and 2387 m (7832ft) respectively(mean annual temperature at surface = 21°C).

Applying similar procedure, the collected samples would have subjected to temperature range from 41°C to 112°C. These would correspond to burial depth range from 800 meter to 3640 meter respectively (considering 25°C/km geothermal gradient). Hence this coal bearing strata will possibly act as source bed at deeper depth in the basin. This may be validated by further study of formations of various depth range within the basin.

Rock-eval Pyrolysis and Proximate Analysis

The rock-eval pyrolysis method is the most basic organic geochemical analysis of organic matter based on steady heating of

Table 2. Values of conversion, oil yield and petrofactor calculated for the Barail Formation using empirical formula

Sample ID	Huminite	Liptinite	Inertinite	Min Matter	Reactive Macerals	Rmax	Conversion %		Oil Yield %	RF
							Conv-1	Conv-2		
PRA10C4	60.1(70.0)	22.7(26.4)	3.1(3.6)	14.1	96.4	0.55	95.88	94.21	66.01	5.71
PRA13C1	83.4(87.7)	2.7(2.8)	9(9.5)	4.9	90.5	0.53	94.7	94.08	64.71	5.86

Values in parenthesis are recalculated on m.m.f. basis. Rmax – Maximum reflectance of Vitrinite; RF –Petrofactor. Conv-1 = 99-0.84RF (Jin and Shi 1997); Conv-2 =0.2 RM+76.6 (Guyot 1978); Oil Yield% = 0.22 RM + 44.8; RF = 1000 Rmax/RM

rock samples so that the total evolved hydrocarbons can be monitored as a function of temperature. This method permits rapid evaluation of the organic matter type, quantity and maturity. The thermal maturity is deduced from Tmax value. The governing relation between Tmax and Vitrinite reflectance is given by Jarvie et al., (2001). The equation is mentioned below:-

$$\text{Estimated \%VRo} = 0.0180 \text{ x } T_{max} - 7.16 \qquad (2)$$

Where, VRo = Vitrinite reflectance, estimated from T_{max}. T_{max} = Pyrolysis temperature in degree Celsius.

The chemical composition of coal is analyzed by proximate and ultimate analyses (ASTM, 1993). Proximate analysis is carried out to determine basic four constituents namely fixed carbon, volatile matter, ash and moisture. Ash is a measure of coal purity and highly variable; related to depositional environments and cleat mineralogy. Fixed carbon increases with rank; Volatile matter and moisture decreases with coal rank. Coal rank is defined according to vitrinite reflectance value of coal. The value of vitrinite reflectance steadily increases with maturity of coal and coal rank is generally determined by it. The correlation between volatile matter and vitrinite reflectance was proposed by Rice, 1993. The formula is as follows:

$$\text{Estimated \%VRo} = -2.712 \times \log(VM_{(daf)}) + 5.092 \qquad (3)$$

Where, $R_o\%$ = Vitrinite reflectance (%). $VM_{(daf)}$ = Volatile matter (dry ash free basis) (%)

From above discussion it is evident that there is relationship between Tmax and $VM_{(daf)}$ for coal and it is derived from equation (1) and (2) as below:

$$T_{max} = -150.67 \log(VM_{(daf)}) + 680.67 \qquad (4)$$

Therefore, coal will have liquid hydrocarbon generation potential when volatile matter (daf) percentage is in the range of 25 to 45 (T_{max} values are 470°C and 430°C respectively) and dry gas when volatile matter (daf) percentage is lower than 25 percent. So, bore hole 2 coal samples are prosperous compared to bore hole 1 coals.

CONCLUSION

The investigation presents basic information on organic matter and its characterisation through Rock-Eval Pyrolysis. This laboratory technique has been applied to rock samples of Oligocene Barail Formation to study the nature of the organic matter across coal bearing zone.

The petrographic analysis shows that samples are rich in huminite (60.1- 83.4%) while liptinite (26.4 – 2.9%) and inertinite (3.1 – 9.0%) have low concentration. Vitrinite reflectance value (0.55 -0.53) place them as low rank coal.

This analysis indicates that the organic matter contained in these coal is immature and of type 2 & 3 and admixture of type 2 -3, which are expected to be sources of hydrocarbon under favourable subsurface conditions. Available data suggest that coal samples are in the late pre-oil phase of maturity. Carbonaceous shale does constitute a potential source rock and are in pre-oil window phase. Report on pyrolysis tabled shows encouraging result on TOC, HI, S2 values.

The amount of fixed hydrocarbons (53.09 to 215.42 mg HC/g) is much higher than the free hydrocarbons (0.58 to 5.66 mg HC/g) in the Barail Formation suggest the good source rock candidate for hydrocarbon generation. The Rock-eval analysis is also supported by the empirical study which shows a very high conversion (94.08 – 95.88%) and oil yield (64.71 - 66.01%).

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