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THE MINERALOGY AND CHEMISTRY OF REFRACTORY CLAY DEPOSITS FROM LAKHRA AND JHAMPEER IN SIND, PAKISTAN

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Abstract. The well-known coal seams at Lakhra and Jhampeer occur in the Ranikot Formation and the Sonahri Beds. The refractory clays are found in the roof rocks and the seatearths associated with the coal seams. Twenty-six samples of roof rocks, fourteen from Lakhra and twelve from Jhampeer were obtained for mineralogical, chemical and thermal investigation. The refractory clays from both localities have dominant kaolinite with some quartz and anatase. Those from Lakhra also contain traces of illite. These contain higher clays amounts of aluminium and titanium and a lower alkali content than clays of similar origin from other regions, and can be used for making sanitary ware, buff tiles, engineering bricks and refractories.

Introduction

The refractory clay deposits are exposed in the Habib coal mines at Lakhra and the Daud coal mines at Jhampeer as roof rocks and seat earths of the working coals. The coal seams at Lakhra are a part of the Rahman Member of the Ranikot Formation (Palaeocene), while the seam at Jhampeer occurs in the Sonahri Beds (Eocene). Six samples were obtained from the roof rocks at the Habib coal mine No. 12, and eight samples were taken from the roof rocks at the Habib coal mine No. 2 as representative of the Lakhra refractory clays. The roof rock clays at Lakhra are about 8 ft thick. Five samples were obtained from roof rocks exposed at Daud coal pit No. 11 and seven were taken from an abandoned coal pit, near pit No. 11, as representative of the Jhampeer clays. The roof rock clays at Jhampeer are about 6 ft thick. It was not possible to acquire samples from the seatearths at Lakhra and Jhampeer due to lack of exposures.

Methods of Investigations

Mineralogical and chemical analyses of the samples were obtained using X-ray diffraction and X-ray fluorescence techniques. The X-ray diffraction studies were performed by making diffractograms of random powder samples and oriented slides were prepared from the less than 5 μ fraction. Chemical analyses were carried out on pellets of finely-powdered material after passing through a 200 mesh sieve. Further details of the instrumentation and methods of samples preparatin can be found in Brown [2] and Baqri([1]. Thermal investigations were carried out by using high temperature furnaces.

Results

X-ray Diffraction. The random powders of the Lakhra clays showed reflections due to kaolinite, quartz and anatase. The reflections of kaolinite in random powder diffractograms indicate its poorly crystalline nature [2]. Figure 1 shows a representative diffractogram of the random powder sample from a Lakhra clay. The oriented slides of less than 5 μ fraction of the Lakhra clays indicate that this fraction contains mainly kaolinite with traces of illite. Basal reflections of kaolinite were observed at 7.13Å and 3.57Å. Illite showed a weak reflection at 10Å. Figure 1 shows the nature of kaolinite and illite reflections in the oriented slides. The three traces are diffractograms of untreated (normal), glycolated and heated clays respectively. The kaolinite reflections were not affected by glycolation, but disappeared on heating at 550°C. The reflections of illite show no change on glycolation and heating at 550°C.

Random powder diffractograms from the Jhampeer clays showed reflections due to kaolinite, quartz and anatase. The kaolinite reflections indicate that this is the dominant mineral and is poorly crystalline. Figure 2 shows a representative diffractogram of a random powder sample of a Jhampeer clay. Orient-

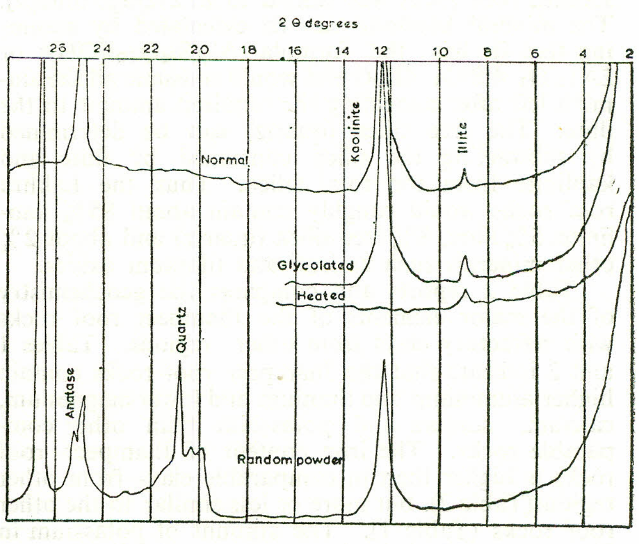

Fig. 1. Reflection of kaolinite and illite from a representative sample of Lakhra clays.

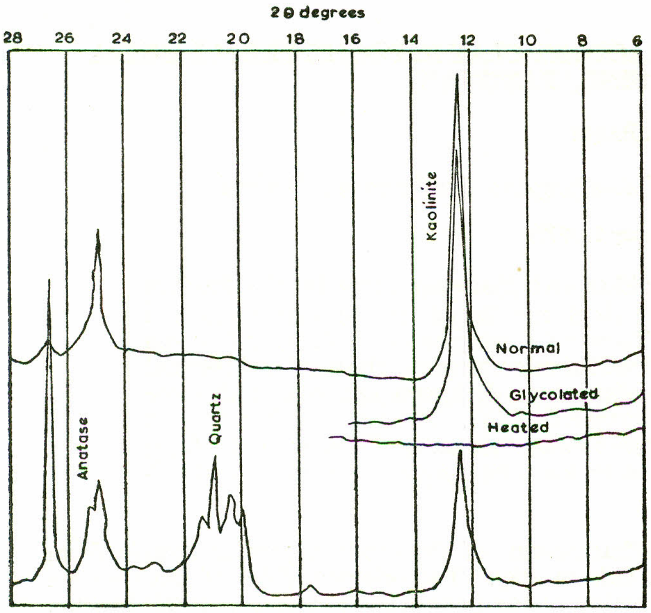

Fig. 2. Reflections of kaolinite from a representative sample of Jhampeer clays.

ed slides of the fine fraction from the Jhampeer clays exhibited erflections due to kaolinite only (Fig. 2). These reflections showed no change on glycolation, but disappeard on heating.

Chemical Analyses. Table 1 reports and compares the geochemistry of the major elements of Lakhra roof rocks with those of other regions. The lakhra roof rocks contain higher amounts of aluminium and titanium and lower iron, magnesium, sodium, calcium and potassium than some roof shales (Table 1) and seatearths (Table 2) of other regions. A mineral composition of these rocks can be calculated by assuming that all the potassium is present in illite. It was assumed that the illite compositions of the Lakhra roof rocks was similar to an average illite[9]. The mineral kaolinite can be calculated by assuming that it has the formula $Al_4(Si_4O_{10})(OH)_8$ or $2Al_2O_3.4SiO_2.4H_2O$ and would consume all the alumina left after allocating the required amount to the illite. The free silica (quartz) can be determined by subtracting the silica consumed by illite and kaolinite from the total silica. Thus the Lakhra roof rocks would roughly contain about 85% kaolinite, 5% illite, 8% free silica (quartz) and about 2% other minerals such as iron and titanium oxides.

Table 2 reports and compares the geochemistry of the major elements of the Jhampeer roof rocks with refractory clays from other regions. Tables 1 and 2 indicate that the Jhampeer roof rocks contain higher aluminium and titanium and lower magnesium, calcium, sodium and potassium than other comparable rocks. The iron content of Jhampeer roof rocks is higher than in comparable clays from other regions(Table 2), but more or less similar to the other roof rocks (Table 1). The amount of potassium in Jhampeer roof shales is very low and illite was not detected by X.R.D. Hence the quantity of kaolinite can be calculated by assuming that all the aluminium

TABLE 1. MEAN ANALYSES OF LAKHRA CLAYS COMPARED WITH THOSE OF OTHER REGIONS.

	1	2	3	4	5	6	7
SiO_2	50·00	50·70	59·66	61·06	54·65	62·20	56·45
TiO_2	3·53	3·54	0·91	0·93	0·80	0·96	1·01
Al_2O_3	35·19	35·36	19·68	22·26	19·00	21·49	25·17
Fe_2O_3	2·53	2·55	5·15	2·96	10·20	7·13	5·18
MgO	0·39	0·36	1·27	0·70	1·45	1·70	1·25
CaO	0·14	0·15	0·36	0·38	0·35	0·37	0·14
Na_2O	0·18	0·16	0·39	0·46	0·60	0·74	0·46
K_2O	0·40	0·41	2·57	2·96	2·97	3·46	3·99

1 Average of six samples from Habib coal mine No. 12 at Lakhra.
2 Average of eight samples from Habib coal mine No. 2 at Lakhra.
3 Average of 28 roof rock [8].
4 Average of three roof shale samples (Bc 7, 8, 9) from the Barsham cyclothem [7].
5 Average of 16 roof rocks associated with Better Bed coals in Yorkshire [10].
6 Average of 9 roof rocks from South Wales [4].
7 Average of 86 roof samples [1].

TABLE 2. MEAN ANALYSES OF JHAMPEER CLAYS COMPARED WITH THOSE OF OTHER REGIONS.

	1	2	3	4	5	6	7
SiO_2	42·66	42·10	57·96	58·5	54·1	61·99	60·95
TiO_2	4·28	4·26	1·19	1·26	1·19	1·06	1·21
Al_2O_3	34·92	34·87	26·93	24·6	24·5	21·14	23·41
Fe_2O_3	4·88	4·92	2·35	2·46	2·71	2·70	1·95
MgO	0·40	0·34	1·02	0·74	1·05	1·56	0·78
CaO	0·02	0·04	0·07	0·27	0·16	0·25	0·22
Na_2O	0·24	0·19	0·49	0·17	0·32	0·75	0·18
K_2O	0·09	0·08	3·48	1·97	2·91	3·56	1·11

1 Average of 7 roof samples from abandoned Daud coal pit at Jhampeer
2 Average of 5 roof samples from Daud coal pit No. 11 at Jhampeer.
3 Average of 82 seatearths from South Wales [1].
4 Mean of 45 seatearth analyses, after Ennons and Scott (1924), taken from [8].
5 Mean of 42 seatearth analyses, after [8].
6 Mean of 19 seatearths, after [6].
7 Mean of 35 seatearths, after [10].

is present as kaolinite with the same formula as given above. Free silica can then be calculated by subtracting the silica in kaolinite from the total silica. The calculated mineral composition of the Jhampeer clays is: kaolinite 88%, free silica (quartz) 2%, and other minerals such as iron and titanium oxides10%.

Table 3 reports and compares the minor and trace elements in ppm from Jhampeer, Lakhra and South Wales (U.K.). As might be expected from the low K content, the Rb content at Lakhra and Jhampeer is very low as compared to the sediments adjacent to coals in South Wales. The values of the zirconium content are relatively high. Comparison between the roof shales from Lakhra and Jhampeer indicates that the Jhampeer roof rocks contain more sulphur and less zirconium.

The elements in Lakhra and Jhampeer roof rocks are located in various minerals. Silica may be present as free silica (quartz) or as combined silica (in clay minerals). Titanium may be present as anatase (as its reflections were observed in X.R.D. diffractograms), but some of it may be present in the kaolinite structure. Baqri [1] found a positive relation between kaolinite and titanium in coal-bearing strata. The aluminium is present mainly in clay minerals, but the possibility of amorphous aluminium oxides cannot be ignored. Iron may be present as iron minerals sush as siderite, pyrite and also as amorphous oxides. Magnesium may be present in the clays or in iron minerals such as siderite. The minor and trace elements may be present in the structure of the clay minerals or absorbed on thier surfaces. Thus rubidium is generally found in the illite structure since it follows potassium in its geochemical behaviour [3].

Differences in the chemistry and mineralogy as between the Lakhra and Jhampeer clays and other refractory clays are probably due to differences in original source material. The much lower illite content and the higher titanium and zirconium contents suggest that more intensive weathering and leaching of the source rocks have taken place in the case of Lakhra and Jhampeer clays.

Industrial Uses

The samples were finely ground and thoroughly mixed with water to make a 10-cm long mould. The moulds were dried in an oven at 60°C overnight and fired at 1200°C for 3hr. The shrinkage after oven drying and firing was recorded. Table 4 records the mean shrinkage of Lakhra and Jhampeer fireclays in comparison with English and Scottish fireclays. The Lakhra clays gave 3–4% shrinkage and Jhampeer clays 2–3%. This is very similar to the values reported from English and Scottish clays The clays assuemed a pinkish colour and did not disintegrate after firing. Table 5 compares the chemical and mineralogical composition of the standard fireclays from England and Scotland, which are mined and used in the clay industry. Worrall [11] stated that for high refractoriness, the alumina content should be high (approaching 30–40%) and the alkali content low (less than 1%), since high alkali contents (in the form of mica) increase firing contraction. Alkali oxides are strong fluxes and therefore produce a high proportion of liquid at firing temperatures. Keeling [5] stated that illitic clays are less refractory than the kaolinitic clays since they contain more alkalies. He also pointed out that a high proportion of quartz in clay often leads to a low strength product. Comparative studies of the Lakhra and Jhampeer clays suggest that these clays are highly desirable refractory clays and could be used for the manufacture of tiles, sanitary ware, engineering bricks and refractories.

TABLE 3. MEAN ANALYSES OF TRACE AND MINOR ELEMENTS FROM JHAMPEER, LAKHRA AND SOUTH WALES ROOF ROCKS.

	1	2	3	4	5
Rb	8	4	18	21	173
Sr	38	33	174	156	143
U	9	5	2	2	..
Zr	499	503	641	635	205
Y	34	33	32	31	34
Nb	37	34	36	34	18
Mo	..	..	..	..	..
Th	8	9	16	15	15
Pb	19	16	21	20	26
As	14	17	1	..	10
Br	..	3	1	..	..
P	147	146	354	359	416
S	7498	7543	376	386	1750
Cl	754	796	605	518	87
V	188	182	209	216	..
Ga	32	33	29	27	29
Zn	53	59	20	24	91
Cu	78	82	..	..	37
Ni	172	181	165	171	64

1 Average of seven samples from abandoned Daud coal pit at Jhampeer.
2 Average of five samples from Daud Coal pit No. 11 at Jhampeer.
3 Average of six samples from Habib coal pit No. 12 at Lakhra..
4 Average of eight samples from Habib coal pit No. 2 at Lakhra.
5 Average of eighty-six samples from SouthWales roof shales (1).

TABLE 4. MEAN SHRINKAGE AFTER OVEN DRYING AT 60°C AND FIRING AT 1200 °C IN LAKHRA AND JHAMPEER CLAYS AND FIRING SHRINKAGE IN ENGLISH AND SCOTTISH CLAYS.

	Total length cm	Length after oven drying cm	Shrinkage after oven drying %	Loss after firing cm	Shrinkage after firing %
1	10	9·3	7	0·3	3·2
2	10	9·2	8	0·25	2·5
3	10	9·3	7	0·4	4·3
4	10	9·1	9	0·3	3·2
5	..	..	..	..	4·6
6	..	..	..	..	2·8
7	..	..	..	..	3·2

1 Average of 6 samples from Habib coal mine No. 12 at Lakhra.
2 Average of 8 samples from Habib coal mine No 2 at Lakhra.
3 Average of 7 samples from Daud abandoned coal pit at Jhampeer.
4 Average of 5 samples from Daud coal pit No. 11 at Jhampeer.
5 Average shrinkage (%) of fireclays from South Midlands, England.
6 Average shrinkage (%) of fireclays from Yorkshire [11].
7 Average shrinkage (%) of fireclays from Scotland [11].

TABLE 5. COMPARISON OF THE LAKHRA AND JHAMPEER REFRACTORY CLAYS WITH THE FIRECLAYS FROM ENGLAND AND SCOTLAND.

	1	2	3	4	5	6
SiO_2	50·35	42·38	56·50	60·10	59·50	57·40
TiO_2	3·53	4·27	1·50	1·20	1·20	1·20
Al_2O_3	35·27	34·89	26·60	25·70	26·00	26·40
Fe_2O_3	2·54	4·90	2·40	1·70	1·60	1·90
MgO	0·37	0·37	0·50	0·50	0·50	0·50
CaO	0·14	0·03	0·30	0·40	0·40	0·30
Na_2O	0·17	0·21	0·20	0·15	0·10	0·10
K_2O	0·40	0·08	1·40	1·30	1·70	1·20
Kaolinite	85·00	88·0	53·3	52·6	50·4	55·5
Mica	5·0	..	14·8	12·6	15·7	12·1
Quartz	8·0	2·0	25·4	30·0	29·3	26·2

1 Average of 14 samples from Lakhra fireclays.
2 Average of 12 sample from Jhampeer fireclays.
3 Average composition of fireclays from South Midlands, England [11].
4 Average composition of fireclays from Yorkshire, England [11].
5 Average composition of fireclays from Northumberland and Durham, England [11].
6 Average composition of fireclays from Scotland [11].

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