



MODE OF OCCURRENCE OF MERCURY IN LIGNITE DEPOSITS OF SINDH, PAKISTAN

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ABSTRACT

Pakistan has huge deposits of 185 billion tons of lignite coal deposits. The major deposits are found in lower Indus Basin, southern Sindh. The coal basins extend westward from Thar coal field, near Islamkot through Badin to Lakhra-Sonda and Meting-Jhimpir-Thatta area. There are 16 coal samples from southern coalfields of Pakistan have been analyzed for the presence of mercury. The results show that the mercury in Thar coal samples varies from 0.38 to 0.56 ppm, with an arithmetic mean of 0.45 ppm. In Lakhra coals, the concentration varies from 1.12 to 1.94 ppm with an arithmetic mean 1.34 ppm. Whereas, in Meting-Jhimpir coal samples, it varies from 1.748 to 1.750 ppm with arithmetic mean 1.75 ppm. The current results indicate that the mercury in Thar coal has originated as silicate bound, in Lakhra coals as sulfide bound, and in Meting-Jhimpir coal, as carbonate bound. Calculated enrichment/depletion factor shows that the mercury in Thar and Lakhra coal samples is significantly enriched, whereas in Meting-Jhimpir coal it is very highly enriched. The comparison of present data with world average values shows that the average mercury contents in coal samples of Thar coalfield relative to Lakhra and Meting-Jhimpir coalfield are very near to the world average and is of silicate bound nature. Therefore, it will pose no environmental threat if proposed coal-fired power station is installed, to overcome the energy crises of Pakistan.

Keywords: Lower Indus Basin, lignite coal, silicate bound mercury, carbonate bound mercury.

INTRODUCTION

Mercury (Hg) has drawn the global attention and concern as hazardous pollutant, due to its toxicity. In nature mercury occurs in three primary forms; elemental, divalent and organic. Elemental mercury stays in atmosphere from six month to two years due to its stability and has a potential to distribute globally (Watras *et al.*, 1994; Clarkson, 2002). In general, coal contains mercury in traces, amounting 0.01-0.5 ppm. Due to coal combustion, mercury spreads in atmosphere, when it is released as exhaust in the form of elementary mercury (Hg⁰) (Meij *et al.*, 2002; Niksa and Fujiwara, 2004., Lee *et al.*, 2006; Park *et al.*, 2008). The combustion of coal generates various trace elements like As, Cd, Pb, Se, and Hg and have raised concern about human health (Clarke and Sloss, 1992; Gibb *et al.*, 2000). In the result certain organizations such as European Pollutant Emission Register (EPER, 2000), have reported various hazardous Air Pollutants like, As, Cd, Cu, Cr, Hg, Ni, Pb and Zn. The USA Clean Air Act Amendments Bill (1990) has also declared Be, Cl, Sc, Cr, Mn, Fe, Co, Ni, As, Cd, Sb, Pb, Th, U, and Hg as hazardous air pollutants, which are produced from the combustion of coal (Gibb *et al.*, 2000, Huggins, 2002).

Booth *et al.* (1999) included mercury in the list of elements of greatest environmental concern. The US national committee for geochemistry (1980) also includes mercury in the list of trace elements (e.g. B, As, Se, Mo, Cd, Hg and Pb) as elements of major environmental concern. Goodarzi (2002) also includes mercury among the elements such as Se, V, Cr, Cu, Zn, As, Mo, Cd, and Pb as elements of prime international concern.

Pakistan has total lignite coal deposits of 185 billion tones in lower Indus Basin, southern Sindh. These coal deposits are found in (1) Sonda, (2) Meting-Jhimpir (in Thatta district) (3) Lakhra (Jamshoro district) and (4) Thar coalfield in Tharparkar. Sixteen coal samples were collected from coal mines of Lakhra and Meting-Jhimpir coalfields, whereas the drill core samples from Thar coalfield, were obtained from Sindh coal authority's field core library at Islamkot.

The Thar coalfield is one of the largest coalfield in Pakistan and is spread over 9100 km². It is situated at the distance of 410 km from the east of Karachi, Sindh. The Lakhra coalfield covering an area of 68 km² is located 225 km North East of Karachi in Jamshoro, district. The Meting-Jhimpir coalfield occupies an area of about 422

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km², and is situated approximately 150 km to the east of Karachi in Thatta district.

It is expected that in the future coal will emerge as the dominant source of energy in Pakistan, as at present country is facing extreme energy crisis, but the coal combustion may result the release of volatile trace elements like mercury along with other pollutants into the atmosphere. It is therefore, important to study the potential impact of mercury on the environment. Considering the environmental concerns about the possible distribution of mercury after its combustion, the lignite coals of Sindh, southern Pakistan have been studied to assess the concentration of mercury in these coal deposits and to evaluate its impact, if any.

Geological Setting of Coalfields of Southern Pakistan

The province of Sindh, is situated in the Lower Indus Basin, where a number of coal field are reported. Geologically, two stratigraphic horizons of coal deposits had developed. One is in the lower part, where it is associated with Bara Formation of Daninian age while the second was deposited in upper part where it occurs within the Sonhari Member of Laki Formation of Ypresian age (Kazmi and Abbasi, 2008).

The Thar coalfield area is mainly covered by sand dunes. Subsurface geology reveals that the Pre-Cambrian basement rocks of igneous origin are uncomfortably overlain by coal bearing strata of Palaeocene-Eocene sediments. The basement complex at Thar coalfield is lying between 110 to 277 metres depth. Generally, granitic and granodioritic rocks constitute the basement, which is highly altered into kaolinite. The Paleocene-Eocene coal bearing strata are generally composed of claystone, carbonaceous clay, sandstone and siltstone with inter-laminated coal beds. The percentage of sand increases below the last coal bed. Claystone is the dominant lithological unit of the formation. It is silty at places and also contains pockets of very fine sand and scattered coal fragments (SanFilipo, 2000). The claystone is generally present as the floor and the roof rock. Pyrite is present in the form of patches upto 2 mm size. Claystone also contains 3 to 30 cm thick sideretic bands and nodules (SanFilipo *et al.*, 1992; Jaleel *et al.*, 2002; Siddiqui *et al.*, 2007).

The Lakhra coal field area is underlain by sedimentary rocks. The rocks exposed in the area belong to Ranikot group (Paleocene), Ypresian (Sonhrari member-Early Eocene), Ypresia (Laki limestone-Early Eocene) and Calabrian (Manchar Formation). These units are composed of shallow marine and fluvial sediments. The oldest exposed rocks belong to the Daninian (Bara Formation) of Early Paleocene age. It is conformably underlain by Thanetian (Lakhra Formation) of Middle to Late Paleocene age and unconformably overlain by the

Ypresian/Laki Formation of Early Eocene age. The unconformity between Lakhra and Laki formations is marked by a 38 to 63 cm thick laterite bed. Rocks of Middle Eocene to Miocene (intervening the Laki limestone and the Manchar Formation) are absent in the area, hence the Calabrian (Manchar Formation of Pliocene age directly overlies Ypresian/Laki Formation. A very thin cover of alluvium rests over the Manchar Formation. The minable coal seam's thickness in the area varies between 1.5 to 3.35 meters (Khan *et al.*, 1988).

According to Outerbridge *et al.* (1991) the deposition of coal in the Meting-Jhimpir area took place on the erosional surface of the Paleocene rocks. It is grouped in the lower part of the Laki Formation of Ypresian age. In Meting-Jhimpir area, coal is associated with lateritic clay and shale with beds of arenaceous sandstone of Ypresian (Sonhari Member of Early Eocene age). The Sonhari coal is of poor quality lignite with high Sulfur (Siddiqui and Shah, 2007). Only one workable coal seam ranging in thickness from 0.3 metre to 1.0 metre with an average thickness of about 0.5 metre is present which is generally thin and lenticular. The coal is of lignite-A to sub-bituminous-B in rank. It is soft, friable and exhibits spontaneous combustion on exposure. It is estimated that 25 million tonnes of coal deposits are found in Meting-Jhimpir area.

MATERIALS AND METHODS

In this study 16 coal samples were collected from the coal mines of Lakhra and Meting-Jhimpir coalfield, while fresh drill core samples were collected from drilling site of Thar coalfield, and grounded to 75 μ (200 mesh size). The samples were air dried as per ASTM D-2013 method (ASTM, 2011). The surface moisture was removed through oven by setting temperature between 10-15°C with maximum temperature upto 40°C. For removing the inherent moisture of the coal the oven was set at 107-110°C temperature for approximately one hour (ASTM, 1985).

ASTM D 6414-99 (ASTM, 2014) method was used for the determination of mercury through acid digestion method described by Jeffery and Hutchinson (1981) was employed. In this method 0.1 g of coal samples were digested with 2ml of HNO₃ and 6ml of Cl in Teflon beakers and then heated at 80°C for one hour. After that 36.5 ml of deionized water was added, followed by 5 ml of 05% KMnO₄. After ten minutes, finally 0.5 ml of Hydroxylamine mixture (NH₂OH) was added for the complete digestion. Stannous chloride was used as reducing agent for the generation of free mercury vapor. Finally digested samples were run on Atomic Absorption Spectrophotometer as per ASTM D3684 method (ASTM, 2006). The samples were run on 3300 Perkin Elmer-700

graphite furnace Atomic Absorption Spectrophotometer equipped with mercury hydride system (MHS).

RESULTS AND DISCUSSION

In Table 1 summarizes the results of mercury analysis in Thar, Lakhra and Meting-Jhimpir coals. The table shows the minimum and maximum concentration of mercury in studied coal. The range, arithmetic and geometric means of mercury for Thar, Lakhra and Meting-Jhimpir coalfields are listed in Tables 2 and 3.

The arithmetic mean of mercury in Thar coalfield has the lowest concentration (AM 0.45ppm). The coal samples from Lakhra coalfield have relatively higher concentration (AM 1.34ppm), whereas the concentration of mercury in Meting-Jhimpir coalfield has even the higher concentration (AM 1.75ppm). The geometric mean is used for the estimation of probable concentration. As per calculation, the geometric mean of mercury content in Thar coal samples is 0.45ppm, in Lakhra coalfield is 1.34ppm and in Meting-Jhimpir coals is 1.75ppm.

Table 1. Analytical data of mercury in studied coal (values in ppm).

	Thar coal	Lakhra	Meting-Jhimpir
	0.450	1.336	1.749
	0.480	1.250	1.748
	0.560	1.115	1.749
	0.450	1.128	1.748
	0.380	1.133	1.748
	0.450	1.630	1.749
	0.420	1.234	1.748
	0.450	1.935	1.749
	0.430	1.255	1.748
	0.450	1.412	1.749
	0.420	1.542	1.748
	0.400	1.320	1.749
	0.450	1.300	1.750
	0.480	1.325	1.748
	0.550	1.121	1.748
	0.450	1.333	1.748
Min	0.380 (0.38)	1.115 (1.12)	1.748 (1.75)
Max	0.560 (0.56)	1.935 (1.94)	1.750 (1.75)
Average	0.45	1.34	1.75

Table 2. Minimum, arithmetic mean, geometric mean, standard deviation of mercury in coalfields of southern Pakistan (values in ppm).

	Thar (N=16)	Lakhra (N=16)	Meting-Jhimpir (N=16)	World	World Av.
Min	0.38	1.12	1.748	0.01	0.01
Max.	0.56	1.94	1.75	0.02	
AM	0.45	1.34	1.75		
GM	0.45	1.34	1.75		
Std Dev	0.05	0.02	0.001		

(N=Number of samples)

Table 3. Range, mean, geometric mean, Clarke value and enrichment factor of mercury in coalfields of southern Pakistan (values in ppm).

	Thar	Lakhra	Meting-Jhimpir
Range	0.380- 0.560	1.115- 1.935	1.748- 1.750
Mean/Average	0.45	1.34	1.75
Geometric Mean	0.45	1.32	1.75
Clarke Value	0.08	0.08	0.08
Enrichment Factor	5.625 or 6	16.75 or 17	21.875 or 22

The geometric mean of mercury in Thar coal samples is compared with rest of analyzed coals and it is found that in Lakhra coals it is three times higher than the Thar coal, whereas the mercury in Meting-Jhimpir coal is four times higher than the Thar coal samples.

Table 4. Average concentration of Hg in studied coal and Hg in lignite of the world.

Country	Hg in coal
Pakistan	
Thar	0.45
Lakhra	1.32
Meting-Jhimpir	1.75
India ^a	2.31
Indonesia ^b	0.03
Germany ^c	0.11
Thailand ^d	0.21
Romania ^e	0.12
Turkey ^f	0.11
UK ^g	0.15
Kosovo ^h	0.11
China ⁱ	0.10
World ^j	0.01

^a. Bhangare et al., 2011 ^f. Finkelman, 2004

^b. US EPA, 2002 ^g. US EPA, 1997

^c. Finkelman, 2003 ^h. Finkelman, 2004

^d. Finkelman, 2003 ⁱ. Haung et al., 2005

^e. Finkelman, 2004 ^j. Ketris and Yudovich (2009)

In addition, the geometric mean values of mercury in studied coal are compared with world average mercury concentration in coal i.e. 0.01ppm. The comparison shows that mercury in Thar coal is nearly equal to the world average. The mercury in Lakhra coals is three times higher than the world averages and the average mercury content in Meting-Jhimpir is four times higher than the world average (Table 4).

Enrichment factor can also be used to estimate depletion and enrichment of the element in coal. The enrichment or depletion factor of the elements is calculated by computing arithmetic mean values (AM) and dividing the resultant means by their Clarke value. Elements are considered highly enriched when the Clarke value is more than 10, while those elements with clark value less than 0.1 are known as depleted (Clarke and Washington, 1924). The comparison of studied coal with Clarke value i.e. 0.08, shows that coals from Thar coalfield are slightly

enriched, whereas coals from Lakhra coalfield and Meting-Jhimpir coalfield are highly enriched. Generally the enrichment factor is used to assess pollution in environment. Sutherland (2000) interpreted the enrichment factor as follows:

Enrichment Factor = < 2 depletion to minimum enrichment

Enrichment Factor = 2 - <5 moderate enrichment

Enrichment Factor = 5<20 considerable or significant enrichment

Enrichment Factor = 20-<40 very highly enriched

Enrichment Factor = >40 extremely highly enriched.

On the basis of Sutherland (2000) assessment, the investigated lignite coal samples reveal that mercury in Thar and Lakhra coal samples is significantly enriched, whereas in Meting-Jhimpir coal is very highly enriched (Table 2). United Nations Environment Program - UNEP (2002) in a study to reduce mercury emissions from coal combustion; published a data regarding mercury content in raw lignite coal from various countries of the world (Table 4). This table shows that mercury content in studied coal ranges from 0.45 (in Thar coal) to 1.75 ppm (in Meting-Jhimpir coal). The coal from India (Phalki, Naugpur coalfield) has the highest concentration of mercury i.e. 2.31ppm (Bhangare *et al.*, 2011). Mercury concentration in German lignite coal is 0.11ppm, the lignite coal of Thailand has the mercury content of 0.21ppm (Finkelman, 2003), and the lignite of Romania has the mercury content of 0.12 ppm. Whereas, the Turkish coal has mercury content of 0.11ppm (Finkelman, 2004). The lignite coals from United Kingdom have the mercury content of 0.15ppm (US EPA, 1997). The lignite coal from Kosovo has the mercury content of 0.11ppm (Finkelman, 2004). The Chinese lignite coal has 0.10ppm mercury content (Haung *et al.*, 2005). According to Ketris and Yudovich (2009), the world average of mercury for lignite coal is 0.10ppm.

The lignite coals of Pakistan are also compared with the lignites of the world in terms of their mercury content (Table 3). The comparison of studied Thar, Lakhra and Meting-Jhimpir coal, with other countries of the world shows that the mean values of investigated coal samples have the lower content of mercury as compared to Indian lignite coal. The mercury content in all studied coalfields has the higher concentration as compared to other countries of the world, except India. Though among the all studied coal samples, Thar coal has relatively the lower mercury content i.e. 0.45ppm.

Origin and Occurrence of Mercury in Studied Coal

The mode of occurrence of mercury within the coals is of two types (Feng *et al.*, 1999; Finkelman; 2004; Yudovich and Ketris, 2005) pointed out that mercury in coal occurs dominantly as sulphide bound. The mercury in coal is also

found as silicate-bound as in Chinese coal (Zhang *et al.*, 2008), the mercury in Thar coals varies from 0.38 to 0.56 ppm, with an arithmetic mean of 0.45ppm. Figure 1 (i) shows the X-ray diffraction analysis investigation of coal samples from Thar coalfield, showing dominant silicate minerals i.e. 86.8% zircon-metamict Zr SiO₄, and 8.5% quartz SiO₂ minerals. It is therefore, observed that mercury in Thar coal is of silicate bound origin. The SEM micrograph shows that the Thar coal contains dominant silicate minerals (Fig. 2-a).

In Lakhra coal samples, the mercury concentration varies from 1.12 to 1.94 ppm, with an arithmetic mean 1.34 ppm. The XRD investigation Lakhra coal samples; identifies the dispersed micrometer-sized grains of pyrite (FeS₂) minerals i.e. 88.41% (Fig. 1(ii)). The scanning electron microscopy of samples also shows the sulphide minerals i.e. 1.48% Fe and 4.52% Sulphur (Siddiqui and Shah, 2007) indicating that, the coal has sulphide bound mercury (Fig. 2-b).

The mercury in Meting-Jhimpir coal samples varies from 1.75 (1.748) to 1.75ppm (1.750) ppm with an arithmetic mean 1.749 ppm (175) (Table 2). The X-ray diffraction analysis of the investigated samples from Meting-Jhimpir coalfield has shown abundant clay minerals (i.e. 58.1% kaolinite-montmorillonite) that may have originated as detrital mineral in coal. The SEM micrographs in Figure 1 (c) also shows, the presence of carbonate minerals (i.e. 8.06% calcium, and 0.62 magnesium), in Meting-Jhimpir coal samples. Figure 2-c shows the SEM micrograph of coal of Meting-Jhimpir coals reveals that coal is of carbonate bound mercury (Siddiqui, 2012).

Potential Impact of Mercury

Mercury has the low boiling point i.e. 356.5°C, therefore higher amount of coal burned for electric generation, will cause emission of elemental mercury in vapor phase that may escapes with flue gases from stack of the power station (Menounou, 2003; Pacyna *et al.*, 2006).

As per US EPA (2002) and US Geological Survey (1970) studies, the lignite coal with low calorific values has high potential for mercury emission. The lignite coal based Lakhra Fluidized-bed-combustion (FBC) power station at Khanot, Sindh-Pakistan is generating 150 MW of electricity from lignite coal at the rate of 52 tons per hour. The Lakhra coal used for combustion has also low calorific value i.e. 3057-5088k cal/kg (5503-9158 Btu/lb) (Siddiqui and Shah, 2007). As this coal is being used in the 150 MW FBC Lakhra power station. Therefore, it is assumed that the coal used here may cause higher amount of mercury emission from the power station.

Another study, Xu *et al.* (2003) suggested that burning of mercury is linked with sulfide minerals, which vaporizes during combustion. As a result, the smallest sized particles of fly ash will cause the volatilization of elements that have tendency to concentrate on these particles when release in the gaseous phase. Moreover, the present pollution control devices are also not capable to detect these pollutants (typically PM₁₀ and finer) and so far, may escape up into the atmosphere (Buhre *et al.*, 2006).

The present study shows that coal used in the power station at Khanot has sulphide bound mercury (i.e. having sulphide minerals), so during combustion, the power station may release finer PM₁₀ airborne particles that may cause mercury related diseases in the surrounding population. It is also established fact that during coal combustion mercury distributes widely in all tissues and may cause mercury poisoning, neurological diseases, kidney related diseases and diarrhea to among the people living in the area (Siddiqui *et al.*, 2000, Finkelman *et al.*, 2003).

CONCLUSION

The mercury concentration of analyzed coal particularly from Lakhra and Meting-Jhimpir coalfield is high as compared to global averages. The comparison shows that mercury in Lakhra coal is nearly 3 (2.5) times higher in Meting-Jhimpir coal it is 3 times higher, while Thar coal is 0.09 times higher than world average. Further, the data also shows that, mercury in Thar coal is approximately equal to world average and slightly enriched as compared to Clarke's value. The Thar coalfield has low mercury concentration because of silicate bound nature. It is therefore, assumed that the proposed power station at Thar coalfield will have lesser impact on environment, and there will be no any serious impact on the health of local community. However, clean coal technology may be installed in the proposed power station to minimize any environmental impact. In the case of Lakhra power station, where sulphide bound mercury is being used, therefore, advance mercury capturing technology may be acquired and clean coal equipment may be installed to protect the local population from the possible threats of mercury pollution.

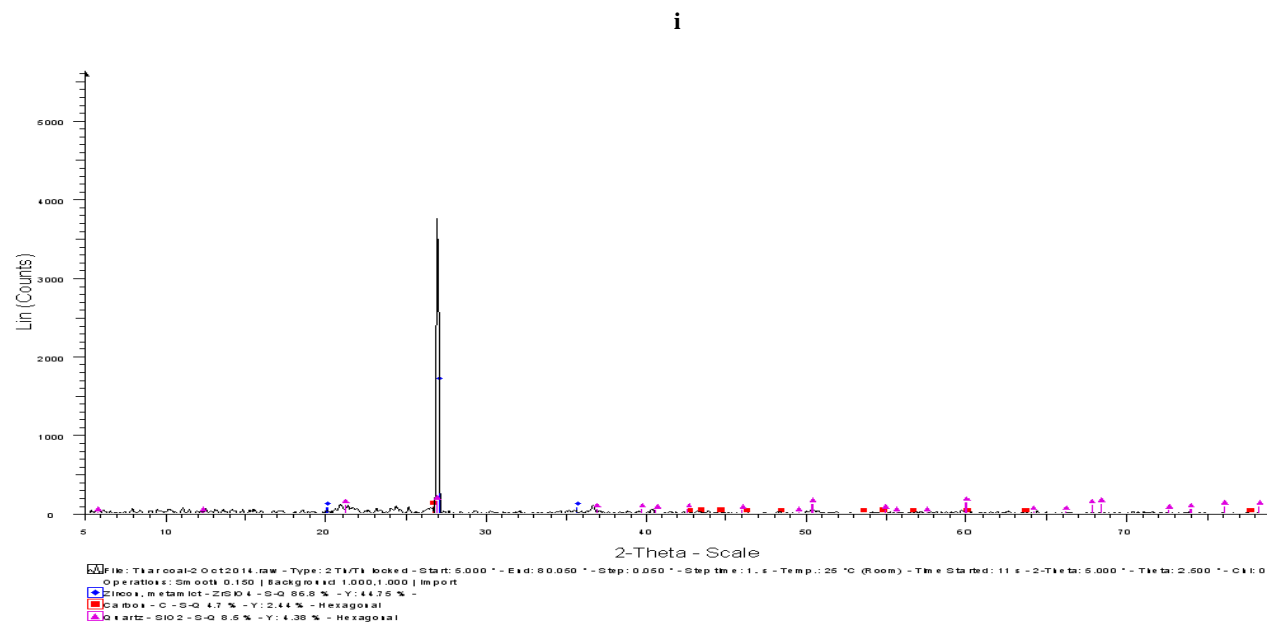


Fig. 1(i). XRD pattern of Thar, coals.

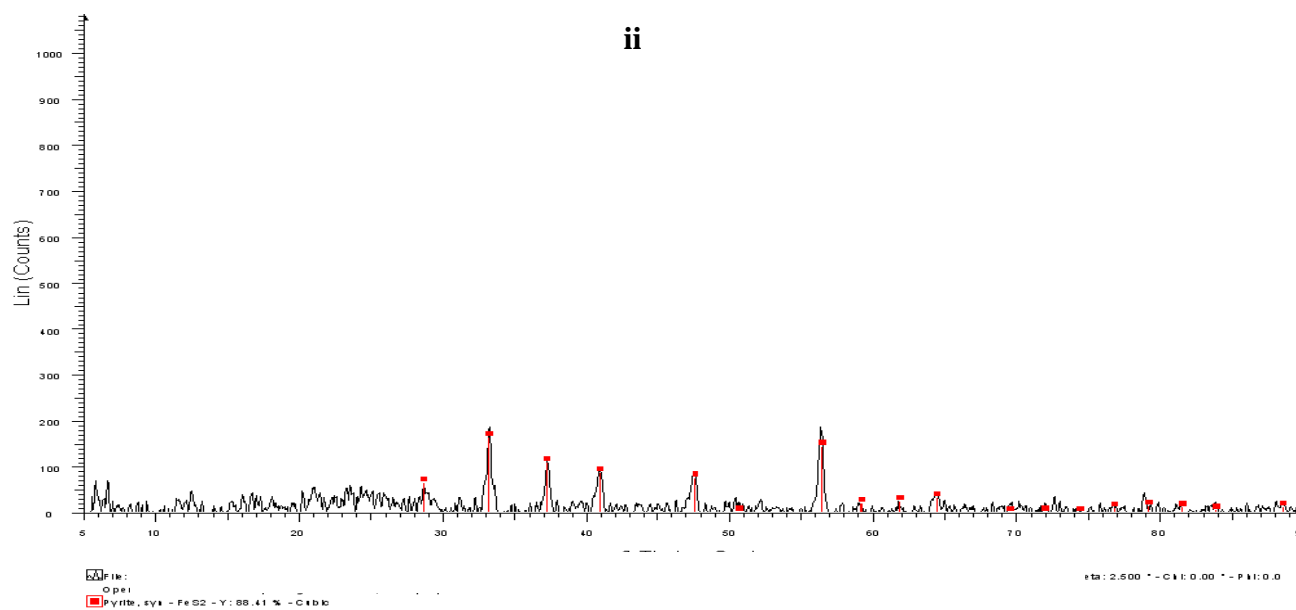


Fig. 1(ii). XRD pattern of Lakhra coal.

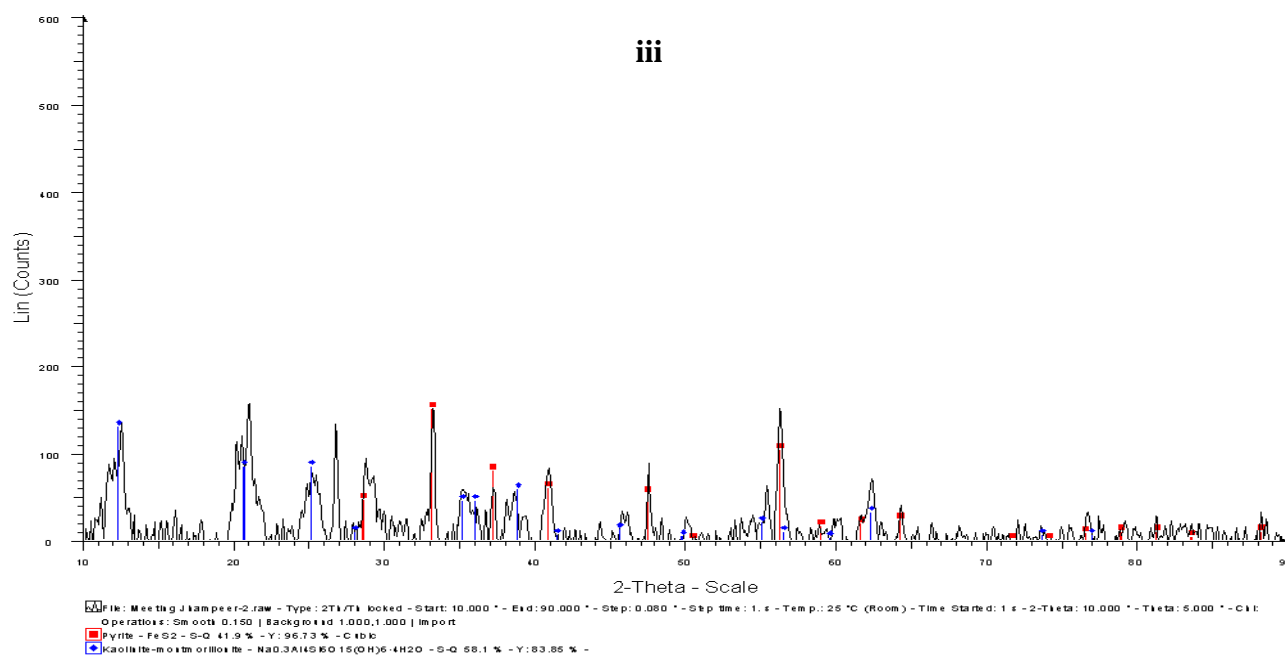


Fig. 1(ii). XRD pattern of Meting-Jhimpir coal.

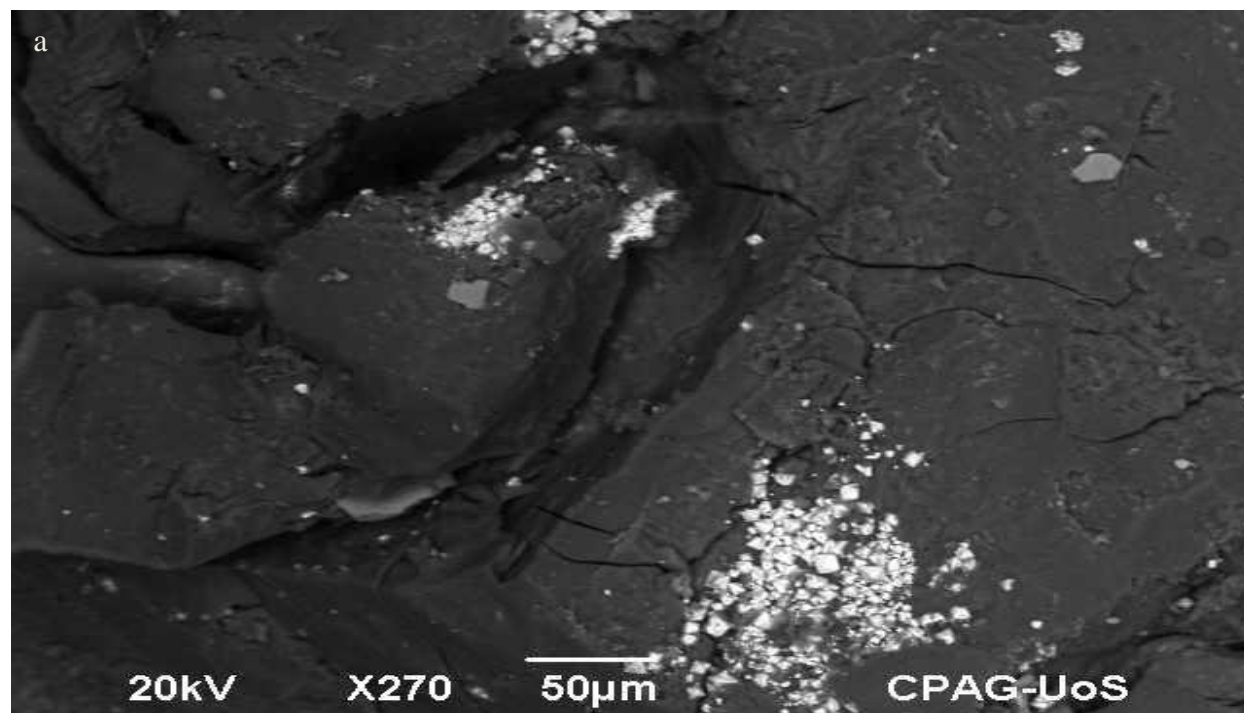


Fig. 2a. Microphotograph of coal sample of Thar coalfield, showing silicate minerals in coal.

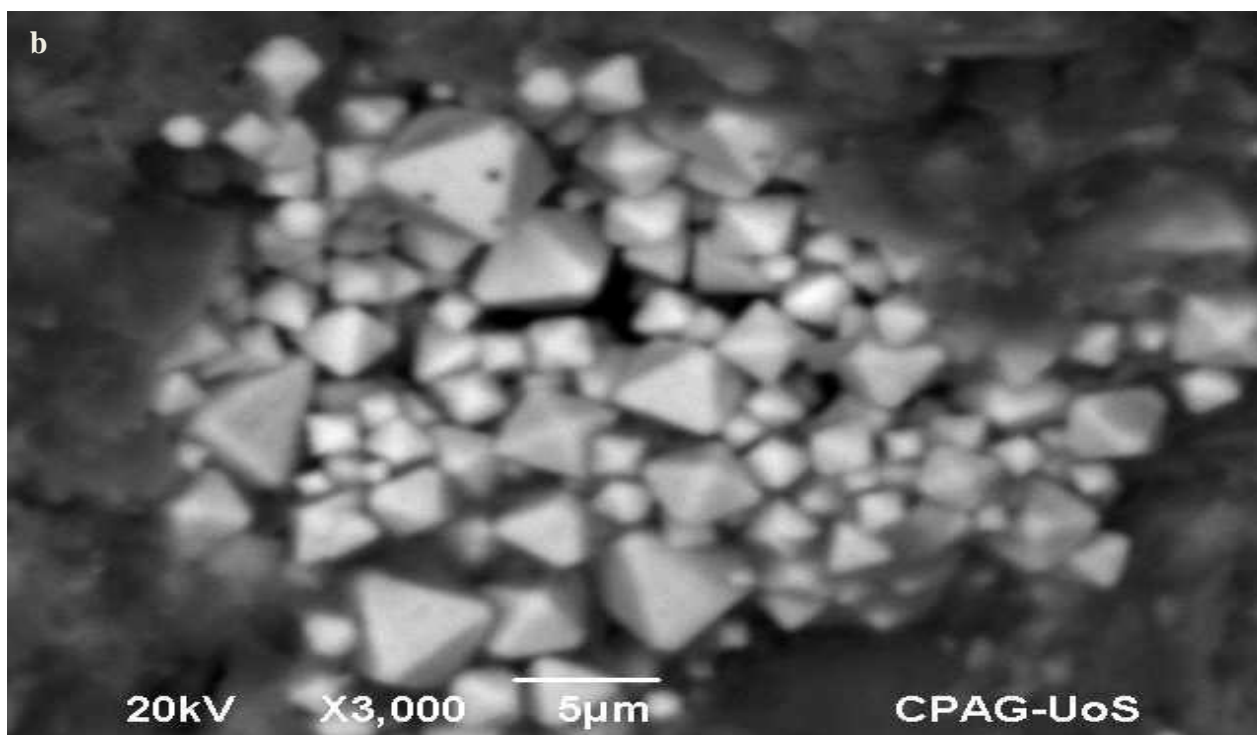


Fig. 2b. Microphotograph showing coal sample of Lahra coalfield, showing iron sulphide/pyrite (FeSO_2).

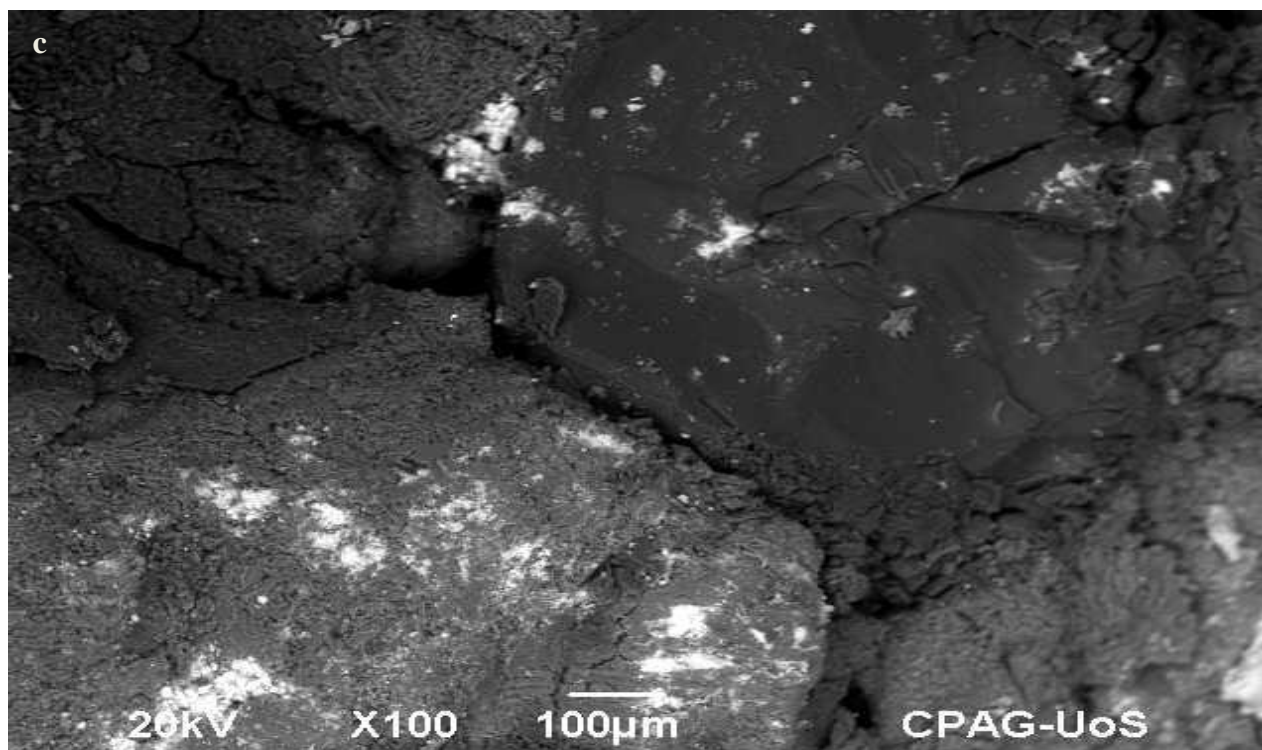


Fig. 2c. Microphotograph of coal samples of Meting-Jhimpir coal, showing carbonate minerals.

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