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Sulphur study of the Palaeogene coals from Jaintia Hills, Meghalaya, NE India: Implications for palaeoenvironment, utilisation prospects, and environmental impacts

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Abstract

Coal samples of Eocene age (Shella Formation) from four different mines (Bapung, Jaintia, Sutunga, and Lakadong) of the Jaintia Hills of Meghalaya, Northeast India, were collected and investigated to observe the sulphur content and to understand the palaeoenvironment, utilisation prospects, and environmental impact. The study reveals that these coal samples contain sulphur in higher concentration (4.46% to 7.26%) both organic and inorganic forms. There are 3 coal seams exposed in the area. The organic sulphur is higher (2.53%-5.49%) than the inorganic forms (1.26%-1.77%). The upper seam is found to contain higher concentration of sulphur than the lower seam. Intra seam pyritic sulphur also shows an upward increasing trend. The high sulphur content in the coal seams suggests the marine influence in the peat-forming swamps. These coals are classified as High Sulphur coal (>1%) which is the main obstacle in the utilization although high volatile matter and hydrogen content strongly suggest that these coals are good for liquefaction. Moreover, during coal combustion emissions of sulphur dioxide produce acid rain, affecting the environment of the mine areas.

Keywords: Sulphur, palaeoenvironment, utilisation, marine, environment, Meghalaya.

INTRODUCTION

The occurrence of sulphur (S) in the Palaeogene coals of India attracted the attention of early workers (LaTouche 1882; Ghosh 1964; Ahmed 1971).The North Eastern coal fields are mainly scattered in Arunachal Pradesh, Assam, Nagaland, and Meghalaya in nearly 70 individual coal deposits (Mishra and Ghosh 1996). The Palaeogene coal deposits are of good quality, having characteristically low ash content and high caking properties. But the only drawback of these coals is the high amount of sulphur (10% or more), mainly in the organic form. (Ahmed and Bora, 1981)

In the northeast region of India, coal contains both organic and inorganic forms of sulphur. Pyritic and sulphate sulphur together commonly referred to as inorganic sulphur. The NE coalfields have been studied for their physicochemical and petrographic characteristics (Ahmed and Bora, 1981; Chandra *et al.*, 1983; Mishra, 1992; Ahmed and Rahim, 1996; Mishra and Ghosh, 1996; Singh and Singh, 2000, 2001, 2003; Singh *et al.*, 2012b). The Geological Survey of India (GSI) studied the regional tectono-stratigraphy of various coalfields of NE India (Karunakaran, 1974; Satsangi and Parlda, 1980; Raja Rao, 1981; Nayak, 2013).

Important contributions have been made by several authors on the stratigraphy, structures, and tectonics of NE India (Mallet, 1876; Biswas, 1962; Bhandari *et al.*, 1973; Dasgupta and Biswas, 2000; Acharyya, 2007). The provenance of the Barail Formation in Assam has been studied by Srivastava and Pandey (2011).

The present study has been carried out on the coalfields of the Jaintia Hills district of Meghalaya. The coal seams occur in the Lakadang sandstone member of Shella Formation of Jaintia Group of Eocene age. The coal deposits of the Jaintia Hills of Meghalaya have been studied by different workers for desulphurization, the environmental impact of mining, assessment of coal quality, etc. (Ahmed and Bora, 1981; Chandra *et al.*, 1983; Sarma, 2005; Nayak, 2013; Sahoo *et al.*, 2014). The objective of this paper is to decipher the palaeoenvironmental conditions of the coal seams, also to observe the utilisation prospects and environmental impacts based on chemical and petrographic studies. The present study will provide a framework that could help future gasification and liquefaction studies.

Geological Setting

The Geological and location map of Maghalaya is given as figure 1. Meghalaya, India, contains coal deposits within Palaeogene strata. Reserves calculated for Northeast India account for 593.81 MT (Indian Bureau of Mines, 2019), out of which Meghalaya alone have 89.04 MT of coal. Seven coal mines occur in the Jaintia Hills. Bapung, Jarain, and Sutunga coal mines are important amongst them (Fig. 2). Together, they are grouped as the "Bapung coalfield" (Singh and Singh, 2000).

A well-developed sequence of the lower Palaeogene sediments is exposed at Jaintia Hills and constitutes the type area of the Jaintia Group where coal seams are associated with the Lakadong sandstone member of the Sylhet Formation (Raja and Rao, 1981) Sutunga and Jarain villages in the valleys. A low regional dip of 3° towards the south is observed in these

Table 1. Geological succession of Jaintia hills coalfield of Meghalaya (modified after Raja Rao, 1981; GSI, 2009)

Age	Group	Formation	Member	Lithology
Palaeocene-Eocene	Jaintia Group	Kopili Formation (50 m)	----------	Shale, Sandstone, marl, and coal
		Shella Formation (600 m) (earlier Sylhet Formation)	Prang Limestone	Fossiliferous argillaceous limestone
			Nurpuh sandstone	Sandstone with subordinate calcareous bands
			Umlatdoh limestone	Foraminiferal limestone containing a few sandstone bands
			Lakadong sandstone	Coal bearing sandstone
			Lakadong limestone	Fossiliferous limestone
			Theria sandstone	Medium to coarse-grained ferruginous sandstone containing thin coal seams, calcareous shales and at places clay bands
		Langpar Formation (100 m)	----------	Calcareous shale, sandstone, limestone
Archaean	Meghalaya Gneissic Complex			Gneisses, Schists and Granites

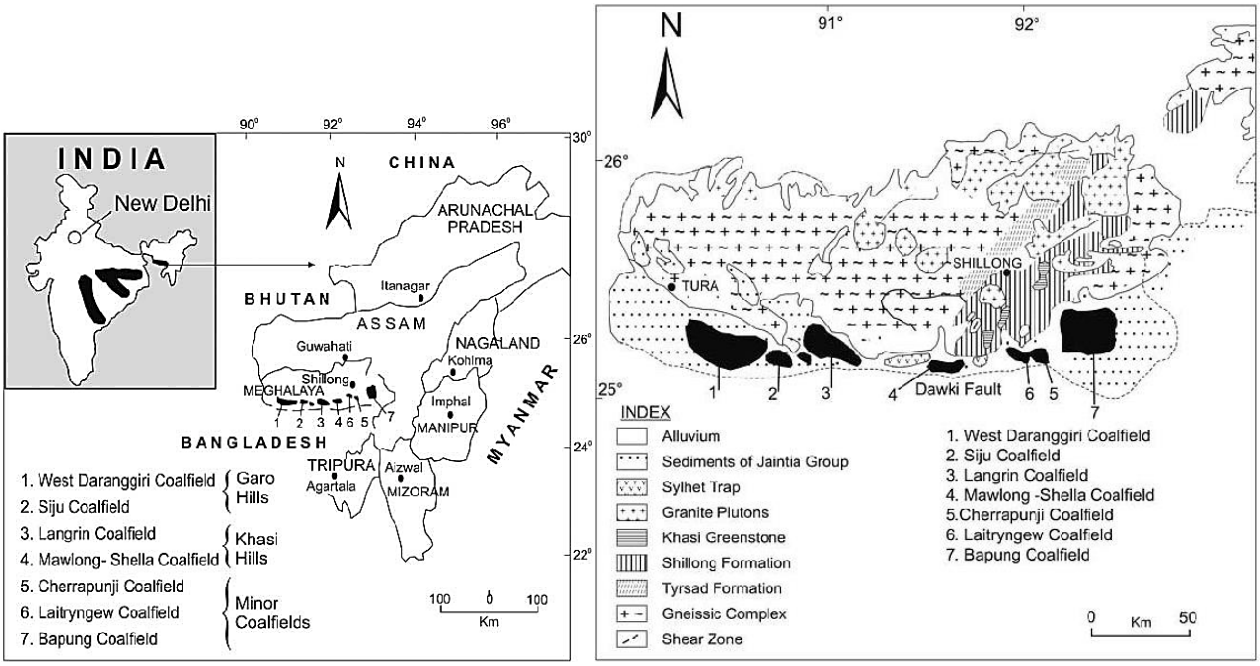

Figure 1. Location and geological map of Meghalaya, North-eastern India (after Singh *et al.*, 2015)

A generalised stratigraphic sequence is shown in table 1.The Meghalaya Palaeogene coal deposits are presumed to have been formed over platform areas under stable shelf conditions (Raja Rao, 1981; Singh and Singh, 2000). In the study area (Fig. 2) eight coal seams have been recognised at Bapung (3), Jarain (2), Sutunga (2), and Lakadong (1). In the Bapung coalfield, only the lower seams are persistent, which vary in thickness from 0.3 to 1.5 m. In some places in the Bapung area, the upper and middle seams become very thin, ranging in thickness from 0.1 m to 0.5 m. All three coal seams are associated with the Lakadong sandstone which strikes in an ESE-WNW direction with south-westerly dips varying from 3° to 6°. Two coal seams are well exposed in Sutunga and Jarain area. The lower seam occurs in the basal part of the Lakadong sandstone over the Lakadong Limestone. These seams are well exposed towards the east and north of the

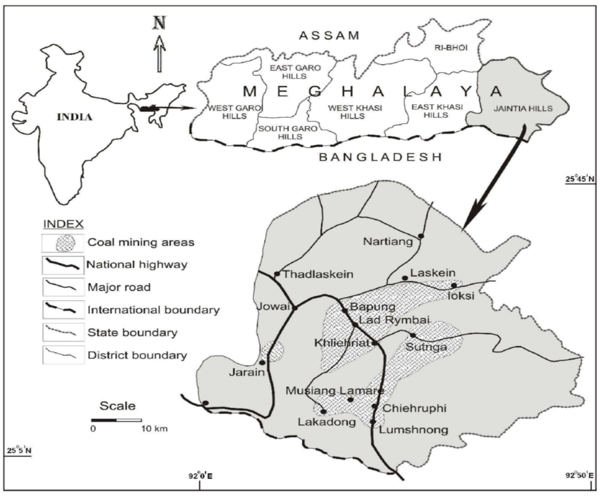

Figure 2. Outline map showing the coal mining areas in Jaintia Hill of Meghalaya in northeast India and the sample locations (Nayak, 2013).

seams. The thickness of the seam varies from 0.3 m to 1 m and 0.1 m -1.7 m in these coalfields respectively. There occurs a 0.3 m to 2.1 m thick coal seam at Lakadong.

following the guidelines of the Bureau of Indian Standards (BIS): 1350 (BIS 2003) and American

Table 2 Results of proximate analysis in weight percent of the coalfields of Jaintia hills

Site/Source	Sample No/Seam	air-dried basis			
		Moisture (%)	Ash (%)	Volatile Matter (%)	Fixed Carbon (%)
Bapung Coalfield	I	2.81	2.20	42.15	52.84
	II	1.81	1.30	40.25	56.64
	III	1.72	1.22	41.20	55.86
Jarain Coalfield	I	1.90	1.11	43.21	53.78
	II	1.50	1.91	44.02	52.57
Sutunga Coalfield	I	2.11	1.10	41.25	55.54
	II	1.62	1.02	40.62	56.74
Lakadong	I	2.20	1.82	42.25	53.73

Table 3: Sulphur content of coal samples (in wt. %) from Jaintia Hills.

Site/Source	Sample No/ Seam	Total S	Pyrite S	Sulphate S	Organic S
Bapung Coalfield	I	7.26	0.97	0.80	5.49
	II	5.25	0.82	0.72	3.71
	III	4.02	0.72	0.62	2.68
Jarain Coalfield	I	5.65	0.86	0.64	4.15
	II	4.12	0.83	0.59	2.70
Sutunga Coalfield	I	4.46	0.69	0.58	3.19
	II	3.79	0.65	0.61	2.53
Lakadong	I	4.78	0.61	0.57	3.60

Material and Methods

Coal samples were collected following pillar sampling method from the exposed outcrops of the coalfields of Jaintia Hills located in deep mines. The samples were crushed to <72 mesh and <18 mesh sizes for chemical and petrographic tests, respectively. Proximate and sulphur analyses were performed by Society for Testing and Materials, ASTM D5373-08 (2008), respectively. A Leica DM 2700 P advanced petrological microscope aided with MSP 200 coal photometer and fluorescence attachment was used for petrography following standard practices and norms of the International Committee for Coal and Organic Petrology as described in Singh and Jha (2018b).

Table 4: Petrographic composition of Jaintia Coals (in volume %).

Site/Source	Sample No/ Seam	Vitrinite	Liptinite	Inertinite	Mineral Matter
Bapung Coalfield	I	81.76	3.11	11.17	3.96
	II	80.05	3.66	13.95	2.34
	III	78.05	4.84	15.25	1.86
Jarain Coalfield	I	79.24	0.68	18.02	2.06
	II	77.92	1.72	18.74	1.62
Sutunga Coalfield	I	78.54	2.39	16.79	2.28
	II	76.25	3.09	18.92	1.74
Lakadong	I	79.32	1.28	17.02	2.38

Results and Discussion

Megascopic characterization

The coals of the area are black and generally are hard and compact but soft and friable varieties are also observed in the field. The coals break with a cubical fracture but the hard one breaks with sub-conchoidal to conchoidal fracture. Most of the coals when exposed to the sun easily disintegrate into small chips. Sometimes very thin pyrite bands are observed. They show dull to glassy lustre. On weathering the coal breaks parallel to the bedding plane. These coals are seen as a homogenous mass of vitrain. Megascopically, the coal indicates a high sulphur content, which occurs as minute yellow particles or granules.

Sulphur content of the coals

The results of the proximate analysis of the Jaintia Hills coals are presented in table 2. From table 3, it is observed that very high sulphur concentration is present in these coals. The total sulphur content of the coals varies from 3.82% to 7.26%, which is much higher than the recommended permissible limit of sulphur (as per the classification scheme of Chou (2012)) with <1%, 1-3%, and >3% are categorized as low, medium, and high sulphur coal, respectively.

The inorganic constitutes pyritic sulphur varies from 0.61% to 0.97% and sulphate sulphur from 0.58% to 0.80%. The organic sulphur, which dominates the inorganic forms, varies from 2.53% to 5.49%. From table 3, it is seen that total sulphur content (of each of the four coalfields) increases from the bottom to the top of a seam (e.g., Bapung 4.02% to 7.26%).

This intra-seam vertical variation of sulphur has been attributed to the fact that pH values decrease with depth (i.e. alkalinity increases upward from bottom to the surface) as a result sulphur content increases upward in the sequence since the alkaline condition is conducive to the deposition of sulphur (Chandra *et al.*, 1983) and also that there was more marine influence at the top of the peat. Some binary plots have been made to explore the possible relationships among the maceral groups and total sulphur (Fig. 3).

Relation of group maceral with total sulphur

The petrographic composition of the Jaintia coals is presented in table 4. A linear relationship is observed when plots are made of vitrinite vs. total sulphur. In the case of the inertinite vs total sulphur plot, an inverse relationship is observed, since leptinites in these coals are very few. From these relationships, it emerges that the precursors of vitrinite and liptinite macerals have evolved under a high water table, high pH, and low Eh conditions. Such conditions allow the reduction of sulphate to sulphide and provide an ideal situation for the formation of pyrite and organic sulphur in good concentration.

Palaeoenvironment

As per the observation of Teichmüller (1962), the coals with marine influence are generally rich in sulphur, hydrogen, and nitrogen contents and are characterized by a higher volatile matter content than other coals of the same rank. In the Jaintia coal mines, similar conditions seem to be applicable, which have relatively high S and volatile matter contents.

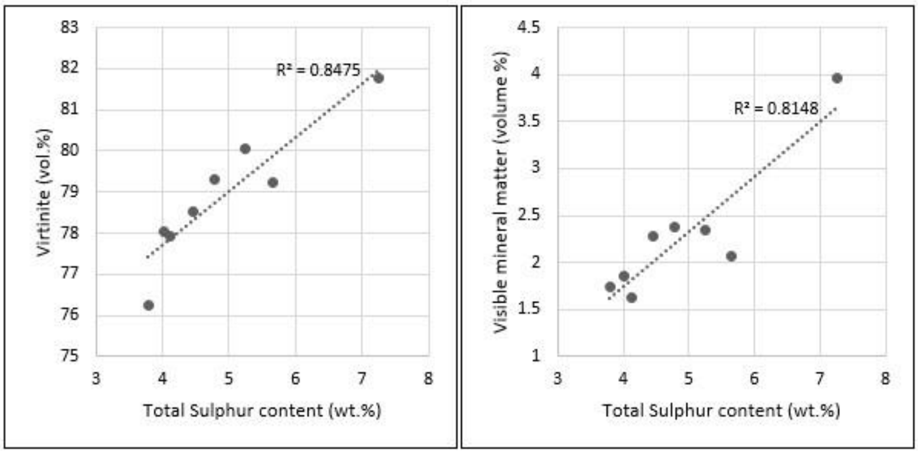

Figure 3. Correlation of microscopic constituents (vol. %) with total sulphur content (wt. %)

Organic sulphur might originate from the complexing of sulphur from sulphate ions and hydrogen sulphide by humic acids during the process of coalification (Casagrande*et al.,* 1979). Irrespective of the form, such a high concentration of sulphur in coal indicates a marine origin because freshwater contains only 0 to 10 ppm sulphur and henceforth, even if there is a prolonged circulation of fresh water through the peat, it cannot account for much sulphur in the coal. Chou (1990) indicates that most of the sulphur in coals with more than one percent sulphur comes from sea water. Price and Shieh (1979) have shown that in high sulphur coals (>0.8% S), about 63% of the sulphur is derived from seawater by sulphate reduction and the rest is derived from original vegetation. Therefore, in the Jaintia Hills, the high sulphur content in the coal seams perhaps can be explained by the repeated influx of seawater (Nayak, 2013) with an intermittent hiatus during which the water table was not enough to sink the swamp vegetation. The findings of Singh and Singh (2000) also corroborate this point of view. The model proposed by Singh and Singh (1996) shows that the coals of the study area formed under alternating oxic and anoxic moor conditions (Fig. 4). Since the Jaintia Hills coal seams occur in the Lakadong sandstone member that is sandwiched between two limestone members: the underlying Lakadong limestone and overlying Umlatdoh limestone, Raja Rao (1981) also suggested that these formations have formed due to intermittent marine transgression and regression during the Eocene period.

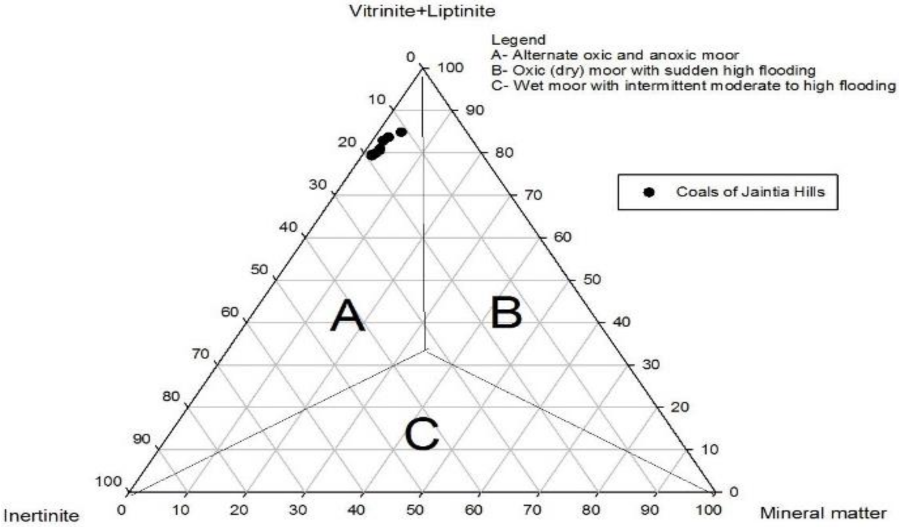

Figure 4. Depositional conditions of Jaintia Hills coals based on maceral and mineral matter content (after Singh and Singh 1996).

Table 5 Result of ultimate analysis of the coal samples of Jaintia Hills.

		Dry, mineral matter free (dmmf) basis				
Site/Source	Sample No/ Seam	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Sulphur (%)	Oxygen (%)
Bapung Coalfield	I	74.28	6.62	2.56	7.26	9.28
	II	75.36	5.13	2.71	5.25	11.55
	III	75.02	6.23	2.80	4.02	11.93
Jarain Coalfield	I	74.58	6.42	1.68	5.65	11.67
	II	73.34	5.92	2.11	4.12	14.51
Sutunga Coalfield	I	72.89	5.03	1.75	4.46	15.87
	II	75.67	5.52	1.32	3.79	13.70
Lakadong	I	72.02	5.86	2.10	4.78	15.24

Utilisation and Prospects

The majority of the coals of Jaintia Hills of Meghalaya are supplied to Assam and Bangladesh for use in brick and other industries. The presence of high volatile matter and significant concentration of hydrogen (Table 5) strongly suggest that these coals may be good for liquefaction and gasification (Singh and Jha, 2018a). In addition, these coals can produce coke or, at least, can be used in a blend for coking purposes. The high sulphur content is the main obstacle in the utilization of these coals; it is strongly felt that the possible method for desulphurization should be evolved through research and development activities. Recently, desulphurization of coal with bacterial biomass (Singh *et al.,* 2012a, 2013, 2018; Singh, 2018) has come up with a new insight into such problems. If this problem is solved, then these remotely located coal deposits may turn out to be an asset for the people of Meghalaya.

Environmental impacts

As coal contains a high amount of sulphur (dominant organic sulphur) the problems of pollution both in air and water is evident. During the burning of coals in the open air, SO_2 gas emits which further reacting with air produces SO_3. The produced SO_3 get dissolved in water and produces H_2SO_4,which causes acid aerosols and acid rain in the area. Besides, a high amount of sulphur present in pyrite also causes acid mine drainage (Graham and Sarofim 1998).

Conclusions

The high organic sulphur content of the Palaeogene Jaintia Hills coals indicates their marine origin and the formation associated with coals have witnessed both marine transgression and regression phases. Besides, as the coal deposits are rich in vitrinite and low in ash, good coke can be produced and can be used as a blend for coking purposes. High sulphur is the only limiting factor and needs to be eliminated before any use. Moreover, sulphur dioxide emitted during coal combustion is a principal source of acid rain which affects the environment badly in the study areas.

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