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Assessment of Hydrocarbon Generation Potential of Gondwana sediments of Permo-Carboniferous, Lesser Himalayan, NE India

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Abstract

This paper represents the geochemical characteristics of Gondwana coals from the Bhalukpong-Bomdila, Arunachal Pradesh in order to investigate the potential for hydrocarbon generation. A total ten (10) coal samples were collected from outcrops of Elephant flat area encountered within Gondwana succession of Permo-Carboniferous age. The coals were analysed to assess the source rock potential, organic matter quality, type and maturity using Rock-Eval pyrolysis, maceral analysis, vitrinite reflectance and Proximate Analysis. The TOC contents of the coal samples range between 2.19 to 73.68 wt.% (average 42.7 wt%). The HI, Tmax, GP and PI values range between 12 - 256 mg HC/g TOC, 439 - 561^{o}C, 5.13 -17.66 mgHC/g rock and 0.01 – 0.18 mgHC/g rock, respectively. The organic matter consists predominantly of type III and type IV kerogen with respect to hydrocarbon generation. And the organic matter is mainly gas prone and thermally early to over mature level. The primary investigation results are showing that these Permo-Carboniferous Gondwana coals are having sufficient hydrocarbon generation potential. As the coal and carbonaceous shale bearing Gondwana rocks extends below the Miri Formation to an unknown depth and lateral extent because of thrusting, there is possibility of occurrence of hydrocarbon reserves under the thrust and also in the Miri Formation in conventional or unconventional form.

Introduction

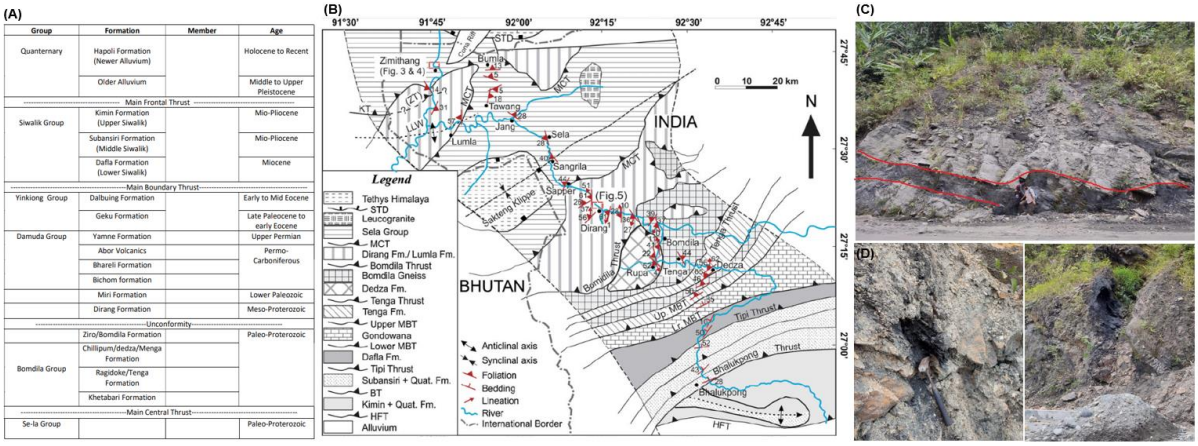
The narrow belt of lower Gondwana sediments of Permo-Carboniferous age occurring along the foot hills of Arunachal Pradesh in the Kameng, Subansiri and Siang districts. This inner belt is bounded by thrusts along the southern and northern boundaries, therefore it is highly folded and deformed. Along the southern boundary, the Gondwana sediments are thrust over the Siwalik Group. The thrust marking the northern boundary brings the metamorphosis in juxtaposition with the Gondwana sequence. The Gondwana sediments are deposited as discontinuous patches in this zone of thrusts. Unlike those associated with the Gondwanas of peninsular India (i.e. Damuda Group), these coal seams are lenticular, crushed and impersistent in nature due to the tectonic setting of the coal belt (Raja Rao, 1981; Tripathi and Roy Chowdhury, 1981). Gondwana rocks are exposed in a linear belt of about 300 km starting from Kameng district in the west (through Subansiri) and Siang districts in Arunachal Pradesh, NE India. Coals of Lower Gondwana age are associated with these rocks and occur as thin seams (0.3 to 2.5 m). Disposition of the stratigraphic groups are tectonically controlled, which in turn is affected by moderately tight NE-SW trending antiformal-synformal folds with axis plunging towards NEand SW direction. The regional strike of all rock units exposed in the area is NE-SW with dip direction towards NW. The average dip of bedding planes varies between 15° and 30°. Departure from these general attitudes has been affected at places by thrusts, faults and shear zones. Coal is an important source rock for both natural gas and crude oil (Petersen, 2006). Available information on the hydrocarbon potential of Gondwana coal in Lesser Himalayan is limited. The present study attempts to improve the understanding of hydrocarbon generation potential through organic geochemical investigation of Lower Permian Gondwana coals in the Assam-Arakan Basin, northeast India.

Geology of Area

A narrow exposure of Gondwana sediments comprising folded and deformed beds occur along the foot hills of the eastern Himalaya in the Kameng District of Arunachal Pradesh. The Gondwana rocks in the area is tectonically emplaced below the Miri Formation occurring to the north and by the Siwalik Group of rocks exposed to the south. This rock sequence is classified as Bhareli Formation in the study area. The Bhareli Formation comprises of gritty, coarse and medium to fine-grained sandstone, massive sandstone alternating with shaly sandstone, silty shale, grey and carbonaceous shale and discontinuous bands and lenses of coal varying from 20 cm to 1.7 m in thickness. The Miri Formation is mainly composed of coarse grained orthoquartzites with a zone of impersistent intraformational conglomerate beds along the thrusted contact with the Gondwanas. Basaltic rocks belonging to the Abor Volcanics is underlying and sometimes intercalated with the Miri Formation. Towards south, the

Gondwana rocks are separated from the Siwalik group of rocks by Main Boundary Thrust. The Siwaliks are represented by coarse to medium grained immature sandstone with minor bands of shale and siltstone. A generalised stratigraphic division of the Arunachal Himalayas (Table -1) as established by GSI (2010).

Table -1: (A) A Generalised stratigraphy division (GSI,2010), (B) Geological map of western Arunachal Pradesh (modified after Kumar 1997; Bhattacharjee and Nandy 2007;Goswami et al.2009;Yinet al. 2010) shows distribution of the different stratigraphic units in the area. Structural databased on traverses from Bhalukpong–Tawang and Tawang–Zimithang. STD: South Tibetan Detachment, KT: KakthangThrust, ZT: Zimithang Thrust, LLW: Lum-La Window, MCT: Main Central Thrust, MBT: Main Boundary Thrust, HFT:Himalayan Frontal Thrust, BT: Bhalukpong Thrust, (C) Coal band intercalated with shale, (D) Pockets and Lenses of Coal



Occurrence of Coal: Exposures of coal has been found within the fine to medium grained sandstone of Gondwana Group of rocks. Most of the coal exposures occur in lensoid shape and in thin bands. The strike extension of individual coal seam/bands ranges from 3m-7m. Due to subsequent deformation and thrusting the coal seams are of impersistent nature and thecoal has been crushed, sheared and powdered making it difficult to correlate the scattered coal lenses as to whether they belong to the same coal seam or not. The coal seams in the Gondwana sediments are insignificant and have little or no economic value. The coal seams occur in the upper part of the Bhareli Formation.

Material and Methodology

Ten (10) coal samples were collected at Elephant flat area of Bhalukpong-Bomdila road section. The thickness of the seam is about 3m-7m. The Gondwana sediments mainly comprise of gritty, coarse and medium to fine-grained sandstone, massive sandstone alternating with shaly sandstone, silty shale, occasionally black micaceous sandstone, grey and carbonaceous shale and discontinuous bands and lenses of coal. The coal pieces were slightly heavier and had brownish and yellowish tinges on the surface.

Rock Eval Pyrolysis is used to measure the quantity, quality, type and maturity of organic matter in rock samples and also determine the hydrocarbon generation potential in rocks. During the pyrolysis a FID monitors the release of hydrocarbon giving the peak S1 (thermos-vaporized free hydrocarbons) and peak S2 (hydrocarbon forms cracking of organic matter). In addition, CO and CO_2 released during pyrolysis can be monitored in real time by mean of an IR (infra-red) cell, giving information of the oxidation state of organic matter. The Rock Eval pyrolyzer consist of a pyrolysis oven where programmed temperature heating in an inert atmosphere (helium) of a small sample (~100 mg) takes place in order to determine the free hydrocarbons contained in the sample and the hydrocarbon and oxygen containing compounds (CO_2) that are volatilized during the cracking of the un-extractable organic matter in the sample i.e. kerogen. At the time of pyrolysis, the oven is kept isothermally at 300°C for 3 min and the free hydrocarbons are then volatilized, which is measured as the S1 peak detected by FID. After that, the temperature is increased to 550°C (at 25°C/min) and the volatilization of the very heavy hydrocarbon compounds as well as the cracking of non-volatile organic matter takes place. The S2 peak gives the hydrocarbons released from this thermal cracking as measured a by FID and the highest temperature at which S2 reaches its maximum depends on the nature and maturity of the kerogen i.e. Tmax at 300°- 390°C temperature range, the CO_2 released from the

kerogen cracking is trapped. The trap is heated, and CO_2 is released and detected on a (Thermal Conductivity Detector) TCD during the cooling of the pyrolysis oven which gives the S3 peak. Residual carbon is also measured and is recorded an S4 peak.

Four basic parameters are obtained by pyrolysis (Tissot and Welle, 1984) are as follows:

- S1 (mg HC/g TOC) - the amount of free hydrocarbons analysis.
- S2 (mg HC/g TOC) - the amount of hydrocarbons generated through thermal cracking of non-volatile organic matter.
- S3 (mg HC/g TOC) - the amount of CO pyrolysis of kerogen.
- Tmax (°C) - The temperature at which the maximum release of hydrocarbons from cracking of kerogen occurs during pyrolysis (top of S2 peak).
- Hydrogen Index (HI) - [100 * S2] / TOC.
- Oxygen Index (OI) - [100* S3] /TOC.
- Production Index (PI) - S1/ [S1+S2].
- Genetic Potential (GP) - S_1+S_2

Proximate analysis gives a measure of relative amount of volatile and non-volatile organic compounds in the coal as well as the percentage of water and non-combustible mineral materials. The proximate analysis of coal is determined by series of prescribed or standard test methods (ASTM D3172, IST1981). The procedure is used as a simple means of determining the distribution of products obtained when the coal sample is heated under specified conditions. The proximate analysis of coal separates the products into four groups: (1) Moisture, (2) Volatile matter (3) Fixed carbon, the non-volatile fraction of coal, and (4) Ash, the inorganic residue remaining after combustion. If corrections are applied to compensate for the non-organic impurities, proximate analysis enables the organic component of one coal to be compared with that of another and hence, provides much of the basic data for systematic coal classification.

Results and Discussion

Rock Eval analysis

Richness of Organic Matter: The results of Rock Eval Pyrolysis of Bhalukpong-Bomdila Coal Samples are given in Table-2. Organic matter richness is the most important parameter for evaluation of hydrocarbon potential. The organic richness and hydrocarbon generation potential of the samples can be evaluated by the Total Organic Carbon (TOC) content, the pyrolysis derived free hydrocarbon (S1) and hydrocarbon generative potential (S2). The TOC ranges from2.19 % to 73.68 % (avg. 42.27), which indicates the coals samples are rich in organic carbon and possess very good hydrocarbon generation potential, (Peters, 1986). The S1 values are low which may indicate free hydrocarbons and may also be related to highly weathered samples (Salleh et al., 2008). The S1 value of coal samples ranges from 0.11 and 1.03 mgHC/gTOC (avg. 0.33 mgHC/gTOC) which represents fair to good source rock for petroleum potential and S2 value ranges between 4.64and 17.57 mgHC/gTOC (avg.9.82 mgHC/gTOC) which represents fair to very good source rock for petroleum potential. These Rock-Eval parameters reflect that the Bhalukpong-Bomdila coal have fair to excellent generative potential (Peters, 1986). The same is also reflected by the TOC versus S2 diagram (El Nady et al., 2015). According to Hunt source rocks with a genetic potential (GP) is considered to have poor, fair, good, and very good generation potential, respectively. The Bhalukpong coal samples of the study area contain very good GP averaging at 10.13 mgHC/g rock, which indicates that the coal deposits could act as a very excellent source rock for petroleum if the burial depth is sufficient. The plot of TOC versus HI (Jackson et al., 1985) mostly shows gas source in the Bhalukpong-Bomdila coal. The plot of TOC versus S1 (Gürgey and Bati, 2018) indicates that all samples are characterized by autochthonous hydrocarbon.

Organic Matter Types: Hydrogen indices (HI) above 200 are generally considered capable of generating some liquid hydrocarbons (Hunt, 1991). Organic matters with hydrogen index between 150-300 mgHC/gTOC have potential to generate both gas and oil, while lower and higher hydrogen indices than this range indicate only gas or oil generation potentials of organic matters respectively (Peters, 1996). HI values below about 150 mg/g indicate the absence of significant amounts of oil generative lipid materials and confirm the kerogen as mainly terrestrial (Hakimi and Abdullah, 2014) Type III or Type IV (Waples, 1985). According to Peters (1986), low HI indicates gas prone nature of the organic matters. The HI of the Bhalukpong-Bomdila Coal samples range from 12 to 63 mg/g with

only two samples (1 & 7) having 211 mgHC/gTOC & 256 mgHC/gTOC with an average of 63 mg/g, indicating dominant presence of Type III/IV kerogen which are potential source of gas. Type IV kerogens usually have less than 50 mg/g of HI (Tissot and Welte, 1984). The TOC versus S2 diagram (Langford and Blanc-Valleron, 1990, Fig. 9) indicates presence dry gas prone kerogens in the Bhalukpong - Bomdila coals. Cross plot of HI versus OI (Espitalie et al., 1977) also shows Type III and Type IV kerogens.

Thermal Maturity of Organic Matter: The hydrocarbon generation from organic matters through its burial history is a part of thermal metamorphism of organic matters (Tissot and Welte, 1984). Tmax and Production Index (PI) are considered as reliable proxy of organic maturation. According to Espitalie et al., (1985), oil generation begins at Tmax between 435 and 465^{o}C, and PI between 0.2 and 0.4 and gas generation at Tmax 470^{o}C, and PI more than 0.4. The Tmax of Bhalukpong-Bomdila coal ranges between 439 to 577^{o}C and PI ranges between 0.01 to 0.16 with an average of 0.04. The Tmax and PI of Bhalukpong-Bomdila coal indicates that the maturity of organic matter ranges within a wide limit from immature to over mature (gas generation). The plot of Tmax versus HI, indicates the maturity of the samples dominantly fall in the mature oil window (Mukhopadhyay et al., 1995). The plot of Tmax versus PI shows the maturity of the samples dominantly fall in the dry gas zone and a few samples in the oil window (El Nady et al., 2015). Also the Tmax versus PI shows the maturity of the samples dominantly fall in the Inert Materials and a few samples are immature (Sengular et al, 2008).

Table-2: Rock Eval Pyrolysis Results of Lower Gondwana coals

Sample No	TOC %	S1 (mgHC/gTOC)	S2 (mgHC/gTOC)	S3 (mgHC/gTOC)	HI (mgHC/gTOC)	OI (mgHC/gTOC)	Tmax (°C)	GP (mgHC/g rock)	PI (mgHC/g rock)
A1	2.19	0.95	4.64	0.62	211	28	439	5.59	0.16
A2	58.4	0.15	14.4	1.36	25	2	539	14.55	0.01
A3	68.84	0.22	8.46	6.05	12	9	577	8.68	0.02
A4	57.55	0.17	15.42	2.32	27	4	532	15.59	0.01
A5	43.42	0.11	9.41	0.94	22	2	538	9.52	0.01
A6	44.15	0.19	6.93	4.27	16	10	555	7.12	0.18
A7	2.24	1.03	5.75	0.36	256	16	440	6.78	0.02
A8	32.01	0.11	5.02	3.43	16	11	561	5.13	0.01
A9	45.29	0.07	10.61	0.52	23	2	528	10.68	0.01
A10	73.68	0.09	17.57	3.66	24	5	535	17.66	0.01
Average	42.7	0.33	9.82	2.01	63	8.5	524	10.13	0.04

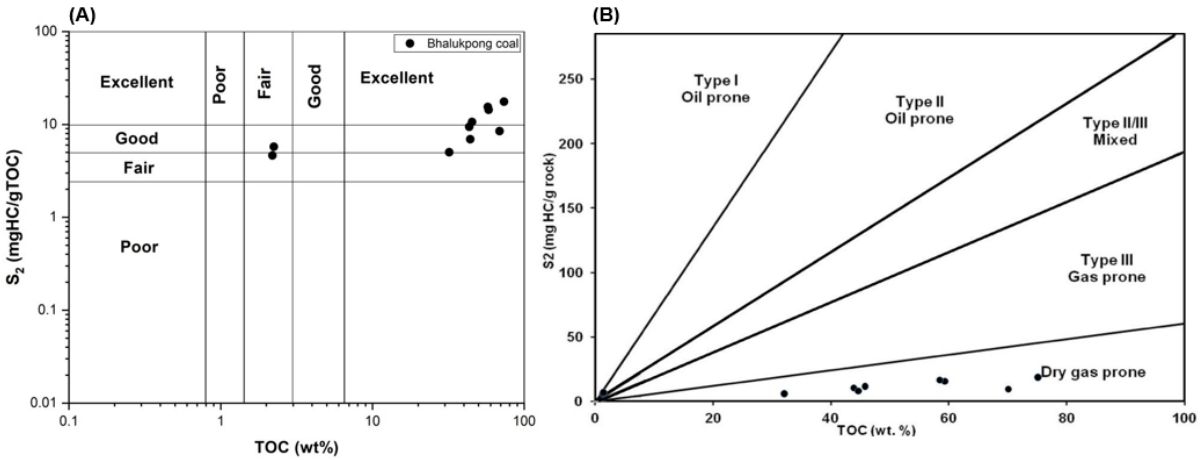


Fig.1. Cross plots of TOC and S2 (A) Organic richness, (B) Organic matter type

Proximate analysis

The results of Proximate analysis of Coal Samples are given in Table-3. The Moisture Content ranges between 0.3% to 1.7% with an average of 1.09%. The Ash Content ranges between 19% to 60.7 % with an average of 37.16 %. The Volatile matter content ranges between 7.25% to 15.36 % with an average of 10.70 % and the Fixed carbon content ranges between 28.33% to 69.26% with an average

of 50.92 %. The Mineral matter present in the coal samples ranges between 20.52% to 65.55% with an average of 40.13%. The Fuel ratio in coal is between 3.05 and 7.03 with an average of 4.87. The calorific value of Coal, determined using Goutal's Formula is given in Table - 4. For Classification of coal deposits the coal classification system adopted by the Indian Standard Institute is used which is mainly based on the yield of volatile matter and calorific value (on dry mineral matter free basis) together with the moisture content. The Range of Volatile Matter percentage for Coal is in the range of 7.25 – 15.36, Range of Calorific Value is between 8624.72 - 8748.28 and the range of moisture percentage in air dried basis is between 1 and 2. The coal deposits of can be established to be of Anthracite to Semi Anthracite type.

Table-3: Proximate analysis results of Lower Gondwana coals

Sample No.	In air dried basis					
	Moisture %	Ash %	Volatile Matter %	Fixed Carbon %	Mineral Matter (MM= 1.08* Ash%)	Fuel Ratio
A1	0.90	19.00	9.84	69.26	20.52	7.03
A2	1.70	20.08	13.70	63.80	21.68	4.65
A3	0.80	29.00	11.11	59.09	31.32	5.31
A4	1.09	49.00	7.83	42.08	52.92	5.37
A5	1.40	47.16	9.60	41.84	50.93	4.35
A6	1.70	60.70	9.27	28.33	65.55	3.05
A7	0.80	50.30	7.25	41.65	54.32	5.74
A8	1.10	33.50	13.52	51.88	36.18	3.83
A9	0.30	33.30	9.60	56.80	35.96	5.91
A10	1.20	29.60	15.36	53.84	31.96	3.50
Average	1.09	37.16	10.70	50.92	40.13	4.87
Minimum	0.30	19.00	7.25	28.33	20.52	3.05
Maximum	1.70	60.70	15.36	69.26	65.55	7.03

Table-4: Calorific value of Bhalukpong Coal (determined using Goutal's Formula)

Sample No	Volatile Matter % (in dry ash free basis)	Fixed Carbon % (in dry ash free basis)	Volatile Matter % (in mineral matter free basis)	α (determined using Goutal's chart)	Calorific value (Kcal/Kg)
A1	12.28	86.46	12.52	125	8624.72
A2	17.51	81.56	17.88	114	8684.06
A3	15.82	84.17	16.36	116	8737.06
A4	15.68	84.31	17.02	115	8716.62
A5	18.66	81.33	20.13	110	8721.66
A6	24.65	75.34	28.30	101	8667.53
A7	14.82	85.17	16.15	117	8717.88
A8	20.67	79.32	21.55	107	8715.93
A9	14.45	85.54	15.06	120	8748.28
A10	22.19	77.80	22.98	106	8732.00
Average Calorific Value					8706.57

Conclusion

The Coal occurs within the Gondwana sediments of Lower Permian age along the foothills of Himalayan are of impersistent nature and has been moderately crushed and sheared due to prolonged tectonic disturbance in the area during the Himalayan orogeny. Rock Eval analysis of the coal samples indicate that the coal samples are rich in organic carbon with an average Total Organic Carbon (TOC) of 42.27wt% which indicated that the coals have good hydrocarbon generation potential. The Coal deposits also have a good Genetic Potential (GP) averaging at about 10.13mg/g which indicate that the coal deposits act as a very good source rock for petroleum if burial depth is sufficient. However, Tmax and Production Index (PI) indicate that the maturity of organic matter ranges within a wide limit from early to over mature and indicates presence of dry gas kerogens in Coals.The proximate data indicate that the coals have low moisture (average 1.09%), low volatile matter (average 10.7%), moderate ash (average 37.6%), and a fixed carbon content of average 50.92%. The coals have a high calorific value of 8706.57 Kcal/kg. These coals have reached semi-anthracite to anthracite stage. The results indicate that the coal deposits of Bhareli Formation of Lower Gondwana Group in Bhalukpong-Bomdila have good potential to act as a source rock and also sufficient hydrocarbon generation potential. As the coal and carbonaceous shale bearing Gondwana rocks extends below the Miri Formation to an unknown depth and lateral extent because of thrusting, there is possibility of occurrence of hydrocarbon reserves under the thrust and also in the Miri Formation in conventional or unconventional form.

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