Chapter I

General introduction

1.0 Introduction to coal

Coal is a non-renewable energy source formed from the decaying dead plants which were died about 100-400 million years ago. Among all coal producing countries, India is the second largest coal producer in the world (Statistical Review of World Energy, 2017) and it is the fourth largest country in terms of world coal reserves (Statista, 2019; Rai, Paul, and Singh, 2010). Out of the total India's coal reserves of about 220 billion tons, only 0.90 billion tons occur in the North Eastern region which is about 0.4% of the total coal reserves of India. By detecting the depth, thickness as well as the configuration of minerals resource present in coal, the mining technology (open cast or underground) can be selected. Depending upon mode of formation of coal, fixed carbon limits, volatile content, and gross calorific value it is classified into different types (rank) as per ASTM standard (ASTM D388-12). The term 'rank' is used to refer the steps in a slow, natural process of coal formation called coalification (USGS, 1983; Coal and Peat Fires, 2011). During the process of coalification, buried plant matter changes into an ever denser, drier, carbon rich, and harder material. The classification of coal is generally based on the content of volatiles (Standards catalogue 73.040 – Coals, ISO*)*. The coals are most generally classified by rank that depends upon the degree of transformation from the original source like decayed plants and therefore is a measure of a coal's age (Green and Perry ,1997; Kreith, 1998). International Organization for Standardization (ISO) classification system divides coals into three main categories, namely low rank, medium rank, and high rank. This classification of coals into the primary ranks and subcategories is on the basis of the parameters are vitrinite reflectance, vitrinite content, moisture, and ash yield (ISO Standard 11760). The four ranks of coal are: anthracite, bituminous, sub-bituminous and lignite. Out of these four categories, anthracite is the highest ranking coal, bituminous is the middle rank coal between subbituminous and anthracite , subbituminous coal has black colour with a dull appearance and it has higher heating value than lignite . Lignite coal is the lowest grade coal with a brown colour and it has the lowest concentration of carbon. With decrease in the moisture content and volatile matter and increase in the fixed carbon results in increase in the rank of the coal

(USGS, 1983). Higher is the ranking higher will be the heating value of coal. The Figure 1.1(a), 1.1(b), 1.1(c) and 1.1(d) show the pictures of different types of coal found in India and other coal producing countries.

Figure 1.1(a): Anthracite

Figure1.1(b): Bituminous

Figure1.1(c): Subbituminous

Figure1.1(d): Lignite

The coals from Northeast India are subbituminous form (Baruah et al., 2006; Mukherjee and Borthakur, 2003; Chabukdhara and Singh, 2016). Subbituminous type of coal has numerous applications including power generation and industry. In Northeast region (NER) of India the coal deposits occur only in five states namely Assam, Arunachal Pradesh, Meghalaya, Nagaland, and Sikkim (Energy and Energy Resource Management,1999). According to the reports of Department of Mines, Minerals and Geology, Government of Sikkim, Gangtok (2014), there are coalfields in Sikkim, but coal occurrences are widespread in the ' Rangit Tectonic Window' zone. Also, the Geological Survey of India (G.S.I.) studies around Namchi, South Sikkim, established that a reserve of 1.40 Lakh tons of coal is found in this zone. Coal is composed primarily of carbon, along with various amounts of some other elements, mainly hydrogen, oxygen, sulfur, and nitrogen (Blander, 2011). The NER coals have high sulfur content and low ash content and most of its sulphur is present as organic sulfur (Chandra et al.1983). This organic sulphur is formed due to the influence of marine matters during coalification (Ward et al., 2007; Widodo et al., 2010). Coal can be classified as low sulfur, medium sulfur, and high sulfur coals based on their sulfur contents within the range < 1wt%, 1-3 wt%, and □ 3wt %, respectively (Chou 2012).

Although the NER coals possess high sulphur contents (3-8 wt %) (Barooah and Baruah, 1996; Ali et al., 2009, Saikia et al., 2015a), but the sulphur content in NER coal differ from one colliery to another. In NER of India, Assam is recognized as the one of the major coal producing states which has an enormous deposition of high sulphur Tertiary rank of coal (Saikia et al., 2009). Assam coals mainly present in Makum coalfield of Tinsukia District contains 2.7-7.8 wt % sulphur (Barooah and Baruah, 1996). The coals from Meghalaya possess 1.61-8.92 wt % sulphur content (Chandra et al., 1983). The average sulfur content in Nagaland coals is 6.9 wt % (Singh et al., 2012). Coals from Arunachal Pradesh is also reported to have high sulphur content (1.23-4.84 wt %) (Chandra et al.,1984). On the basis of the age of coal formation, coal reserves in India can be classified into two classes – Gondwana and Tertiary. Gondwana type of coal is mainly found in the river basins of the Damodar, Mahanadi, Godavari, and Wardha and most of the coal of India belong to this type. The Tertiary coal is younger in age. The Tertiary coals are located in Assam, Meghalaya, Nagaland, and Arunachal Pradesh (Qaisar and Ahmad, 2014; Chabukdhara and Singh, 2016; Chandra et al.,1983 ; Singh et al.,2012). The deposition of subbituminous Tertiary type of coal in Northeastern India was found to be due to the influence of marine environment (Rajarathnam et al. 1996). Due to the high sulphur content the Assam coal is not useful for metallurgical as well as power plant operations. The Assam coals was termed as abnormal coals (Iyenger et al.,1959) and the abnormal behaviour of these coals is attributed to its high sulphur content and other physico chemical parameters. The distribution pattern of sulphur in Northeast Indian coals was reported by several workers (Chandra et al.,1980; Mukherjee,1984; Barooah and Baruah,1996; Chou, 2012; Calkins, 1994). The upper coal seams contain more sulphur content than the lower seams (Chandra et al., 1983). Sulphur is present in coals mainly in three forms namely, Pyritic, organic and sulphate sulphur (Barooah and Baruah, 1996). But Baruah and Gogoi (1998) reported that sulphur in coal can be present in five forms namely pyritic, sulphate, organic, elemental and secondary sulphur. Out of these five forms, elemental sulphur is not found in Assam coals (Baruah, 1992). The major part of inorganic sulphur present in coal consists of pyritic sulphur (FeS_2). The pyritic sulphur in coal can occur both in microscopic as well as macroscopic forms (Baruah et al., 2006). Coal mining process is recognized as very essential process for economic growth of a country and also the power sector is the largest consumers of coal from last four decades (Chaulya and Chakraborty, 1995). Apart from this positivity, the coal mining leads to extensive environmental degradation and it should be taken with an equal importance because it creates an enormous amount of adverse and hazardous impact on soil, and water resources (ground and surface water) by destroying human being specially the health condition of mine workers and the habitant rendering near the coal mine areas (Sahoo and Bhattacharjee, 2010) and aquatic life system (Tiwary, 2001). Coal mining imparts a long term effect on the landscape and ecosystem also (Baruah, 2010; Giri et al., 2014).

The environmental problems associated with the coal mining in Northeastern India (before , after, and during mining), coal storage , coal transportation, and production of waste products and their health impact on the human beings and other life systems has been already discussed by many research workers (Finkelman et al. 2002). The adverse effects resulting from coal mining are due to mine waste and dust generation, land subsidence, deforestation etc. (Eqeenuddin Md. S.K., 2013). It is experimentally found that the underground land mining is very expensive and because of the soil condition of Assam, open cast mining has been adopted as the main method for coal mining over underground method. But the open cast coal mining destroys the environment, leaving large scars instead of fertile paddy fields and forests. Neighboring areas are also polluted with large quantities of overburden (OB) as the opencast mining involves the displacement of large volumes of OB as mine waste to excavate coal from the earth (Nand et al., 2015). The OB gets washed away by rains water, joins streams, and drains and ends up at the large water resources like river causing severe water pollution. Despite these problems, opencast mining has been accepted at a phenomenal rate in India and other coal producing countries like Australia, Canada, Russia, Spain, New-Zealand, South-Africa etc. due to its increasing energy demand (Ghose, 2004; Kumar, 1995). Also, some problems created in case of underground mining have reduced in opencast mining. Open cast mining is principal mining process in India, which contributes about 85% of total coal production, while underground mining system contributes only15% (TERI, 2001; Gupta and Paul, 2015). If the opencast mining is not planned properly it may result in a number of environmental degradation in and around the mining area. The environmental effects of mine waste from open cast mining include release of potentially hazardous elements (PHEs); air, water, and soil pollution; expose of sulphide minerals causing acid mine drainage. Also, drastic change of physico-chemical properties of land as a result of environmental degradation (Prasad et al., 2016), Overburden is the top soil consists of stone particle which is laid above the coal seams which are removed from coal during mining (Maiti et al., 2001). These dumped OB are left over the land. It occupies a large amount of useful land, which loses its originality results in degradation of soil qualities (Barapanda et al., 2001). Thus in opencast mining, the unwanted by-products of mining activities carry a high risk of contaminating the nearby water resources. During open cast mining, enormous amount of the mine OB wastelands are generated. Also, OB contains an elevated concentration of trace and heavy elements (Chandra and Jain, 2013).

OB dumps are manmade problems causing manyfold environmental problems including erosion and increasing sediment load in receiving water resources, dust pollution etc. The OB dumps are responsible for changing the natural land structure. It also disturbs the natural drainage system and prevents natural development of plant growth (Bradshaw and Chadwick, 1980; Wali, 1987), creating long term problems of soil erosion as well as the environmental pollution (Singh et al., 1996) In India, a large area of mine OB wasteland is generated annually due to open cast mining which are dumped on nearby area of mining site. From literature (Gupta and Paul, 2015), it is found that the Northeastern coalfields of Coal India Limited (NECF-CIL), Margherita, Assam, have generated > 1000 ha of mine OB wasteland. The major problem arises from OB dump is the formation of acid mine drainage (AMD). The physico-chemical characteristics of OB dump materials are regionspecific which differ from one dump to another dump because of deposition of rocks under different geological environment (Lovesan et al., 1998). Mine OB creates an adverse condition for the microbes present in soil. Also, it effects in the plant growth due to its low concentration in organic matter and unfavorable and poor soil structure (Rai et al, 2010).

1.1 Major types/grades of coal found in Margherita

Coal is one of the significant natural resources of Assam (Northeast India). The major coal reserves of Assam are found in two coal belts: the Makum coalfields and the Dilli-Jaipore coalfields. Excluding them, some small coal deposits exist in the Karbi Anglong, North Cachar Hills, and Dhubri districts of Assam. It is found that Makum coal of Assam has about 453 million tons of coal reserves (Coal Directory of India 2012-13). The coal from Makum coalfield of Assam has high calorific value, low ash content and high sulphur and low oxygen content. As per ASTM standard (ASTM D388-12) classification of coal, the Makum coals are categorized under the categories of sub-bituminous A to high volatile bituminous B in rank which is basically a hydrocarbon rich rock. The Tertiary coal are found in NER India, distributing over the states of Assam, Arunachal Pradesh, Nagaland, and Meghalaya among which the Assam Coal fields are the protruding ones (Coal directory of India 2013–2014). The NER coals are subbituminous Tertiary coals which were deposited under the influence of marine environment (Rajarathnam et al.,1996). These coals are characterized by high sulphur and low ash content. Most of the sulphur present in these coals is organic sulphur because of the influence marine materials during its formation by physical and chemical alteration of sedimentary rocks (Chandra et al., 1983; Ward et al., 2007; Widodo et al., 2010). NER of India contains total of 105 million tons of Gondwana coal and 1492 million tons of tertiary coal reserves (India's inventory of coal resource, 2014). The tertiary coal is 15 to 60 million years old where carbon content is very low. In Northeast India, Meghalaya and Assam contain 73% Tertiary coal and Nagaland

and Arunachal Pradesh contain 21% and 6% tertiary coal from the total coal reserves. The coals of Tiru valley, Nagaland (India) have subbituminous to bituminous-D rank. These coals are characterized by high sulphur content (5-11 wt %), medium moisture content (4-7%) and moderately high volatile matter content (22-42%) (Singh et al., 2012a, 2012b).

1.2 Coal resources and its distribution in NER India

Coal India Limited (CIL) and its eight subsidiaries are incorporated under the companies Act, 1956 and are wholly owned by the Central Government. The coal mines in Assam and other Northeastern states of India are controlled directly by CIL under the unit Northeastern coalfields. The Tertiary coals are found in Northeastern part of India. In Assam, it is distributed in Makum, Nazira, Mikir Hills, Dilli-Jaipore and Lakhuni among which Makum coalfield of Tinsukia district, Assam, is the largest coalfield in Northeast India. Assam coals comprise with very low ash content, and high coking potentials. Moreover, the sulphur content is high, as a result of which this coal is not suitable for metallurgical purposes but these coals are very suitable for hydrogenation process and for producing making liquid fuels. The upper Assam coal belt extends towards east as Namchick-Namrup coalfield in Arunachal Pradesh. These coals are high in volatiles and sulphur. In Meghalaya, these tertiary coals are dominated on Garo, Khasi and Jaintia Hills.

As per Geological Survey of India (GIS) Coal Inventory (5^{th} January, 2018), Coal resources are found in Northeastern region (NER) of India (Table 1.1).

Table 1.1: Coal resources in NER of India

Sl. No.	State	Coalfield	Resource (MT)
1	Assam	Singrimari	14.49
2		Makum	452.79
3		Dilli-Jeypore	54.02
4		Mikir Hills	3.71
5	Sikkim	Rangit Valley	101.23
6	Arunachal Pradesh	Namchik-Namphuk	84.23
7		Miao Bum	6.00
8	Meghalaya	Balphakram-Pendenguru	107.03
9		Siju	125.00
10		Mawlong-Shella	6.00
11		Bapung	33.66
12		Jayanti Hills	2.34
13		West Daranggiri	125.00
14		East Daranggiri	34.19
15		Langrin	133.16
16		Khasi Hills	10.10
17	Nagaland	Borjan	10.00
18		Jhanzi-Disai	97.12
19		Tuen Sang	3.26
20		Tiru Valley	6.60
21		DGM Area	293.47

MT: million tons

1.3 Present scenario of coal utilization

In the past five decades, the developments in the production of primary commercial energy indicate that coal is the most abundant resource among all commercial energy sources (TEDDY, 2004-2005; Energy and Resources Institute, 2006). Coal is responsible for around 30.3% of the world wide primary energy needs and it generates 42% of the world's electricity. Since 2011, coal was one of the fastest growing source of energy after renewable sources (The Indian coal sector, 2012) India is the 5th largest electricity producing country and is the 6th largest energy consumer accounting for 3.4 % of world total energy consumption (Kumar et al., 2014).Upto the year 1970, the increasing demand of coal was due to the rapid growth of thermal power plants. During these years, about 13 million tons of coal was consumed in electricity production which was about 20% of total consumption whereas during the period of 2009-10, it was consumed about 411.06 million tons which was nearly 75% of total power consumption (India Energy Book, 2012). 68% of coal is consumed in electricity production all over the world whereas in India, power sector is mainly based on coal (55%) (Kumar et al., 2014). In India, the demand and consumption of coal have grown enormously which is primarily dominated by the electricity sector. Coal utilization in U.S industrial is found to be mainly in two kinds: one in coke plants which produce high-purity carbon for steel production and the other in coal combustion that provide heat and power generation for industrial purposes (Coal Research and Development, 2007). Since 1970, the demand for coal has increased due to the rapid installation of thermal power plants. About 13 million-tons of coal is consumed in electricity generation in 1970-71, which is about 20% of total consumption whereas in the year 2009-10, it is consumed about 411.06 million tons which is nearly 75% of total consumption (India Energy Book, 2012). Other major coal-consuming sectors include iron and steel production, cement production, chemical, base metals, cookeries, paper& pulp, textile& rayon, bricks, etc. The iron and steel industry, which primarily consumes coking coal and some high-grade non-coking coal, is the second largest consumer of domestic coal, although its consumption has decreased from 19% of total consumption in 1970-71 to about 7% in 2009-10. The third largest consumer of coal in

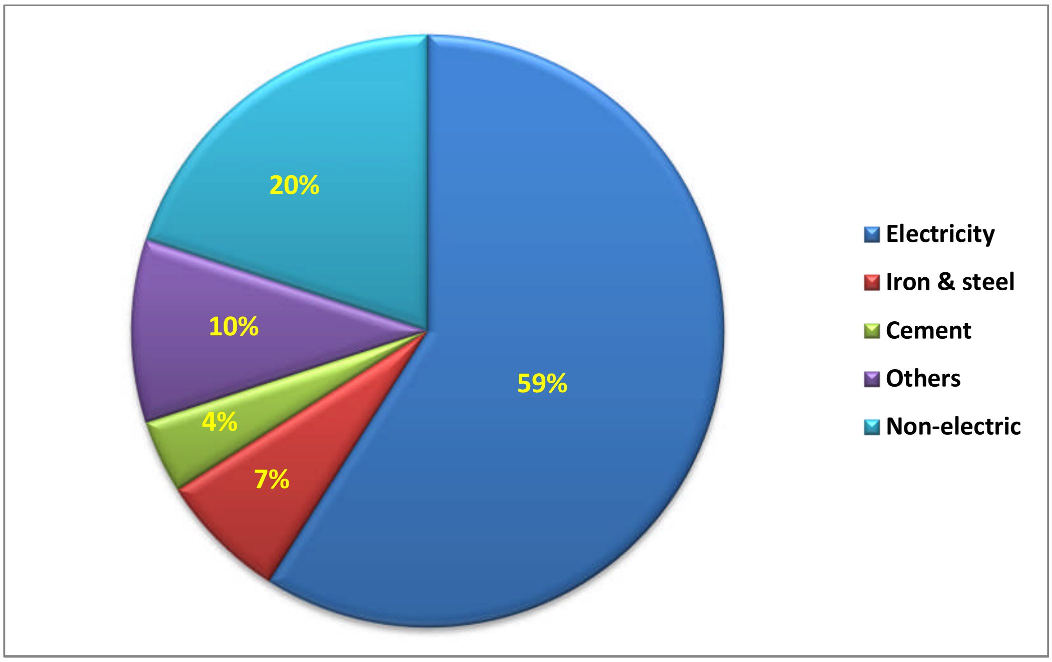
India is the cement industry, which accounts for 4 to 5% of total consumption. Other smaller consumers include the fertilizer industry, the textile industry (including jute and jute products), the paper industry and the brick industry. India is the fifth largest energy consumer in the world which is responsible for production of 4.1% of the global energy consumption. Maharashtra is the foremost state in case of electricity generation. The current per capita consumption of energy in India is 0.5 toe (ton of oil equivalent, an unit of energy) over the global average energy consumption of 1.9 toe, which indicates that India has high potential for growth in the energy sector. In India, out of the total electric energy consumption, approximately 80% is generated from coal (The Indian coal sector, 2012). Coal is assumed as an important input in the production of steel. It was found that, in the year 2011, the world steel production reached 1,518 million tons which reflects a growth of 6.2% over the year 2010. The key raw material for the production of steel is the cooking coal which is very limited in India. Coking coal is found in only 15% of the India's overall recognized coal reserves. Due to this, the Indian steel industry has been facing an serious scarcity of coal for the last several years. The coal in Jharia coalfield of Jharkhand is found to hold the major part of the coking coal reserves. As per the report, the steel production by 2016-17 was projected to be 105 million tons, but the conforming requirement of coking coal for this quantity of steel was 67.2 million tons in 2016-17 (Indian Minerals Yearbook 2017). Apart from these, major utilization of coal, Significant quantities are also utilized by the railways, and as a domestic fuel (Indian Minerals Yearbook 2017). The coal consumption/ utilization in India from the year 2013-14 to 2015-2016 is given in Table1.2 (Coal Directory of India, 2014-15 and 2015-16; Provisional Coal Statistics, 2016-17).

Table1.2: Consumption of coal in different industries (in million tons)

Industry	2014-15	2015-16	2016-17
Iron and steel	12.34	12.36	12.50
Sponge iron	12.05	7.76	5.68
Fertilizer	2.29	2.30	2.14
Cement	11.06	8.98	6.43
Electricity	485.95	502.28	527.26
Others*	80.08	98.76	96.31

*Chemical, base metals, cookeries, paper and pulp, textile and rayon, bricks, etc.

Sector wise percentage of coal consumption in the year 2016-2017 is described by the Figure1.2



Others: Chemical, base metals, cookeries, paper and pulp, textile and rayon, bricks, etc.

Figure 1.2: Sector wise coal consumption (2016 -2017)

1.4 Geological settings of Ledo open cast mining area

Ledo colliery where open cast mining takes place is located in Makum coalfields, Margherita in Tinsukia District of Assam. The Makum coalfield extends from Naga patkai hills which ranges from Sibsagar (Assam) to Noa Dihing (Arunachal Pradesh) at a distance of about 250 km. and lies between latitude 27°15´ and 27°24´N and longitude 95°40´ and 95°59´E covering an area of about 100 sq.km. It is the largest Tertiary coalfield in India and consists of two opencast (Tikak and Tirap) and three underground (Baragolai, Ledo, and Tipong) coal mines The Makum coalfield is located in Tinsukia district, Assam. Mining of coal in the Makum Coalfield was first started by the Assam Rail-ways & Trading Company at Ledo Colliery in 1882. Total reserve of 280.69 million ton was estimated by the Northeastern coalfield, Coal India Ltd. The Makum coalfield contains five workable coal beds. Out of these, two are persistent across the coalfield whereas others are sporadic in their occurrence. The Tertiary coal of this coalfield is very friable in nature. Seams are highly gassy and liable to spontaneous heating. These coals are characterized by low moisture (2-5%), low ash (3-10%), and highly volatile matter (>40%) and high sulphur content (2-6%). In Makum coalfields, four coal seams known as 5 feet, 8 feet, 20 feet, and 60 feet seam occurs. Out of which 20 feet and 60 feet seams are persistent and are being worked.

. The satellite picture of ledo open cast mine collected from Google Earth shows the proper location of the mine area (Figure 1.3).

Figure 1.3: satellite picture of Ledo open cast mine

1.5 Generation of acid mine drainage (AMD)

Acid mine drainage (AMD) is the outflow of acidic water from coal mine (Vyawahre and Rai, 2016). A numerous studies (Kalin et al., 2006; McDonald et al., 2006; Saarinen et al., 2013; Dold, 2014) evidenced that AMD generation has been widely accepted as one of the major environmental problems created by coal mining industries worldwide. AMD results from the oxidation of sulfide minerals mainly pyrite which is characterized by low pH (high acidity) with high concentrations of SO_4^{2-}, Fe, metalloids, and different metals and non-metals (Larsen and Mann, 2005; Envis Newletter, 2005; Baruah et al., 2005; Equeenuddin et al.,2013. The sulphide oxidation associated with AMD generation along with the dissolution and precipitation of metals , non-metals and minerals, has been a major attention of investigation over the last 50 years (Nordstrom, D.K., 1982; Jambor, 1994) Minerals like pyrite (FeS_2) and pyrrhotite (S(x=0-0.2) and some microorganisms like *Leptospirillum ferroxidans*, *Thiobacillus ferroxidans* etc. (Johnson, 2003) are responsible for the generation of AMD which are stable and insoluble in absence of water and atmospheric oxygen(O_2) (Cruz and Monroy, 2006). The chemical process of AMD formation can be given in different steps as follows:

1. Oxidation of FeS_2

   $$\mathrm{FeS} \ + \frac{7}{2}\mathrm{O} \ + \mathrm{H} \ \mathrm{O} \rightarrow \mathrm{Fe} \quad + 2\mathrm{SO} \quad + 2\mathrm{H} \ \ldots\ldots\ldots \ (1)$$

2. Oxidation of ferrous ion (Fe^{2+})

   $$\mathrm{Fe} \quad + \frac{1}{4}\mathrm{O} \ + \mathrm{H} \quad \rightarrow \ \mathrm{Fe} \quad + \frac{1}{2}\mathrm{H} \ \mathrm{O} \ \ldots\ldots\ldots\ldots\ldots \quad (2)$$

3. Hydrolysis and precipitation of ferric complexes and minerals

   $$\mathrm{Fe} \quad + 3\mathrm{H} \ \mathrm{O} \ \rightarrow \mathrm{Fe(OH)} \ (\mathrm{S}) + 3\mathrm{H} \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

Equation (1) refers the initial step of pyrite (FeS_2) oxidation in the presence of atmospheric oxygen. The presence of SO_4^{2-} in mine drainage is the first indicator of pyrite mineral oxidation (Lapakko, 2002), in an available oxidizing condition which is entirely dependent on atmospheric O_2 concentration, pH (□ 3.5) and microbial activity. Fe^{2+} released in Equation (1) is oxidized to Fe^{3+}according to the Equation (2) (Blowes et al., 2003; Akcil and Koldas, 2006). If the concentration of O_2 is low then the Equation (2) will not occur until the pH of the mine water has the value of 8.5 (Fripp et al., 2000). Generally, under various conditions, Equation (2) is considered to be rate limiting step in the pyrite oxidation process because the transformation of Fe^{2+} to Fe^{3+} ion is a slow process at pH value of less than 5 under abiotic conditions (Skousen et al., 1998). When pH values of the mine water lie within the range 2.3-3.5, then the Fe^{3+} ion formed in Equation (2) can precipitate as $Fe(OH)_3$ (and to a small amount of jarosite, $[KFe^{3+}{}_3(SO4)_2(OH)_6]$, leaving a minimum portion of Fe^{3+} in solution while simultaneously lowering the pH (Espana et al., 2005). At a pH value less than 2, the precipitate $Fe(OH)_3$ formed in the Equation (3) is not stable. Under such conditions Fe^{3+}ion remains in solution (Dold, 2010). However, the residual Fe^{3+} present in Equation (2) which does not take part in the formation of $Fe(OH)_3$ from solution as in Equation (3), can participate in the oxidation of additional pyrite which can be shown by the Equation (4) (Akcil and Koldas, 2006) .

$$\mathrm{FeS} \ + 14\mathrm{Fe} \quad + 8\mathrm{H} \ \mathrm{O} \rightarrow 15\mathrm{Fe} \quad + 2\mathrm{SO} \quad + 16\mathrm{H} \ \ldots\ldots\ldots\ldots\ldots\ldots \ (4)$$

When Fe^{2+} is formed (Equation 4) and adequate amount of dissolved oxygen is present then the Equation (2) and Equation (3) are continued (Younger et al., 2002). In absence of dissolved oxygen the Equation (4) will be continued to completion and the AMD water will indicate an elevated amount of Fe^{2+} ion (Younger, et al., 2002). The Equation (2) and Equation (4) are accelerated by low pH of the mine water and microbial activity of some acidophillic bacteria *Thiobacillus ferroxidans*, *Leptospirillum ferroxidans* etc. by producing large amount of SO_4^{2-} (Singer and Stumm, 1970; Jennings et al., 2008; Baruah and Khare, 2010). Another bacterium, *Ferroplasma acidarmanus*, has been identified recently which is responsible for the generation of acidity in mine waters (McGuire et al., 2001). These bacteria have the ability to increase Fe transformation rate 100 times more over an abiotic environment (Blowes et al., 2003).

The generalized overall equation for the whole process of acid mine drainage formation can be given as follows:

$$4\mathrm{FeS}\ (\mathrm{S}) + 15\mathrm{O}\ (\mathrm{g}) + 14\mathrm{H}\ \mathrm{O(l)} \rightarrow 4\mathrm{Fe(OH)}\ (\mathrm{S}) + 8\mathrm{H}\ \mathrm{SO}\ (\mathrm{aq})$$

This oxidation process of pyrite indicates two opposing significances, the first one is that high acidity of AMD increases the mobility and toxicity of different metals, and the second one is that the $Fe(OH)_3$ precipitate produces co-precipitation as well as the adsorption of metals in solution. (McGregor et al., 1998).

From literature (Caruccio and Geidal, 1985; Nordstrom, 1982), it was found that the pathways of bacterial pyrite oxidation are classified into two reactions: direct and indirect metabolic reactions. The direct metabolic reactions involve physical interaction between the bacteria and pyrite mineral particles, while in indirect metabolic reactions the physical interactions of both are not necessary. In this type of reactions, the acidophilic bacteria oxidize Fe^{2+} to $Fe^{3+}$by reproducing the Fe^{3+} required for the oxidation of pyrite (Singer and Stumm, 1970).

1.6 Environmental consequences of AMD

Acid mine drainage (AMD) is one of the most focusing consequences of coal mining in Northeastern region of India that affects surface and ground and surface water. AMD runs off active and abandoned coal mine sites into nearby water systems and causes extensive contamination that is spread downstream (Gray 1997). The oxidation of sulphide minerals (such as pyrite, pyrrhotite etc.) increases the formation of sulphuric acid which consequently promotes the release of a number of trace and heavy metals like Fe, Cu, As, Cd, Hg, Co, Ni, Zn, Pb, Se, Mn etc. (Baruah et al., 2003; Baruah et al., 2006; Baruah et al., 2016). Heavy metals are those which have atomic density higher than 6 g/cm3 (Gardeatorresdey et al., 2005; Gorhe,et al., 2006). In other words it can be defined as the elements having metallic properties with an atomic number more than 20 (Tangahu et al., 2011). This toxic/hazardous mixture flows towards streams and rivers as well as into the ground water producing an enormous amount of environmental problems. AMD imparts toxic effect to aquatic organisms by destroying the ecosystem and stains water in regions near the mine (Ruihua et al., 2011; Singh, 1987). AMD containing heavy metals has a number of serious health implications to human beings as well as to other animals due to their critical and long term toxicity (Ndlovu et al., 2013). The heavy metals are hazardous to human beings and animals due to their ability to persist in natural environment for a long period and also, due to their ability to accumulate in continual process of the biological chain causing severe and chronic health problems. High level of heavy metals in water or soil can enter into food chain easily and disrupt the metabolic functions by accumulating in vital organs and glands like brain, heart liver etc. (Singh et al., 2011).

High concentrations of heavy metals in plants over the desirable limit can disturb plant growth in different ways like plants face an oxidative pressure because of exposure to heavy metals like Cd, Hg, Zn, Pb, Cr etc. This causes their cellular damage with altering the plant physiology and morphology (Yadav, 2010). High acidity of AMD also affects the different aquatic life because the brown or yellow stain of iron hydroxide which is very fine in nature can deposit on the river, stream or drainbed by bolstering substrates (Fripp et al., 2000). Thus, small aquatic animals (benthic organisms) which live at the bottom of the rivers or streams cannot live for a long time which affect in the food chain of fish (Fripp et al., 2000). Also, the heavy precipitate of iron hydroxides coats the gills of the fish by hampering their respiratory system and diminishes the availability of clean gravels for their spawning (Dutta et al., 2017). Since, a proper amount of micro and macronutrients should be present in the soil for the proper growth of a plant. Thus, soil pH is an important determinant factor for the availability of plant nutrients (Halcomb and Fare, 2002). At low pH, the elements like N, P, K etc. which are present in soil can tie up in soil and they no longer available for plants. The essential plant nutrients like Ca, Mg etc. are absent or present in very low concentrations in soil at low pH. A lower pH value leads to decrease in activity of soil microorganisms which are very essential to break down the organic matters present in soil (Schrock et al., 2001). The decrease in pH of stream or river waters near coal mining area can decrease the sodium ions in blood of fishes which have an adverse effect on the function of gills (Evans, et al., 2005).Continuous exposure to high acidity is dangerous to plants and animals as it can stunt the plant's growth, decrease the reproduction rates and increase the deformation rates of animals (Vyawahre and Rai, 2016). Most of the Northeastern coalfields where Tertiary coals are found, discharge a large amount of acid mine water whereas the Gondwana coalfields are almost free from AMD (Tiwary and Dhar, 1994; Tiwary, 2001). From literature (Chabukdhara and Singh, 2016), it was found that the metals such as Fe, Mn, Cu, Ni and Pb in mine water of Northeastern coalfield are present in higher concentrations than the other coal mining sites of India. A comparative study by Tapadar and Jha (2015) showed that the quality of mine affected soil deviated from undisturbed forest soil by means of pH, heavy metal concentrations etc. The coal mine soil dumps has high acidity which was also supported by the reports of Bisws et al. (2013). According to Johnson and Hallberg (2005) the low pH (high acidity) of coal mine soil is due to AMD generated during and post mining processes. The other Indian coalmines that affected by serious acid mine drainage problems are Churcha, West Chirimiri, Rkhikol and Ambora (WCL), Gorbi (NCL) (Rawat and Singh, 1982; Sangita et al.,2010). From the elemental analysis of sediment samples from the Simsang river, Garo hills, Meghalaya it was found that the levels of Fe, Zn, Ni and Pb were significantly the permissible limit especially in the monsoon than the non-monsoon season in the order of Fe > Ni > Pb > Zn. The high concentrations of these metals in the Simsang river sediments were due to inflow of untreated AMD water nearby natural sources. Also, from the investigation by Talukdar et al., (2016), it was revealed that the degradation of soil quality was continuous as a result of this some agricultural areas around the Simsang river became unfertile. Since last few decades, AMD from coal mining activities poses severe threats to the fish and other aquatic lives of the river. Some researchers (Li et al.,2018) reported that Zhijin coal-mining district, located in Midwestern Guizhou Province, China has been significantly exploited for several years. The discharge of AMD from coal mining in this area has created a severe problem to the local water environment, which greatly affected the normal use by the local people. From the study of Gaolan River in China, it was found that the availability of macroinvertebrates in the AMD affected area is very low compared to unaffected area. Also, the population of scrapers were found to be low at AMD impacted sites due to their low tolerance capacity for contamination (Barbour et al., 1996) and due to the scarcity of benthic algae which is an important food reservoir for scrapers in the AMD effected water (Niyogi et al. 2002; Jia et al. 2009). In Appalachian region, the AMD generated from coal mining produces severe problems including destruction and reduction of productivity of aquatic life, reduction of water based recreation, high acidity, hardness, iron and manganese in municipal water supplies manifestation, increased corrosion of metal structures on highway and navigation facilities etc. Reports from The Appalachian Regional Commission study, it was found that more than 10,000 miles of streams have been affected by AMD (Hill and Bates, 1979). The United State Bureau of Mines reported that the abandoned coal mines and the associated piles of coal mine waste severely affect more 12,000 m of river and streams along with more than 180,000 acres of lakes and water reservoirs in the U.S. (Kleinmann, 1989). Acid drainage from underground, open cast coal mines and from coal wastes is the most chronic pollution problem in Appalachian Coal Region. In the year 1995, about 3902 km of streams in Pennsylvania could not meet EPA guideline in case of stream water quality standard because of loss of water quality through acid mine drainage due to long term mineral extraction (PADEP,1996). Also, 19,308km of stream water of United States was reported as degraded by acid mine drainage in 1970 (Warner, 1971). Thus, from our literature study, AMD is responsible for economic losses and making imbalances in water, soils and biota. Accordingly, a proper assessment of environmental effects of coal mining and the remediation process is very important for the socio-economic growth of the region and the restoration of healthy environment for the rural inhabitants. On support of this demand, the present research work is carried out with the following objectives.

1.7 Objectives of the study

The main research objectives of the study are:

1. Chemical characterization of coal, mine rejects, overburden, water, and soil samples from Ledo colliery of Northeastern coalfield, Margherita by using X-ray diffraction (XRD), Fourier transform infrared (FTIR), inductively coupled plasma optical emission spectrometry (ICP-OES), atomic absorption spectroscopy (AAS), ion chromatography (IC), Raman, scanning electron microscopy-energy dispersive spectroscopy (SEMEDS), Mossbouer, high resolution transmission electron microscopy (HRTEM) etc.
2. To study the seasonal change (monsoon and non-monsoon seasons) of physico-chemical parameters as well as elemental composition in soil/ water system in and around Ledo colliery and remediation measure of AMD.
3. To study the association and modes of occurrences of PHEs present in coals, mine rejects and overburden and their transformation and mobilization in different systems.
4. To study the physico-chemical and elemental characterization of aqueous leaching of coal and mine overburden from the Ledo colliery of Northeastern coalfields, Margherita.
5. Environmental assessment and nano-mineralogical characterization of coal, overburden and sediment from Ledo colliery of Northeastern coalfields, Margherita.

Deterioration of environmental conditions is the major contributory factor to poor health and quality of life that hinder sustainable development. It is expected that the findings from this thesis work will provide a base on overall environmental impact from the open cast coal mining activities around Ledo colliery in NER. It will provide the extent of environmental degradation by the coal mining activities around the colliery and will help to understand the probable management measures to combat such problem.

1.8 Arrangement of chapters

The important findings of this thesis work are summarized in different chapters are summarized below:

Both the open cast and underground coal mining activities in Northeastern coalfield of Assam have been carried out from a long time ago. However, in Ledo colliery, recently open cast mining process has been adopted which produces a major threat to the environment of the surrounding area near Margherita town. The validation of this study is to generalize the problem faced from open cast coal mining in an international and national ground.

Chapter I contains a general description of the work along with the literature survey. It describes the major forms/types of coal found in Margherita, Assam, with the information about different coal resources and distribution of coal in Northeastern region India. The present scenario of coal utilization is also discussed in this chapter. The geological settings of Ledo open cast mining area are described. The acid mine drainage (AMD) is the major burning problem created by coal mining processes all over India. The biochemical process of AMD generation from different sources including fresh mining, abandoned mining, overburden (OB) dump, coal storage etc. and its characteristics are summarized in the chapter. The environment concerning problems arises from AMD in coal mine area and its impact on life and plant through water and soil systems are also discussed in this chapter. The comparative literature study of environmental effect of AMD in Indian coal mine with other coal mines is also included.

Chapter II includes the details of study area and different analytical characterization techniques used in the entire study. It includes all the procedures adopted in selecting the sample sites, standard methods used during sample collection, preservation of sample for analysis etc. The methods and details of equipments used in characterization of the samples are also explained in this chapter. The procedures and instruments used for chemical analyses like sulphur analysis, CHN analysis, proximate analysis, ion-chromatographic analysis etc. of coal and OB/mine rejects are discussed in this part of the work. Other advanced analytical techniques like atomic absorption spectrometry (AAS) and inductively coupled plasma-optical emission spectrometry (ICP-OES), Fourier transform infrared (FTIR), X-ray diffraction (XRD), Mossbauer spectroscopy, and Raman spectroscopy along with the sample preparation are included in this chapter. This chapter also gives the methods used in x-ray diffraction (XRD), Raman, field emission- scanning electron microscopy (FE-SEM), and high resolution-transmission electron microscopy (HR-TEM) analytical techniques.

Chapter III describes the variation of physico-chemical properties of coal, OB, soil and mine water samples with the seasonal change. The results are reported on two specific seasons namely monsoon and non-monsoon periods in mining and nearby areas of Ledo colliery, Margherita. The physico-chemical parameters of different mine water samples are discussed. The results derived from the different physico-chemical study including pH, total dissolved solid (TDS), electrical conductivity (EC), inductively coupled plasma-optical emission spectrometry (ICP-OES), ion-chromatography (IC) and sulphur analysis are presented in this chapter. It revealed that there is a prominent change in the parameters like pH, TDS, EC, heavy metals including PHEs like Pb, Cr, Cu, Al, Fe, Co, Ni, Mn, and Zn and metal ions like Na^+, Mg^{2+}, K^+, NH_4^+ ions in monsoon season over the non-monsoon season. The results of this study will be helpful to interpret the water quality in mine source and nearby water resources in monsoon and non-monsoon seasons. Moreover, this study helps in indicating the higher contaminant level of hazardous metals in water system in monsoon season than in non-monsoon season which may pose a threat to survive the flora and fauna.

Chapter IV relates to the study of the aqueous leaching of coal and OB from Ledo coal mine and the characterization of the leachates in terms of their physico-chemical parameters. The physico-chemical parameters including pH, EC and TDS were determined for the aqueous leachates obtained at different time periods and temperatures. The acid mine water (AMD) generated in Ledo coal mine contain hazardous elements in different level of concentrations. The Northeastern coalfield produces considerable amounts of AMD. The AMD generation and metal leaching processes from mine over burden (OB) dumped near the mining area occur in a parallel way. In order to recognize the AMD potential, the leaching experiments of a few coal and OB samples from the Ledo colliery of the Northeastern coalfield, Mergherita (India) in aqueous medium at different time periods and at different temperatures(25□ C, 45□ C, 65□ C, and 90□ C) were investigated. It was revealed that the concentrations of trace and hazardous elements like Na, Mg, Fe, Al, Si, Hg, Pb, Cd, Cr, Mn, As, Se considerably change with leaching time and temperature. Alteration of the physico chemical structure of the coal and OB samples resulting from leaching effect was also studied by field emission- scanning electron microscopy-energy dispersive spectroscopic (FESEM/EDS) methods. The release of the potentially hazardous elements (PHEs) from the raw coal and OB during leaching time periods to the leachates was determined by ICP-OES and ion-chromatographic analyses. The major minerals associated with coal and OB are found to be mainly quartz (SiO_2), pyrite (FeS_2), hematite, marcasite and kaolinite. The overall study discussed in this chapter will be useful in relating the characteristics of aqueous leaching of coal and mine OB results from mining in the laboratory condition with the natural weathering condition at mine area.

Chapter V deals with the environmental assessment and nano-mineralogical characterization of coal, OB, and sediments of Ledo colliery has been carried out by using electron beam techniques like FE-SEM and HR-TEM analysis to evaluate the different nano-minerals/nanoparticles present in coal, OB samples etc. The AMD formation in open cast coal mining is significantly responsible for the deterioration of environmental conditions in and around the coal mining area which is the major contributory factor to poor health and quality of life that hinders sustainable development in the region. The chemical parameters of coal, overburden, soil and sediments along with the acid mine drainage (AMD) were investigated in order to understand the overall environmental impact from high sulphur coal mining at Northeastern coalfield. The high sulphur content in Ledo colliery is the determinant factor for generation of AMD in this area. The mine affected water samples have comparatively high electrical conductivity (EC) and high total dissolve solid (TDS) than unaffected water. Lower values of pH indicate the dissolution of minerals present in the coal as well as other minerals in the mine rejects/overburden. The observations obtained from other analysis high resolution-transmission electron microscopy (HR-TEM) with energy dispersive spectroscopy (EDS), selected area electron diffraction (SAED), field emission-scanning electron microscopy (FE-SEM)/EDS, X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), Raman and ion-chromatographic analysis, and Mössbauer spectroscopy are included in this chapter. From different geochemical analysis it has been found that the mine water sample from Ledo colliery has the lowest pH value of 3.30. This study was also extended to other nearby colliery (Tirap) to compare the effect by mining in both the collieries. The water samples have the lowest pH value of 3.6. Both Ledo and Tirap raw coals have total sulphur contents within the range 3-3.5 wt%. The XRD analysis revealed the presence of minerals including quartz and hematite in the coals from Ledo colliery. Mineral analysis of overburden (OB) reveals the presence of pyrite and marcasite which was also established by XRD and Mossbauer spectral analysis. The ^{57}Fe Mössbauer spectra of coal, overburden, and soil/sediment samples were measured in order to obtain information about the iron containing minerals present in them. Pyrite was found to be the only Fe-mineral present in coal, indicated by the 100% relative abundance. Although other Fe-minerals were found to be present the overburden, soil, andsediment of Ledo colliery like illite $(K,H_2O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2.(H_2O)]$, jarosite and marcasite, but the jarosite and iron sulphate are the oxidation products of pyrite. The high relative abundance of pyrite in the coal samples indicates the higher possibility of AMD formation. Both FTIR and ion-chromatographic analyse showed the presence of high concentrations of SO_4^{2-}. The nano-mineralogical analysis of coal and overburden revealed the presence of Si minerals like kaolinite $[Al_2Si_2O_5(OH)_4]$ with other minerals including sulphate minerals such as barite $[BaSO_4]$, jarosite $[KFe_3(SO_4)_2(OH)_6)]$, pickeringite $[MgAl_2(SO_4)_4.22(H_2O)]$; sulphide mineral galena$[PbS]$, pyrite $[FeS_2]$; oxide mineral hematite $[Fe_2O_3]$, and organic matters which are combined with pyrite. These minerals were also reported to be common in Brazilian and Chinese coals. HR-TEM analysis of the coal and mine overburden reveals the presence of nano-minerals containing potentially hazardous elements. Galena containing high concentrations of Pb, kaolinite having high concentrations of Al and Si can cause health implications. The presence of nano-minerals like nano-hematite in coal samples was revealed by HR-TEM technique. These nano-hematites with high surface area and reactivity are very environmentally sensitive. The nano-toxicity indicate that particle size and the different properties linked to the particle size, have an effect on both the bioavailability and the subsequent level of toxicity to organisms including human being. The presented data of the minerals and nano-particles present reflects their ability to control the mobility of potentially hazardous elements, suggesting conceivable use in environmental management technology as well as restoration of the delicate Indian coal mine areas.

Chapter VI describes about the major problems created by acid mine drainage water to the environment of nearby areas and the remediation study to minimize the effect of AMD. AMD generated problem is the well-known problem in all coal producing countries. Its ability to dissolve large amount of trace and heavy elements including Fe, As, Pb, Hg, Cd, Cr, Co, Ni, Mn, Zn, and Cu due to its high acidity is the major cause for its environmental concern. Due to the long lasting effect of AMD, it is necessary to protect and recover the environment in the coal mining areas to ensure that the environment be protected from a spreading pollution problem. Although, different countries take different remediation processes, but sometimes these are often not sufficient for controlling the degradation of the environmental quality in these mine sites. In our study, a new concept for a lab-scale nano- remediation process, by using nano- limestone treatment for AMD. Coal leachates have been used which have potential field applications.

Chapter VII describes the summary and recommendation of the present work. This chapter brings out an overall summary from **chapter I** to **chapter VII**. The significant results found in all chapters are included in this chapter from which the environmental implications from coal mining activities can be highlighted. All the results found in this study give a better picture of environmental condition of Ledo open cast coal mining area of Northeastern coalfield. The major findings of the thesis work and the recommendations will be helpful for local habitant to know about the environment related problems created by Ledo open cast mining activities in various fields like economic, social as well as their health effects. Thus proper precautions should be taken to overcome these problems.

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