JOURNAL GEOLOGICAL SOCIETY OF INDIA
Vol.81, February 2013, pp.203-214

Mineral Matter and the Nature of Pyrite in Some High-sulfur Tertiary Coals of Meghalaya, Northeast India

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Abstract: Coal samples collected from four different sources in the Jaintia Hills of Meghalaya, northeast India, have been investigated for their sulfur content, mineral matter, and to assess their potential behavior upon beneficiation. These coals contain high sulfur which occurs both in organic and inorganic forms. The organic sulfur content is much higher than the inorganic sulfur. Studies on different size and gravity fractions indicated that the mineral phases are concentrated in higher density fractions ($d > 1.8$) and in general are fine grained (<50 μm). Data of reflected-light optical microscope and electron probe micro-analysis (EPMA) revealed that minerals in these coals are *sulfides*- pyrite, marcasite, sphalerite, pentlandite; *sulfates*- barite, jarosite; *oxides*- hematite, rutile; *hydroxides*- gibbsite, goethite; *phosphate*- monazite; *carbonate*- calcite, siderite and *silicates*- quartz, mica, chlorite, and kaolinitic clay. The disulfides of iron occur in two modes – mainly pyrite and occasionally marcasite with wide size ranges and in various forms, such as: framboid, colloidal precipitate, colloform-banded, fine disseminations, discrete grains, dendritic (feathery), recrystallized, nuggets, discoidal, massive, cavity-fracture- and cleat-fillings. Framboidal pyrite has formed primarily due to biological activities of sulfur reducing bacteria in the early stages of coalification. Massive and other varieties have formed at later stages due to coalescence and recrystallization of the earlier formed pyrites. Sulfur isotopic values indicate a biogenic origin for the pyrites. Association of trace metals, such as Ni, and Zn has been recorded in these pyrites. Given the large fractions of organic sulfur present, these coals can be upgraded only partially to reduce the sulfur content by beneficiation.

Keywords: High-sulfur Tertiary Coal, Pyrite, Mineral matter, Jaintia Hills, Meghalaya.

INTRODUCTION

Indian coal deposits belong to two different stratigraphic levels and basinal occurrence: (1) Permian sediments deposited in intra-cratonic Lower Gondwana basins, and (2) Early Tertiary coal (and lignite) formed in near shore basins in shelf environments having mainly peri-cratonic set up (Acharyya, 2000). The majority of the coal resources occur within the Gondwana basins that are confined to the peninsular part of the country. The Tertiary coals constitute only a small portion of the total coal resources and mostly occur in the north-eastern states of Assam, Meghalaya, Arunachal Pradesh, and Nagaland. Out of a total 211 billion tons of coal reserves, the Tertiary coal accounts only for about 1 billion tonnes (Indian Minerals Yearbook, 2008). However, when the geographical distribution of Tertiary coal is considered, its importance becomes obvious because the deposits are situated in the extra-peninsular region far away from the main coal producing provinces of the country. Therefore, these resources have special significance in meeting local and regional coal demands of the northeastern Indian states. These coals are characterized by relatively low-ash compared to Indian Gondwana coals and in some cases have good coking properties. However, coal from this region is not being fully exploited due to its high sulfur content; sometimes exceeding 10 wt.% (Ahmed and Rahim, 1996; Mishra and Ghosh, 1996; Singh and Singh, 2006). As sulfur and mineral matter are undesired elements in coal, a detailed characterization with respect to their mode of occurrence and distribution is required for proper assessment of these coals. Accordingly we have investigated coals from four different localities in the Jaintia hills of Meghalaya state and present here a brief account on the associated inorganic mineral phases and their mode of occurrence. In addition, a detailed discussion is given on the character and origin of pyrite, which, is the dominant mineral phase in these coals.

Although macerals are the most important constituents of coal, the abundance and composition of mineral matter often dictates the potential of a coal-type for any specific end use. Most coal-conversion processes have certain limits of acceptance with respect to amount and/or composition of the inorganic components in coal. A detailed knowledge on mineral matter is, therefore, imperative for proper evaluation and maximum use of coal reserves. However, studies related to mineral matter and associated trace elements are rather limited in India and more so in case of the high-sulfur Tertiary coals of the northeastern region (Mukherjee et al. 1982; Chandra et al. 1983; Nongkynrih et al. 1984; Mukherjee et al. 1992; Singh and Singh, 2006; Nayak et al. 2008). No previous study has been done with respect to 'mineral matter' that are associated with these coals. There is no universally accepted definition of the term 'mineral matter'. While some workers restrict the definition to include only discrete mineral grains, such as quartz or kaolinite that are contained within the organic matrix, others extend the definition to include, in addition to the discrete mineral grains, all elements except organically derived or bound C, H, N, O, and S (Renton, 1982; Ward, 2002). In the present investigation, we restrict our studies to mineral phases and sulfur content only.

GEOLOGICAL SETTING OF THE COAL SEAMS

The State of Meghalaya in northeast India contains coal deposits within Tertiary strata. The geological reserve of coal in the state is about 460 million tons (Indian Minerals Yearbook, 2008). However, unofficial sources indicate a total reserve of more than 600 million tonnes. The Jaintia Hills alone have about 40 million tons of coal. There are seven significant coal producing areas in this district where sporadic mining is going on. Of these, Bapung and Lakadong are the most important (Fig. 1). The others are: Musiang Lamare, Sutnga, Jarain, Ioksi, and Khliehriat. All these coal mining areas are grouped under 'Bapung coalfield' in the literature (Singh and Singh, 2000). The Jaintia and Eastern Khasi Hills expose a well developed sequence of the Lower Tertiary sediments and constitute the type area of the Jaintia Group, where, coal seams are associated with the Lakadong Sandstone Member of the Eocene Sylhet Formation (Raja Rao, 1981; Krishnan, 1982). A generalized stratigraphic sequence is given in Table 1.

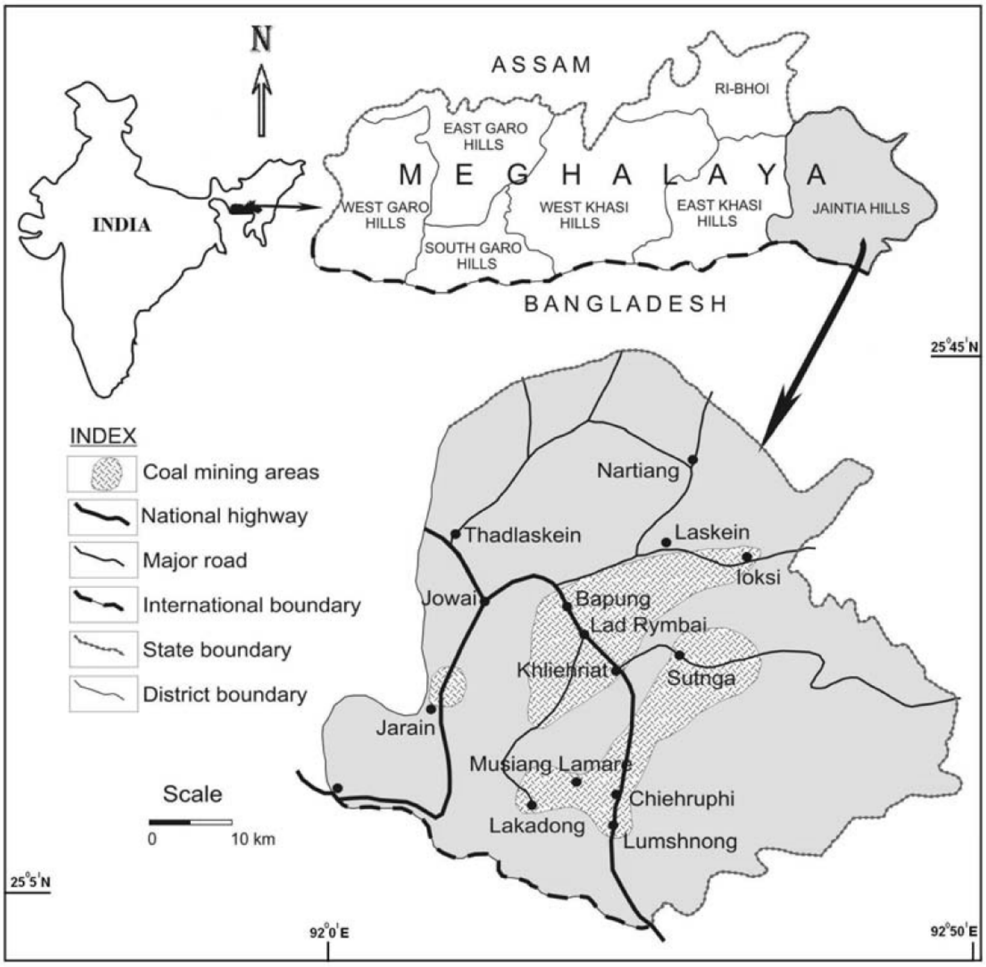

Fig.1. Outline map showing the coal mining areas in Jaintia Hills of Meghalaya in northeast India and the sample localities.

Table 1. Geological Succession around Bapung and Sutnga, Jaintia Hills, Meghalaya

Group	Formation	Rock-types	Age
Barail Group		Sandstones and shales	Oligocene
JAINTIA GROUP	Kopili Formation	Alternations of shales and sandstones with bands of calcareous sandsones and shales	Upper Eocene
		Prang limestone: Fossiliferous limestone	Middle Eocene
		Nurpuh sandstone: Sandstone with subordinate calcareous bands	
	Sylhet Formation	Umlatdoh limestone: Foraminiferal limestone containing a few sandstone bands	Lower Eocene to Upper Paleocene
		Lakadong sandstone: Sandstone with *coal-seams*	
		Lakadong limestone: Fossiliferous limestone	
	Therria Formation	Upper: Hard sandstones	Lower Paleocene
		Lower: Limestones and calcareous sandstones	
Mahadek/Langpar Formation		Boulder bed, sandstone, limestone, shale	Upper Cretaceous

Unlike the abundant Lower Gondwana coals of India that are interpreted to have been formed in rifts and have drifted origin and, therefore, contain high ash (Sharma and Ram, 1963; Acharyya, 2000), the Tertiary coals of Meghalaya are presumed to have been formed over platform areas under stable shelf conditions (Raja Rao, 1981; Singh and Singh, 2000). In the study area (e.g. Bapung, Fig.1), three coal seams have been recorded. The upper and middle seams are thin (0.1 to 0.6 m thick) and are impersistent. However, the lower seam is persistent and varies in thickness from 0.3 to 1.2 m. All the three seams are associated with the Lakadong sandstones striking in an ESE-WNW direction with south-westerly dips varying 4° to 7°. Bore-hole data of the Geological Survey of India indicates pinching and swelling nature of the seams even within a distance of 50 m (Raja Rao, 1981). The overburden thickness in Bapung area ranges from 3 m to 19 m. The coal seams near Khliehriat though slightly different in thickness than those of Bapung, appear to be extensions of the same seams. Two coal seams are also exposed near Sutnga with an intervening parting of 2 to 4 m. The lower seam occurs in the basal part of the Lakadong sandstone over the Lakadong limestone. These seams are well exposed in the valleys towards east and north of Sutnga village. The seams show a low regional dip of 3° towards south. Only one coal seam is recorded near Musiang Lamare.

MATERIALS AND METHODS

Bulk coal samples weighing ~50 kg each were collected from four different sources viz., Bapung, Khliehriat, Sutnga and Musiang Lamare. These were grab samples drawn from the heaps adjacent to the pits that were freshly excavated. The samples were collected after thorough mixing and following the conventional cone-and-quarter method (with a starting material of >1.6 tonnes). The samples at the pit-head were wet and moist. Though it is expected that the collected samples are representative of that particular locality from a geographical point of view, their exact underground location (from particular seam) cannot be traced back. The collected samples were air-dried in the laboratory and then each sample was crushed in two stages using jaw- and roll-crusher to below 6 mm size. About 500 g of head sample (representative split of the whole coal) was drawn from each sample following the conventional cone-and-quarter method and was analyzed for different parameters. Sulfur content was determined according to ASTM D2361-95. Proximate analysis was done using a coal analyzer (Anamed make; Model 490), where, fixed carbon was calculated by difference. One-fourth of the bulk sample was screened using standard Tyler-series screens, generating four size fractions (-6+1.65 mm, -1.65+0.42 mm, -0.42+0.15 mm, and -0.15 mm), and five density fractions (<1.4, 1.4-1.6, 1.6-1.8, 1.8-2.0 and >2.0) from each size fraction using heavy liquids (bromoform and benzene). All the size and density fractions were studied under a Leica zoom stereo-microscope to cross-check the consistency of screening and gravity separation. In contrast to the conventional approach for identifying minerals in coal by X-ray diffraction (after low-temperature ashing), we have used optical and electron microscopy to identify the minerals and show their mode of occurrence. In the case of X-ray diffraction, in some cases, minerals with low abundance are not detected. Therefore, polished sections were prepared on the higher density fractions (d =1.8-2.0, and >2.0) of all size-classes and studied under the optical microscope. Selected samples were studied under EPMA using facilities at IMMT (earlier RRL)-Bhubaneswar (JEOL Superprobe JXA-8100) and the Indian Bureau of Mines, Nagpur (Cameca SX-100), respectively.

Selected pyrite samples were investigated for sulfur isotopic studies at the Indiana University, Bloomington (USA). For sulfur isotopic analyses, sample powders and small amounts of V_2O_5 were loaded into tin cups and analyzed using Elemental Analyzer-Continuous Flow Isotope Ratio methodology (Studley et al. 2002). Samples were measured using a Finnegan MAT252 isotope ratio mass spectrometer. Analytical precision was better than ±0.05‰, whereas, sample reproducibility was ±0.2‰ (2 sigma). NBS-127 ($BaSO_4$, $\delta^{34}S$ = 20.3‰) and IAEA standards (S1 = -0.3, S2 = 20.8) were used as standards (values on the SO_2 scale). Sulfur isotopic compositions are reported in standard notation relative to Vienna Canon Diablo Troilite (VCDT).

RESULTS AND DISCUSSION

Mineral Phases and their Modes of Occurrence

Optical microscopic studies indicated that most of the discrete minerals in the Jaintia coals are fine grained (<50 µm) though, pyrite and marcasite occur as coarser grains that are visible to the naked eye. With the exception of massive occurrences of pyrite or marcasite, few mineral grains exceed 100 microns in size. A thorough scanning of the different size and gravity fractions reveals the presence of various mineral phases in the coals of Jaintia Hills, many of which are reported for the first time from the Tertiary coalfields of northeast India. The detected minerals are listed in Table 2. Some of the mineral occurrences have been described in Figs. 2 to 5.

Because of the autochthonous nature of the Meghalaya coals, most mineral matters are concentrated along bedding. However, some mineral components such as pyrite framboids, clays, gibbsite, and quartz are found disseminated throughout the coal matrix. Sphalerite (~35 µm, Fig. 2) and pentlandite (~10 µm, Fig. 3) occur either as inclusions or as intergrowths with pyrite. Nongkynrih et al. (1984) have reported high titanium content (2-4%) in coal samples from Bapung area. However, mode of occurrence of titanium is not reported. In the present case, Ti-bearing phase- rutile (<10µm) is recorded to occur in close association with Al-bearing phases – gibbsite and kaolinitic clay (Fig. 4). This Ti- and Al-bearing association perhaps hints towards a detrital origin of both rutile and clay phases. Gibbsite might have formed in a supergene lateritic condition. Pyrite framboids though are in close contact with clay, a displacement relation is not observed like that of Meigs Creek coal of Ohio where networks of interstitial kaolinite (having imprint of pyrite crystallites) were recorded within pyrite framboids and a displacive growth for pyrite was inferred (Scheihing et al. 1978). Gibbsite and clay occur as isolated patches (~100µm). Disseminated monazite (~35µm) is recorded within the parallel (syngenetic) layers of coal maceral (Fig. 5). Besides the minerals discussed above, isolated grains of quartz, barite, calcite, siderite, hematite, and mica occur scattered in the coal matrix. In the oxidized and weathered portions goethite, jarosite, and chlorite have been recorded. Mukherjee et al. (1992) have shown that Ni and Ba are associated with the organic matter of these Tertiary coals. But our studies reveal that some part of the Ni and Ba is inorganic affiliated.

Nature and Origin of Pyrite

Among the discrete mineral phases recorded, pyrite (and marcasite) dominates. It occurs in wide size ranges and in various forms. In hand specimens, recognizable morphotypes are: discrete grains (<1 to 2mm), nuggets (~3-4 mm), discoidal (~1-2 cm), massive (>1 cm). Sometimes the pyrite clusters are big enough to be separated manually. In the present case we have recorded massive pyrite of >3 cm in dimension. Under the microscope pyrite appears as: framboid, colloidal precipitate, colloform-banded, fine disseminations, discrete grains, massive, dendritic (feathery), recrystallized, cavity- fracture- and cleat fillings (Fig. 6). The average grain size for most of the microscopic discrete pyrite grains is ~50 microns. Earlier the present author has reported association of gold with the pyrites (Nayak et al. 2008). A maximum of 6 ppm gold has been recorded and it has been inferred that during processes of formation (viz. biological and/or inorganic recrystallization) gold has been captured from the coaly matter and adsorbed

Table 2. Minerals associated with the high-sulfur coals in Jaintia Hills

Groups	Sulfide	Sulfate	Oxide	Hydroxide	Phosphate	Carbonate	Silicate
Minerals	Pyrite Marcasite Sphalerite Pentlandite	Barite Jarosite	Hematite Rutile	Gibbsite Goethite	Monazite	Calcite Siderite	Quartz Mica Chlorite Kaolinite

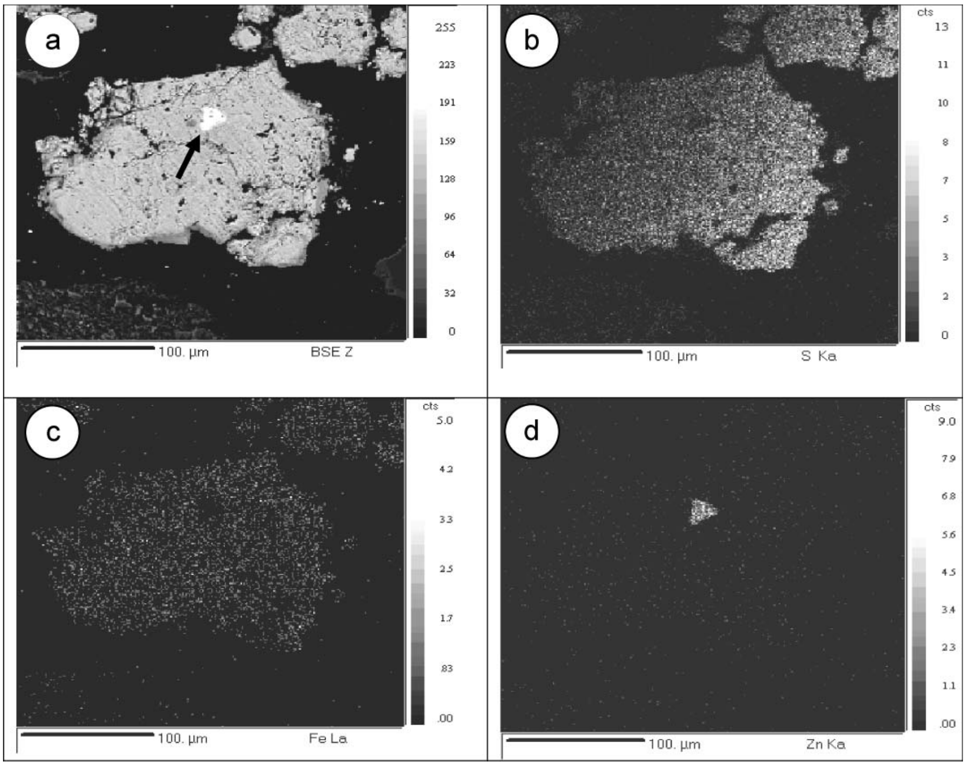

Fig.2. Back scattered electron image of pyrite in Bapung coal showing inclusion of sphalerite (arrow) **(a)**. Other images **(b)**, **(c)** and **(d)** are x-ray image maps showing elemental distribution of S, Fe and Zn respectively corresponding to the image (a).

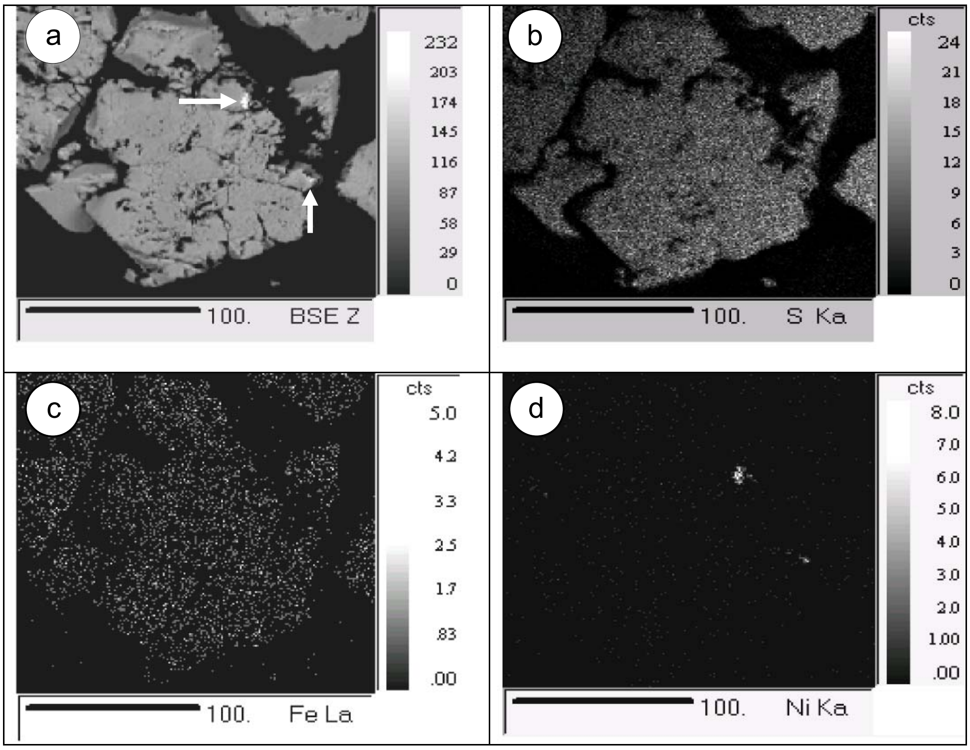

Fig.3. Back scattered electron image of pyrite in Musiang Lamare coal showing inclusion/intergrowth of pentlandite (arrow) within/ with pyrite **(a)**. Other images **(b)**, **(c)** and **(d)** are x-ray image maps showing elemental distribution of S, Fe and Ni respectively corresponding to the image (a).

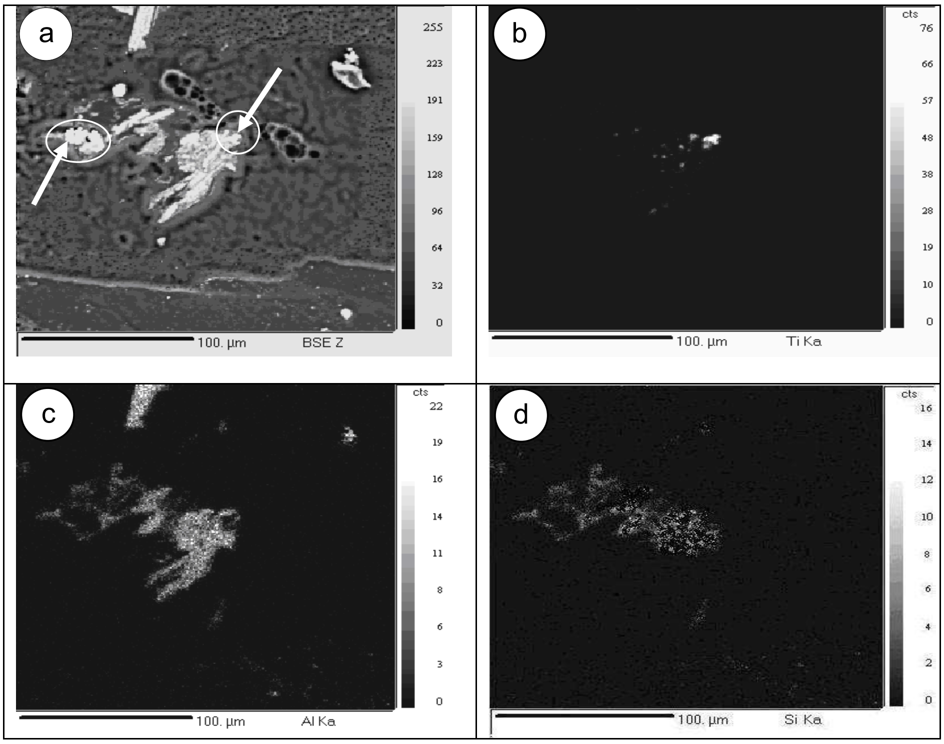

Fig.4. Back scattered electron image showing association of gibbsite (white elongated crystals), clay and rutile (right arrow, within circle) in Sutnga coal that inherits impressions of some bacterial activity **(a)**. White clusters within the ellipsoid (left arrow) are pyrite framboids. Other images **(b)**, **(c)** and **(d)** are x-ray image maps showing elemental distribution of Ti, Al and Si respectively corresponding to the image (a).

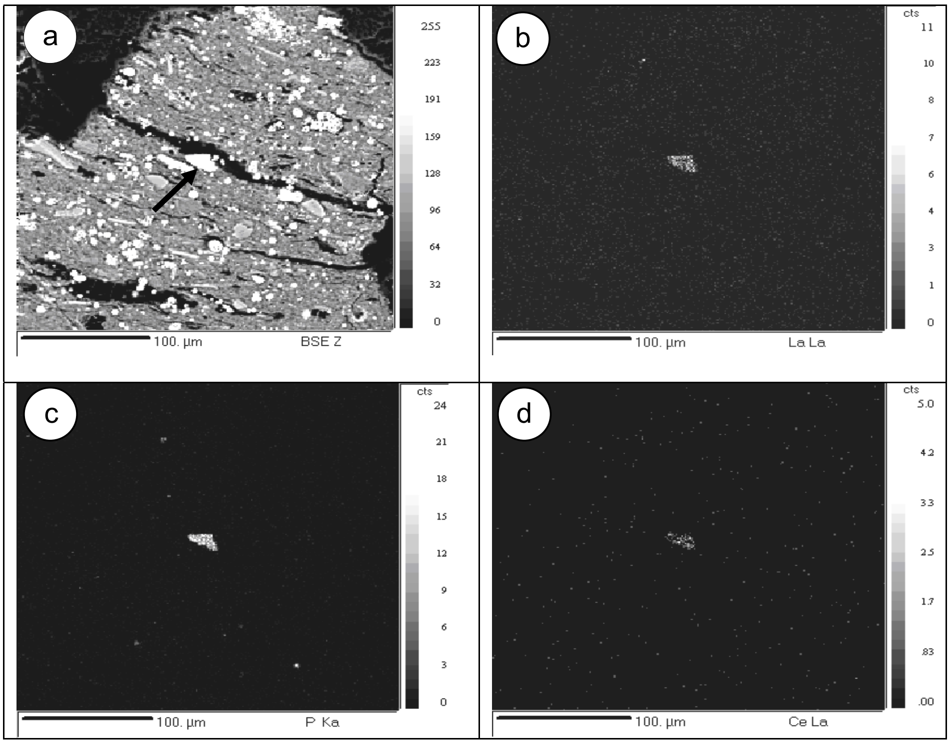

Fig. 5: Back scattered electron image of Sutnga coal showing numerous disseminated grains of pyrite (white) and a grain of monazite (arrow) **(a)**. Other images **(b)**, **(c)** and **(d)** are x-ray image maps showing elemental distribution of La, P and Ce respectively corresponding to the image (a). Traces of other rare earths such as Nd and Gd are also detected in the composition of monazite.

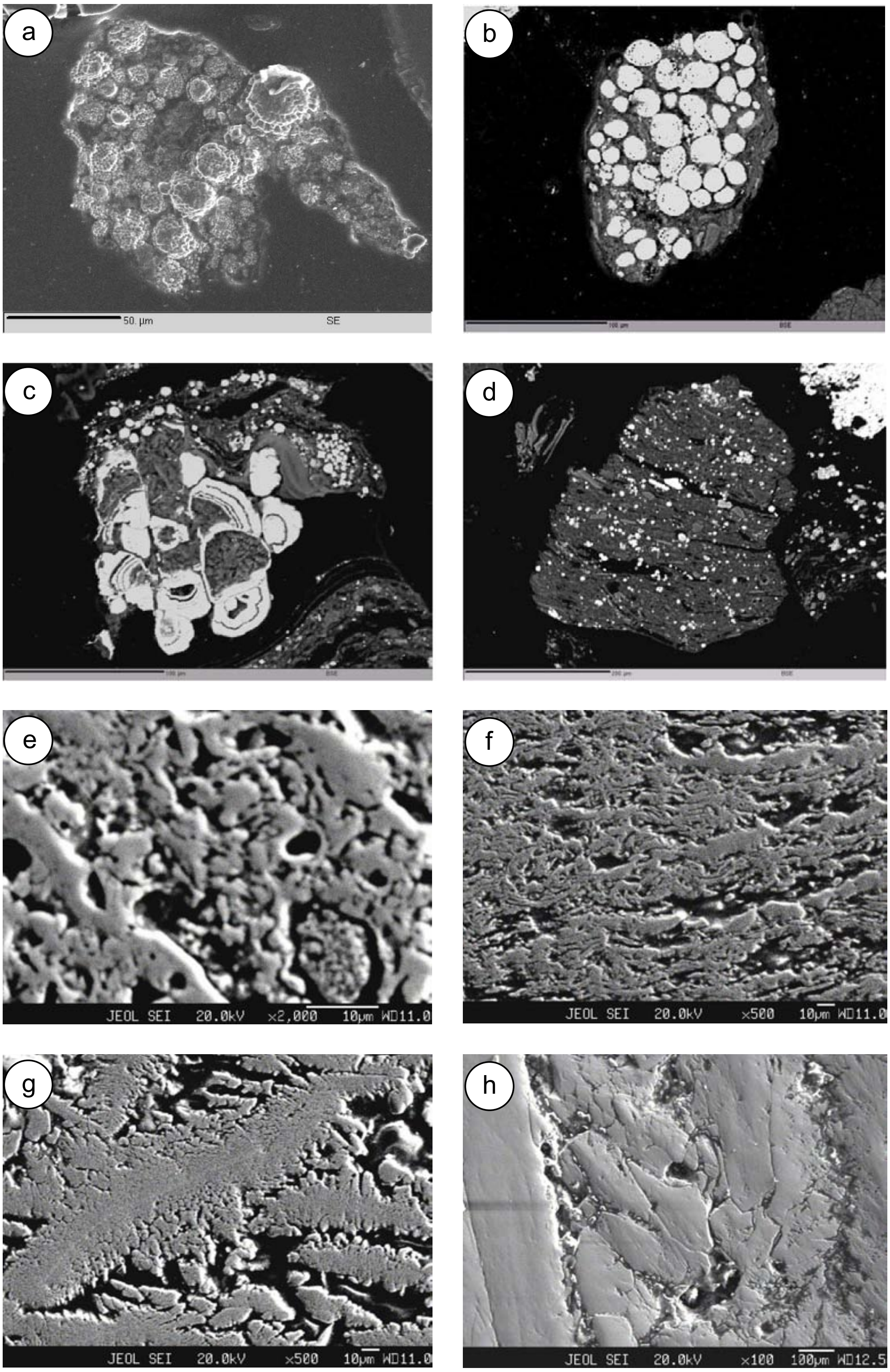

Fig.6. Photomicrographs of various pyrite forms: **(a)** fossilized bacterial colony showing honeycomb structure consisting of pyrite framboids in coals of Musiang Lamare; **(b)** foraminiferal tests (Nummulite?) in lenticular shape that has been replaced by pyrite in coals of Khliehriat; the other circular forms may also be replaced tests of foraminifera or simply fused framboids of pyrite; **(c)** colloform pyrite in coals of Khliehriat; **(d)** disseminated grains of pyrite in coals of Sutnga; **(e)** colloidal precipitates of pyrite in coals of Bapung; **(f)** banded/bedded pyrite in coals of Bapung; **(g)** dendritic/feathery pyrite in coals of Musiang Lamare; and **(h)** recrystallized marcasite in coals of Musiang Lamare.

into the structure of iron sulfides and the selective adsorption of Au into pyrite has depleted the gold content in the carbonaceous organic portion of the coal. The role of bacteria in capturing and incorporating gold into the pyrite structure was envisaged.

Pyrite is a very widespread mineral both in sedimentary and igneous rocks. Sedimentary pyrite generally forms during the stage of shallow burial by the reaction of detrital iron minerals with H_2S (Dai et al. 2002; Dai et al. 2006). The sources of H_2S may be due to the reduction of dissolved sulfate (SO_4^{2-}) that is derived from seawater by bacteria and/ or volcanic activity, organic compounds or other sulfate minerals (Robert, 1984; Hans et al. 1995). Stable sulfur isotopes may be used in distinguishing pyrites of different geological origins such as volcanic or biogenic. We have studied two pyrite samples for stable sulfur isotope analysis. Sample-1 was a disc-shaped pyrite (~2 cm in size) that consists of fine crystals of pyrite and sample-2 was a massive pyrite (~3 cm in size). The results obtained were -36.4‰ and -3.4‰ VCDT respectively. These are depleted values and such low values are generally characteristics of bacterial sulfate reduction. Sulfate reduction and pyrite formation are ubiquitous in modern marine sediments. Under anoxic conditions, seawater sulfate is reduced to H_2S by bacteria, which reacts with detrital iron minerals and ultimately forms pyrite (Robert, 1984; Hans et al. 1995; Dai et al. 2003). Thus this kind of pyrite is bacteriogenic and the $\delta^{34}S$ value of bacteriogenic pyrite is much lower than that of marine sulfate. Therefore, the sulfur isotope studies of our samples clearly indicate that these pyrites were formed by bacterial sulfate reduction.

Pyrite is the major end product of sulfate reduction and sulfate reduction is the major form of respiration in the salt-marsh ecosystem (Howarth, 1979). Pyrite in coal typically forms from H_2S and Fe in solution. The process involves bacterial reduction of SO_4 to H_2S at pH values of 7 to 4.5 followed by the combining of H_2S, elemental sulfur and ferrous iron oxide (FeO) to form pyrite and water. This is the only way pyrite can form in peats and low-rank coals. The SO_4 may come from seawater or vegetal matter, and iron may have been derived from the breakdown of clay minerals and is possibly carried in solution as stabilized organic colloids (Price and Shieh, 1979). In the present case during the peat formation and early humification stages pyrites may have been formed by two different processes: (1) inorganically due to 'diagenetic crystallization' from iron bearing sol/gel (eg. colloform-banded) and (2) biologically due to fossilization of bacterial colonies (e.g., framboidal). Though apparently there is no confusion on the inorganic derivation of some varieties of pyrite, the formation of framboidal pyrite has been controversial over the years. Despite its abundance in various natural occurrences and experimental investigations, the formation mechanism of framboidal pyrite has been debated (Wilkin and Barnes, 1997). Theories proposed for the formation of framboidal pyrite range categorically from 'biogenic', i.e., fossilization of bacterial colonies by iron sulfide (Schneiderhohn, 1923; Love, 1957), to inorganic, based on laboratory syntheses over a range of thermal conditions (Berner, 1969; Farrand, 1970; Sunagawa et al. 1971; Sweeney and Kaplan, 1973; Graham and Ohmoto, 1994). Indirect biogenic models suggesting formation of framboids due to the replacement of an organic spherical globule, or alternatively, a gaseous vacuole have also been proposed (Kalliokoski and Cathles, 1969; Rickard, 1970). Irrespective of whether biogenic or inorganic, it is now widely accepted that pyrite framboid generally forms from iron monosulfide precursor and the minerals mackinawite (FeS) and/ or greigite (Fe_3S_4) play important part in the process (Sweeney and Kaplan, 1973; Taylor, 1982; Morse et al. 1987; Schoonen, 2004). These two minerals can form by magnetotactic bacteria, and also by sulfate-reducing bacteria. In the studied samples we could not trace back the nature of the precursor mineral. However, petrographic and other evidences hint towards a biogenic origin of the pyrite framboids. Figure 6a and 6b clearly reveals bacterial activity in precipitating pyrite. Bacterial impressions have also been recorded in the coaly matter (Fig. 4a). In addition, the sulfur isotopic evidences support a biogenic origin for the pyrites of the Jaintia Hills coals. Bacterial origin for framboidal pyrite in coal has been supported by many (Altschuler et al.1983; Southam et al. 2001; Dai et al. 2002; Dai et al. 2003; Stachura and Ratajczak, 2004; Jiang et al. 2006).

From the various modes of occurrence of pyrite (and marcasite) and their petrographic characters it is quite evident that these di-sulfides have formed in three different stages. Framboid, colloidal precipitate, colloform-banded and fine disseminated grains in the larger part of the coal mass are syngenetic with the coal during peat formation and early humification stages. On the other hand the recrystallized, dendritic, nuggetty, discoidal and massive pyrites, though, syngenetic in nature have formed at later stages due to coalescence and recrystallization of the earlier formed pyrites of first generation. Cavity- fracture- and cleat-filled pyrites are of epigenetic nature and have formed much later due to remobilization and reprecipitation; but these are locally confined within the coal mass.

Sulfur Content of the Coals

Head samples drawn out of the bulk-coal samples were

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

Source	Total S	Pyritic S	Sulfate S	Organic S
Bapung (BP)	7.31	0.97	0.80	5.54
Khliehriat (KH)	6.48	0.86	0.61	5.01
Sutnga (ST)	5.92	0.69	0.53	4.70
Musiang Lamare (ML)	6.21	0.60	0.66	4.95

analyzed for their sulfur contents and the analytical results are given in Table 3. It is observed that these are very high-sulfur coals with total sulfur sometimes exceeding 7 wt.% out of which the organic sulfur content accounts for about 75% and the rest is inorganic sulfur (sulfide + sulfate). The sulfate sulfur in original coals at the pit-head may be slightly less than the results presented in Table 3, because there was a time-lag between sample collection at Jaintia Hills and sulfur analysis in the laboratory at Jamshedpur. We have also marked white stains developed on the surface of coal pieces indicating partial conversion of sulfides to sulfates due to oxidation. The high organic sulfur in these coals might indicate a syngenetic contribution from fluids with high sulfate content (Price and Casagrande, 1991; Querol et al. 1991; Dai et al. 2008).

Sulfur content in coal is thought to originate within the precursor peat environment of the coal (Casagrande et al. 1977; Chou, 1990) and high organic sulfur in most cases has been associated with marine influence (Gayer et al. 1999; Shao et al. 2003; Ward et al. 2007; Widodo et al. 2010). However, detailed basin analysis including paleoclimate, surface and ground water, and tectonic accommodation, in some cases indicate that high sulfur content in coal may not be solely a function of marine influence (Greb et al. 2002; Turner and Richardson, 2004). Organic sulfur might originate from complexing of sulfur from sulfate ions and hydrogen sulfide by humic acids during coalification (Casagrande and Nug, 1979). Irrespective of the form, such a high concentration of sulfur in coal clearly indicates a marine origin because fresh water contains only 0 to 10 ppm sulfur and therefore, even if there is prolonged circulation of fresh water through the peat, it cannot account for much sulfur in the coal. Various studies summarized by Chou (1990) indicate that most of the sulfur in coals with less than 1 % sulfur comes from the original vegetation. For coals with more sulfur, an increasing proportion comes from seawater. Based on sulfur isotope studies Price and Shieh (1979) have also indicated that for high sulfur coals (>0.8% sulfur), about 63% of the sulfur is derived from seawater by sulfate reduction and the rest is derived from original vegetation. If a swamp contains 40 % seawater, then this can account only for about 0.035 % introduced sulfur. Assuming a water-saturated peat can compact by a factor of 5 or more as it is transformed into bituminous coal and if all the sulfur from the water is taken up by the coal, then 0.035% sulfur in the peat could increase to a concentration of 0.2% sulfur only or slightly more in bituminous coal. Therefore, the high sulfur content of the Jaintia Hills can perhaps be explained by repeated influx of sea water with intermittent hiatus during which the swamp vegetation did not sink into a hydrological environment isolated from seawater. This corroborates the findings of Singh and Singh (2000) who based on the micro-lithotype composition of coals of Bapung area and using the facies diagram proposed by Hacquebard and Donaldson (1969) interpreted that these coals have formed in the forest moor facies of the telmatic to limno-telmatic zones. Since the coal seams of Jaintia Hills occur in the Lakadong sandstone member that is sandwiched between two limestone members: the underlying Lakadong limestone and the overlying Umlatdoh limestone, Raja Rao (1981) has suggested that these formations have formed due to intermittent marine transgressions and regressions during the Eocene period. In the absence of detail basin analysis and temporal factors, the influx of sulfur from other sources cannot be commented upon at present.

Ash Yield of the Coals

Proximate analysis was done on a moisture-free basis. The ash yield of the head samples varies from about 15 to 23 wt% (Table 4 and Fig. 7a) that are on a higher side than usually reported values by others (Sharma and Ram, 1963; Raja Rao, 1981; Indian Coals, 1982) and the fixed carbon is relatively low. As mentioned earlier, each coal was crushed and screened to four size fractions and from each size fraction five density fractions were generated. All the size and density fractions were subjected to ash-analysis. It was found that in case of each coal, the four size fractions did not deviate much in their ash yield except in the finest size (i.e., -0.15 mm). This indicates that the mineral matter is getting liberated and concentrated below 0.15 mm in a normal crushing and screening operation. From Fig. 7b, it is evident that the crushing characteristics of the four coal samples are more or less similar generating higher quantity in the size class -1.65+0.42 mm. In case of Bapung this size

Table 4. Proximate analytical data of coal samples (in wt.%) from Jaintia Hills

Source	Moisture	Volatile	Ash	Fixed Carbon
Bapung (BP)	1.04	38.94	16.45	44.61
Khliehriat (KH)	1.69	38.75	14.56	46.69
Sutnga (ST)	0.95	37.41	23.04	39.55
Musiang Lamare (ML)	1.26	36.13	16.22	47.65

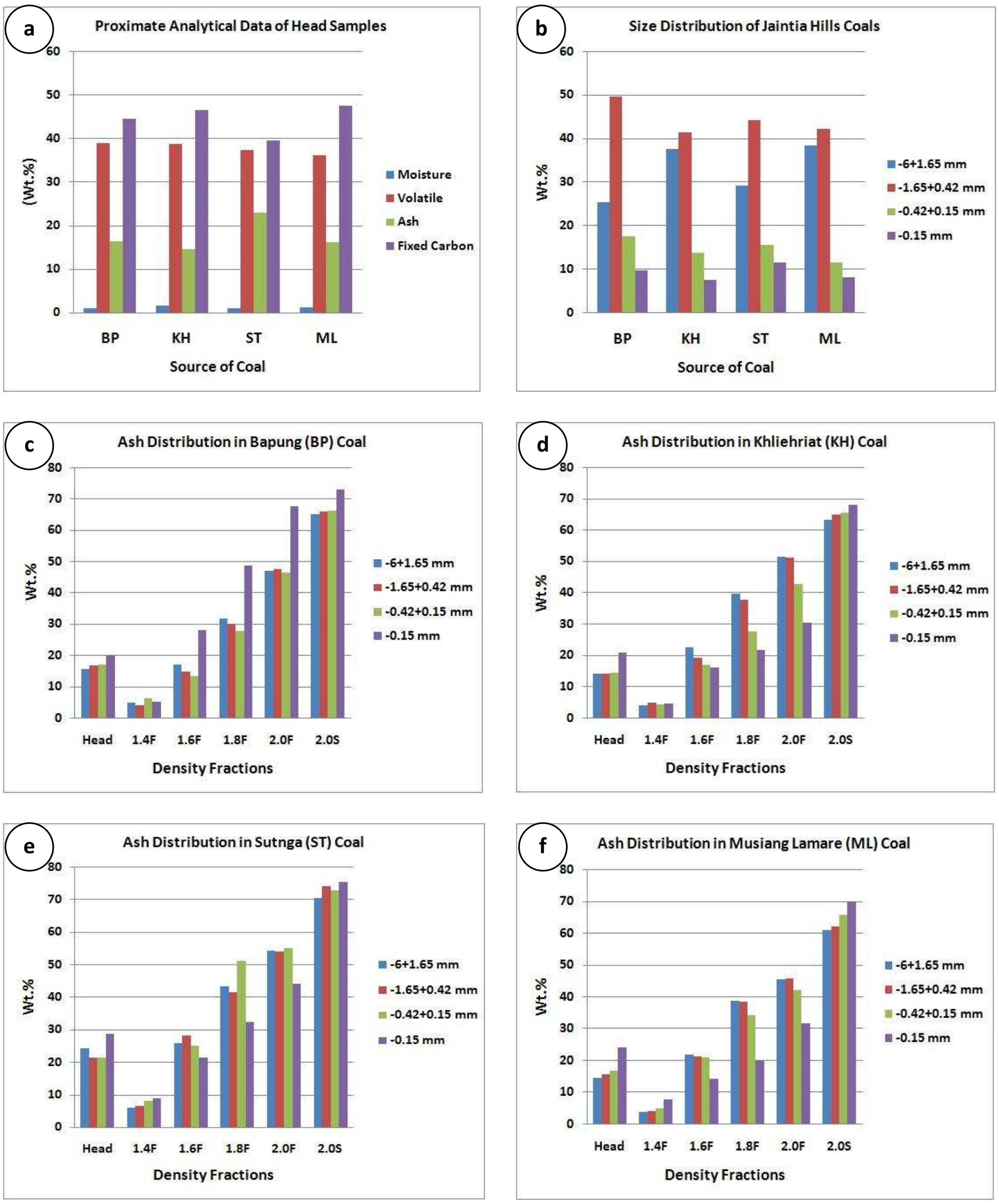

Fig.7. **(a)** Proximate analytical data of the four coal samples; **(b)** Size distribution of the four coal samples in a normal crushing and screen analysis; **(c)** to **(f)** Ash distribution in different size and density fractions of Bapung, Khliehriat, Sutnga and Musiang Lamare respectively.

fraction is quite high (~50 wt%). The finest size fraction (-0.15 mm) is always lowest accounting to around or less than 10 wt%. A critical observation through various density fractions indicates that the Bapung coal has slightly different pattern (Fig.7c to 7f). The -0.15 mm fraction of Bapung in all the density classes (except d<1.4) shows higher ash yield relative to other size fractions. This indicates that though the mineral matters are concentrated in -0.15 mm size, still they are not liberated properly in the case of Bapung. In other coals the trend is reverse indicating a good liberation of mineral matter below 0.15 mm.

CONCLUSIONS

High organic sulfur content of the Tertiary coals of Jaintia Hills indicates their marine origin and the formations associated with the coals have witnessed both marine transgression and regression phases that may be related to

Himalayan tectonic events during the Eocene. Iron sulfides (mostly pyrite and very less marcasite) are detected as the major mineral impurities in these coals. Pyrite in three generations though occurs in various modes and morphology, its origin is dominantly biogenic. Cleaning of these coals shall not be easy because majority of the microscopic mineral phases are ~ 50 mm in size. While some pyrites that are present in the bedding planes of coal can be liberated relatively easily, most of the pyrite framboids and blebs of kaolinite that are found disseminated throughout the coal matrix cannot be removed by conventional coal-cleaning processes and will remain part of the cleaned coal.

Acknowledgements: This article is an outcome of CSIR Network Project No. CMM-0012 during the Xth five year plan. The author is thankful to the Director, NML for permitting to publish this paper. Mr. K. K. Bhattacharyya and other colleagues of MNP Division had supported during the project work. I am grateful to Prof. Edward M. Ripley of Indiana University, USA for providing the sulfur isotope data. Dr. NVC Rao of IBM, Nagpur and Dr. B. K. Mohapatra of IMMT, Bhubaneswar had extended electron microprobe facilities. The constructive comments of Dr. Shifeng Dai and an unknown reviewer have significantly improved the quality of the present paper.

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(Received: 13 September 2011; Revised form accepted: 31 January 2012)