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ORIGINAL RESEARCH ARTICLE

Genesis of Langrial Iron Ore of Hazara area, Khyber Pakhtunkhwa, Pakistan

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ABSTRACT

In this paper, a detailed mineralogical and genesis investigation have been carried out in the seven locations of the Iron Ore in Hazara area. Thick bedded iron ore have been observed between Kawagarh Formation and Hangu For-mation i.e., Cretaceous-Paleocene boundary. At the base of Hangu Formation, variable thickness of these lateritic beds spread throughout the Hazara and Kohat-Potwar plateau. This hematite ore exists in the form of unconformity. X-ray diffraction technique (XRD), X-ray fluorescence spectrometry (XRF), detailed petroghraphic study and scanning elec-tron microscope (SEM) techniques indicated that those iron bears minerals including hematite, chamosite and quartz, albite, clinochlore, illite-montmorillonite, kaolinite, calcite, dolomite, whereas ankerite are the impurities present in these beds. The X-ray fluorescence (XRF) results show that the total Fe_2O_3 ranges from 39 to 56%, with high silica and alumina ratio of less than one. Beneficiation requires for significant increase in ore grade. The petroghraphic study re-vealed the presence of ooids fragments as nuclei of other ooids with limited clastic supply, which indicate high energy shallow marine depositional setting under warm and humid climate. The overall results show that Langrial Iron Ore is a low-grade iron ore which can be upgraded up to 62% by applying modern mining techniques so as to fulfill steel re-quirements of the country.

Keywords: Langrial Iron Ore; Cretaceous-Paleocene Boundary; Geochemistry; Hazara Area; Hangu Formation

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1. Introduction

Iron ore plays a significant role in the development of modern society, which can be used to extract iron, steel and manufacture other alloys. Iron usually occurs in a number of geological environments from deep seated basic igneous intrusion to late stage hydrothermal igneous and metamorphic and sedimentary environment[1]. Iron occurs in banded sedimentary rocks of Precambrian age as residual or replacement deposits. Iron also occurs as oolitic ironstone in Paleozoic to Cretaceous sedimentary successions of the world. The reported massive iron ores in Precambrian igneous rocks of Kirana Hills are magnetite and hematite[2]. Pyrometasomatic deposits formed due to replacement of limestone or volcanic rocks by magnetite.

Occurrences of iron ores are widespread in Pakistan. Mainly these are along unconformities, volcanogenic ores or contact metasomatic deposits. Iron ores have been reported from Indus platform zone, Foreland sedimentary belt, Himalayan crystalline belt, Ophiolitic thrust belt and Chagai magmatic arc[1]. Massive iron ore of Precam-brian age from Kirana Hills near Sargodha consist of magnetite and hematite[2,3].

In Himalayas Foreland Fold and Thrust Belt (HFFTB) of Pakistan, ironstones of oolitic textures are mainly reported in Mesozoic to early Tertiary formations[2,4-6].

The main objective of the study is to explain the texture, mineralogy and depositional environment of the Langrial iron ore in Hazara-Abbottabad area.

2. Geological and tectonic framework

The Hazara area forms the western limb of the Hazara Kashmir Syntax (HKS) and appears as a crescent shape in the northwestern margin of the Indo-Pakistani subcontinent with a North-East to South-East trending. The crescent shaped Hazara area is bounded by Panjal Thrust and the Main Boundary Thrust (MBT) and number of local thrust faults that lie in a similar NE-SW trend (**Figure 1**).

The oldest rock unit of the area is Precambrian Hazara Formation represented by Hazara Group Slates and sedimentary rocks[7]. Hazara Formation is unconformably overlain by Abbottabad Formation of Cambrian age composed of quartzose sandstone, shale, siltstone, limestone and dolomite[8]. Overlain Mesozoic sedimentary rocks are represented by Samana Suk Formation of Jurrasic age and Cretaceous Lumshiwal and Kawagarh Formation[8,9].

The Paleocene Lockhart Formation is lying above the ironstone beds and Cretaceous Kawagarh Formation (**Table 1**) is lying below the Ironstone beds[8,10,11]. The Hangu Formation is overlain by the Lockhart Limestone[8,10,11]. Iron beds also exist at places in the cores of both limbs of anticlines of Paleocene Lockhart Limestone.

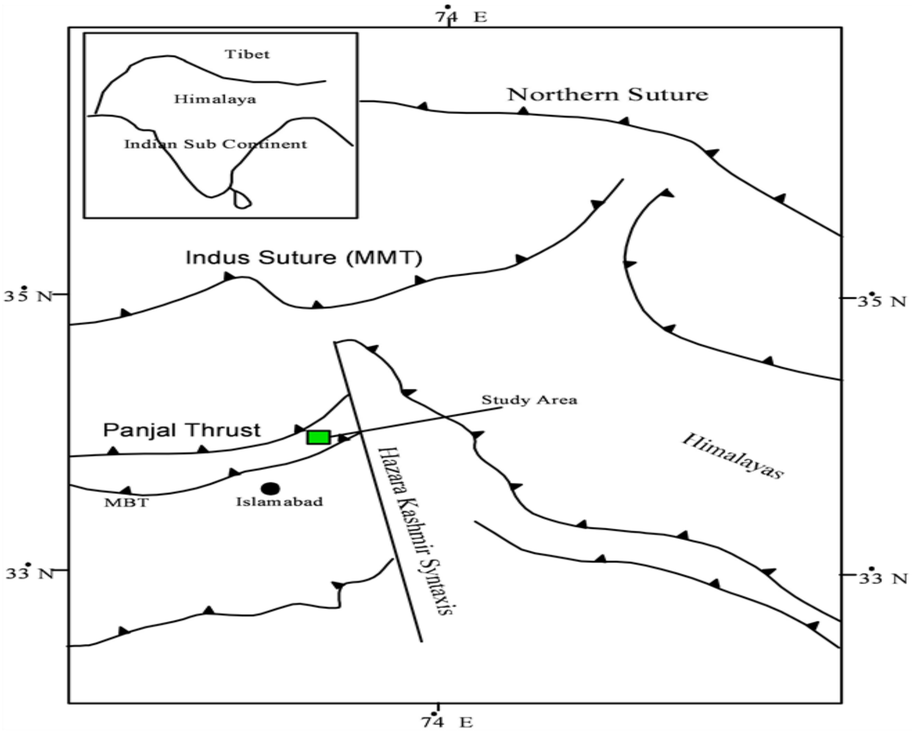

Figure 1. Tectonic sketch map of northern Pakistan[12].

Table 1. Lithostratigraphic unit of the Hazara-Abbottabad area[8]

Age	Formation	Description
Paleocene	Lockhart Limestone	Limestone dark grey to black in colour and containsintercalations of marl and shale.
Paleocene to Cretaceous	Oolitic Iron beds	Ooloitic hematite mixed with chamosite, limonite.
Upper Certaceous	Kawagarh Formation	Grey, olive grey, light grey sub-lithgraphic limestonewith subordinate with marl and calcareous shale.
Upper Certaceous	Lumshiwal Formation	Thick bedded to massive, light grey current bedded sandstone with silty, sandy, gluconitic shale towards the base.
Pre-Cambrian	Hazara Formation	Slate, phyllite and shale with minute occurrences oflimestone and graphite layers.

3. Materials and methods

During reconnaissance survey, seven stratigraphic sections, namely, Bagnotar, Bagan, Durban and Langrial, Danna Noral, Tati Maira, Najaf Pur and Jabri were selected for a detailed study. The study area spreads for about 32 kilometers to the south of Abbotabad. Sedimentary features and orientation and thickness of iron ore beds were examined and 65 samples were collected marked by Global Positioning System (GPS) for subsequent laboratory analysis (**Figure 2**). Bagnotar (Siri) area is located between 34°56'10" N and 73°20' E.

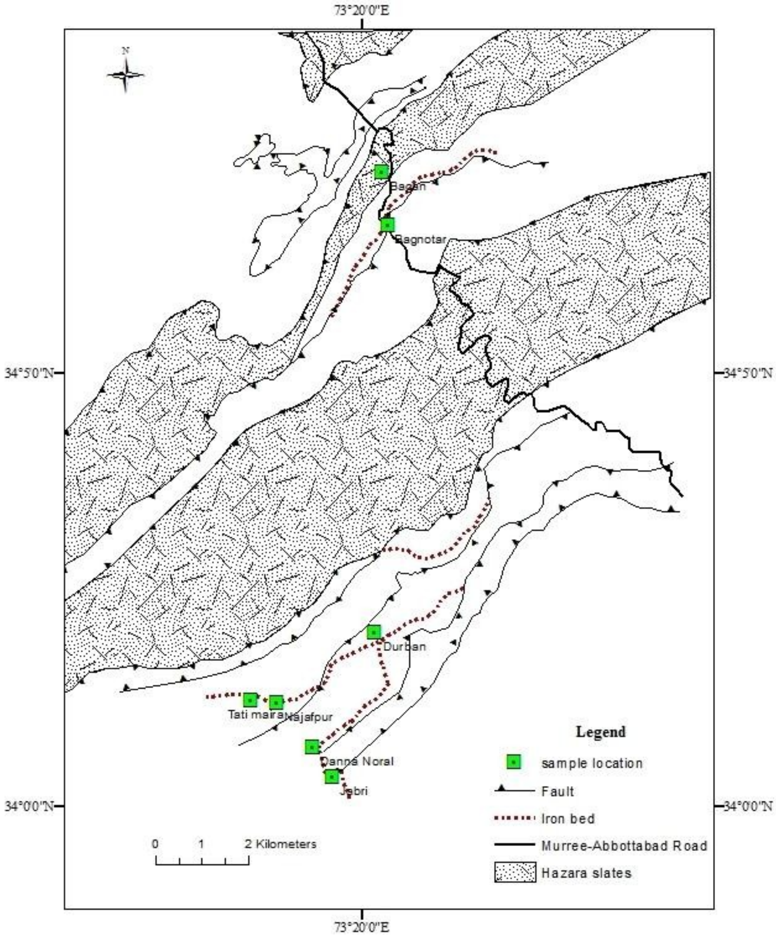

Figure 2. Location map showing geology of the study area.

The strike of the bed is N10° E with a dip varying from 32° SW to 35° SE. Iron bed of 1.5m to 2m have been reported. Iron beds exhibit oolitic to pisolitic hematitic to goethitic appearances, overlain and underlain by Lockhart Limestone due to tectonic disturbance in the area.

Bagan lies between 34°58' N and 73°20'01" E. The strike of the bed is N60° W with a dip of 20° to 45° NE. The thrusted contact between the iron ore and Lockhart formation is observed.

At Durban and Langrial area, the samples have been collected at 34°15' N and 72°17'01" E. The strike of the bed is N35° E and dip is 40° SE. Highly fractured thin to medium bedded iron ore of 2 meter thickness are noted.

Jabri area lies between 34°03'01" N and 73°17' 01" E. The strike of the bed is N60°E and dip is 30° SE. Thin to medium bedded laterite have been reported in this area.

Danna Noral is located between 34°08' N and 72°21' E. The strike and dip of the bed are N60°E, 25° SE respectively. Upper part of iron ore is Kawagarh formation with strike N45°E and 10° SW dip. The thickness of unconformity (i.e., conglomeratic beds) varies from 0.15m to 0.6m at different localities. Upper contact of iron beds is unconformable with Kawagarh formation marked by ferruginous conglomerates whereas the lower contact is sharp with greenish grey to brown flaky shale, the deposit form a synclinal structure.

Tati Maira is located between 34°06' N and 73° 19'01" E with a N30° E strike and a 25 ° NW dip where presents lateritic beds. Najaf Pur area lies between 34°11'04" N and longitude 72°15'01" E with N60° W beds strike and dip 55° SW. The Upper part of the Iron Ore consists of nodular limestone of Lockhart Formation. At Sardhana village near Najafpur, the iron beds show anticline and syncline structure.

3.1 Petrographic techniques

Thin sections and polished slabs of selected samples are prepared at sample preparation section of the Geoscience Advance Research Laboratory, Geological Survey of Pakistan, Islamabad for the Petrographic study under camera fitted Nikon made reflected light polarizing microscope at different magnifications.

3.2 X-ray diffraction analysis

The selected samples were crushed at the crushing and powdering section of Geoscience Advance Research Laboratory of the Geological Survey of Pakistan, Islamabad. The analysis was carried out by Panalytical X'PertPRO Diffractometer (XRD) at 45 Kv and 40 mA with CuK α radiations scanning speed at 0.05°/Sec scanned by X'Pert data collector software and identified with the help of its matching peaks with the existing data base (**Figure 3**).

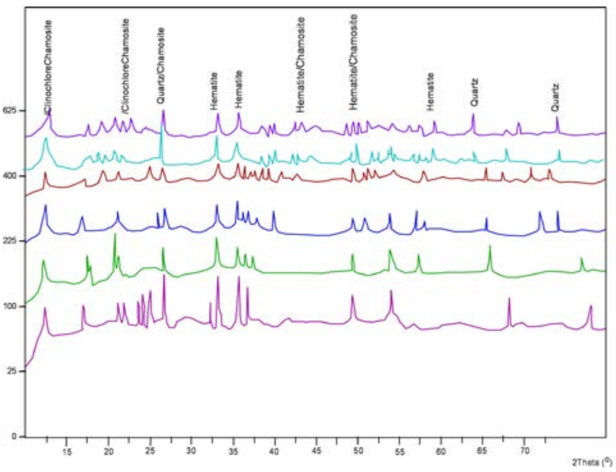

Figure 3. XRD pattern of selected powder samples, where dominant peaks are labeled**.**

3.3 Scanning electron microscope analysis

Thin section of selected samples was analyzed using scanning electron microscope (Model JEOL JSM 6610LV) to study microscopic structures (**Figure 4**). Compositional analyses were carried out using EDS (OXFORD X-MAX 20mm^2). Carbon coating is well applied to the thin section to avoid charging of the particles during observation.

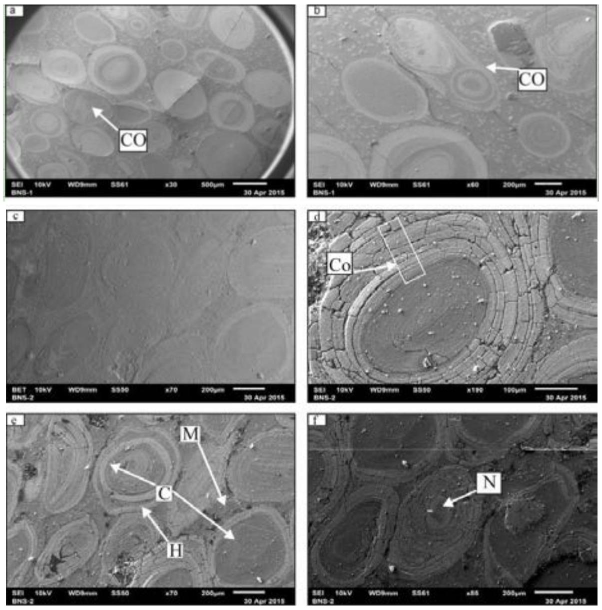

Figure 4. Scanning electron microscopic (SEM) images of the iron ore thin section. **a, c, e)** concentric and elliptical ooids; **b)** composite ooid; **d, f)** concentric lamination in cortex of the ooids**.** (**Note:** CO is composite ooids; Co is cortex; C is Chamosite; H is hematite; M is matrix and N is nucleus.)

3.4 X-ray fluorescence spectrometry

The chemistry of selected samples were analyzed by using Panalytical Axios WD-XRF for all major element using fundamental parameter method with the help of glass bead by taking 1:10 ratio of volatile free sample and lithium tetra borate.

4. Results and discussion

Petrographic and Mineralogical study by XRD indicates the most prominent iron minerals are hematite and chamosite (**Figure 5**). After interpretation of mineral on XRD with the help of its crystal system and d-spacing values, other minerals are quartz, albite, chamosite, clinochlore, illite-montmorillonite, kaolinite, calcite, dolomite and ankerite (**Figure 3**).

XRD study has direct influence on the commercial value of the ore and processing of the ore regarding carbon footprint and coal consumption. The study of the gangue minerals is necessary for taking steps for beneficiation of ores. According to results of X-ray diffraction analysis, it can be safely suggested that all ore samples from studied area were observed to be mainly of a hematitic nature.

Certain localites like Danna Noral and Najafpur shows hematite ores with iron content (40-50% Fe), such hematite deposits require little beneficiation.

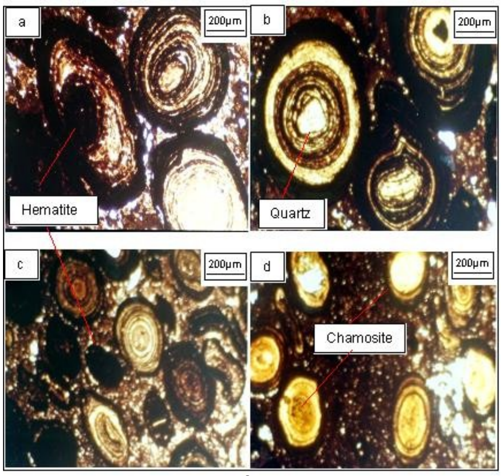

Figure 5. Photomicrographs of selected samples collected from the study area showing **a)** concentric ooids with hematite and chamosite rich concentric lamination. **a & d)** Elongated ooids due to early compaction. **b)** Nucleus of oolites consists of quartz, feldspars, hematite and rock fragments. **c & d)** ooids consist of pure hematite whereas others are comprised of entirely chamosite**.**

The geochemical analysis shows high silica content in the iron ore from Bagnotar area as compared to Danna Noral and Najafpur (**Tables 2** and **3**; **Figure 6**). SiO_2 ranges between 9-35% here, it is necessary to mention that three samples BNS-4, 9 and 10 belongs to Lockhart Limestone having low SiO_2 and Al_2O_3 varies between 10-18%. Beneficiation requires for reducing high alumina content. Crushing, scrubbing and washing of iron ores techniques are being adopted to reduce alumina in some extent[13].

The total Fe_2O_3 ranges from 37.2 to 72.74%. Average iron (Fe) concentration in samples from Bagnotar area is 37.89%. The trace constituents are calcium, magnesium, sodium, potassium, manganese, and phosphorus oxides. These are low-grade iron ore which can be upgraded up to 62% by applying modern mining techniques[14].

Table 2. Chemical composition of samples collected from Bagnotar area

	BNS-1	**BNS-2**	**BNS-3**	**BNS-5**	**BNS-6**	**BNS-7**	**BNS-4**	**BNS-9**	**BNS-10**
SiO_2	20.24	17.32	18.52	28.04	27.46	23.3	14.53	6.20	0.00
TiO_2	0.7	0.45	0.42	0.63	0.79	0.58	0.14	0.348	0.065
Al_2O_3	12.44	10.53	10.46	10.98	17.63	11.88	0.56	2.04	1.18
Fe_2O_3	48.25	54.21	56.31	45.06	39.45	45.24	2.81	4.40	3.68
MnO	0.04	0.1	0.08	0.13	0.03	0.03	0.26	0.04	0.189
MgO	2.58	2.31	2.28	2.27	2.0	2.01	7.42	7.77	0.62
CaO	4.12	3.06	2.62	3.07	2.98	2.96	37.3	42.13	52.5
Na_2O	0.23	0.77	0.39	0.31	0.13	0.41	0.25	0.00	0.08
K_2O	0.09	0.1	0.14	0.17	0.21	0.07	0.18	0.86	0.26
P_2O_5	1.34	1.41	1.41	1.17	0.7	1.10	0.03	0.009	0.124
SO_3	-	-	-	-	0.08	-	-	0.90	0.025
LOI	9.88	9.27	7.36	8.17	9.28	12.43	36.53	35.00	41.30
Fe	33.73	37.89	39.36	31.50	27.58	31.62	1.96	3.08	2.57
Al/Si	0.61	0.60	0.56	0.39	0.64	0.51	-	-	-

Table 3. Chemical composition of samples collected from Danna Noral, Najafpur Jabri and Tatimaira areas

	DN-29	**DN-30**	**DN-31**	**NF-62**	**NF-63**	**NF-64**	**JB-26**	**JB-27**	**TM-33**	**TM-34**
SiO_2	12.51	12.61	9.75	21.04	11.37	13.54	18.72	19.49	35.38	30.76
TiO_2	0.84	0.68	0.27	0.84	0.52	0.55	0.60	0.57	0.54	0.51
Al_2O_3	9.70	10.30	7.62	18.84	8.47	10.79	7.31	12.71	10.02	10.53
Fe_2O_3	63.93	59.13	72.74	37.20	69.73	57.75	52.94	50.58	38.82	46.59
MnO	0.02	0.02	0.01	0.04	0.53	0.05	0.01	0.30	0.16	0.09
MgO	1.55	1.50	1.19	4.79	2.65	3.20	1.78	3.47	1.31	1.29
CaO	2.29	5.80	1.32	1.26	3.34	3.54	1.05	1.09	1.79	2.20
Na_2O	0.48	0.25	0.38	2.11	0.00	0.18	1.94	0.0	2.12	0.0
K_2O	0.02	0.02	0.03	-	0.01	-	0.04	-	0.02	0.02
P_2O_5	1.53	1.37	0.78	0.24	0.55	2.41	0.15	0.30	1.08	1.38
LOI	7.15	8.32	5.91	13.65	8.64	7.99	15.96	13.22	8.78	9.33
Fe	44.69	41.33	50.85	26.00	48.74	40.37	37.01	35.36	27.14	32.57
Al/Si	0.77	0.82	0.78	0.89	0.74	0.79	0.39	0.65	0.28	0.34

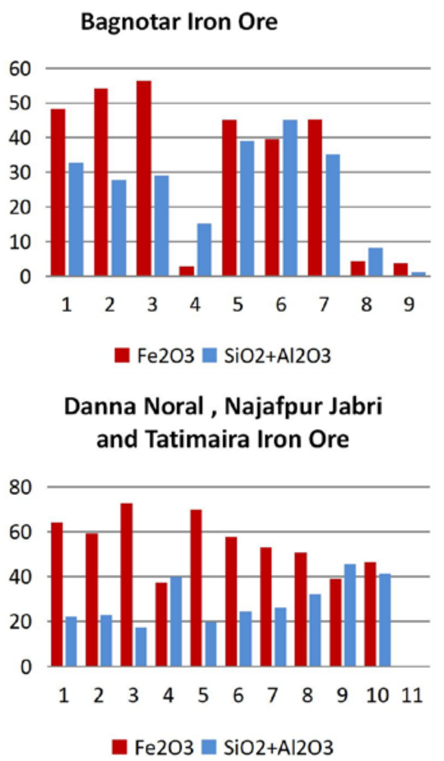

Figure 6. Iron, aluminum and silica ratios of selected samples collected from northern part, Bagnotar area and southern part including Danna Noral, Najafpur Jabri and Tati Maira areas**.**

Samples from Danna Noral (41-50% Fe) contain high iron content as compare to the Najafpur (26-48% Fe), Jabri (35-37% Fe) and Tati Maira (27-32% Fe). Concentration of SiO_2 and Al_2O_3 are also low as compared to Bagnotar area. The beneficiation should require gravity separation as well as magnetic separation in Langrial Iron ore deposits.

The adverse effects of high alumina to silica ratio (ideally it should be < 1) is harmful to blast furnace as well as sinter plant[13]. Aluminum silica ratio in our samples is less than 1, which is good for beneficiation (**Table 2** and **3**), while alumina raises the melting point of slag and increases fuel consumption in the furnace.

Microscopic mineralogical studies indicated that hematite is the major iron oxide mineral with quartz and chamosite. XRD studies revealed hematite as the major minerals with subordinate amounts of quartz, albite, chamosite, clinochlore, illite-montmorillonite, kaolinite, calcite, dolomite and Ankerite confirming the microscopic findings.

These oolites consist of concentric layers of the iron and iron rich clays chamosite, so these deposits contain an iron oxide hematite with iron clay (chamosite). The photo micrographs show that the ratio between hematite and chamosite varies. Thus, some ooids consist of almost pure hematite (**Figure 5c**) whereas others are comprised of almost entirely chamosite (**Figure 5d**). The nucleus of the ooids are variable and consist of quartz, as well as feldspars, iron oxides, broken pieces of older ooids and other rock fragments (**Figure 5b**). These compositional variation are also noted through back scattered image of scanning electron microscope (**Figure 4**).

The presence of ooids fragments as nuclei of other ooids indicates that the formations of ooids are the product of a high energy shallow marine environment with limited clastic sediment supply.

5. Depositional settings

The hematite-chamositic composition of the deposits indicates intense chemical weathering under humid continental conditions[15]. Such conditions are usually common in humid equatorial settings[16]. Paleo-geographic reconstruction studies indicate that the area occupied an equatorial position during the deposition of the Langrial iron ore[10]. Ooids are generally considered to be the product of high energy shallow water and agitated environment. Such oolitic texture is the characteristic of the Phanerozoic iron deposits[17]. These ooids consist of concentric rings of iron bearing minerals and are formed by chemical process in sedimentary rocks[18]. The presence of oolitic ironstones usually shows high relative sealevel, warm and humid climate as well as favored chemical weathering[10,19,20]. In the study area, the iron ores occur in the lower part of the Palaeocene Hangu Formation. In the Salt and Surghar ranges, this lower part of the Hangu Formation is interpreted to have been deposited in terrestrial conditions, while the upper part of the formation indicates transitional settings with the overlying marine Lockhart Limestone[21]. On the contrary, recent studies have interpreted shallow marine and deltaic environment of deposition for the lower part of the Hangu Formation[16]. The present study reports abundance bioturbation and biogenic activity. Further, abundance of burrowing in the sandstone of the Hangu Formation, to go with the presence of marine fossils in the formation indicates marine deposition[22,23]. However, brackish water pollens of the palm genus Spinizonocolpites are reported from the lower part in these areas[16]. Strong influence of subaerial exposure and lateritic palaeosol development is indicated for the Langrial iron ore[16]. A rise in relative groundwater base level may have triggered swampy/boggy land formation to spread over lateritic paleosol deposits that formed on weathered paleo-surfaces of the Cretaceous/older strata[16]. In addition, tectonic controls probably combined with eustatic and climatic controls, influenced the development of the laterites in the area[24].

The tropical, humid climatic conditions coupled with intense chemical weathering produced the initial Fe-rich material, probably as amorphous hydroxides or goethite during the lateritization processes. Marine transgression along the continental margin favored the preservation of these sediments[10]. The high PCO_2, probably associated with the Deccan Trap basalts[25] may have created a stratified ocean with reducing conditions and negative Eh. Such conditions favor Siderite ($FeCO_3$) precipitation, a phenomena also identified in modern oolitic ironstone samples, precipitating at negative Eh, (under reducing conditions) and low sulfide activity (PS2-) in sediment pore-waters[18,26,27]. However, since there are abundant dissolved sulfates dissolved in marine water, thus low sulfide activity is rarely attained in marine sediments. Further, the Deccan trap activity may also have contributed to additional amount of sulfur in the atmosphere. Hence the initial form of the iron oolite sediment appeared to have been accumulated under non-marine or brackish conditions[10].

Further, the bacterial decomposition of the organic matter, following the Cretaceous/Palaeogene mass extinction event may have created/added to the anoxic conditions that accommodate siderite precipitation[28]. The diagenetic processes, following the formation and consolidation of the initial ooids, may have then converted these into hematite-chamosite ooids[29]. The episodic basaltic volcanic activity of the Deccan Trap may have created a fluctuating oxic to anoxic conditions[28] that were favorable for the formation of chamosite ($(Mg,Fe)_3 Fe_3(Si_3Al)O_{10}(OH)_8$), without sulfide activity as the sulfur rich aerosols settle quickly from the atmosphere[18,30]. These conditions also provided the opportunity for the hematite precipitation as the latter is stable under moderate to strong oxidizing conditions[18,26]. These fluctuating environmental conditions must have had a strong control on the depositional and early diagenetic history of the Langrial Iron Ore.

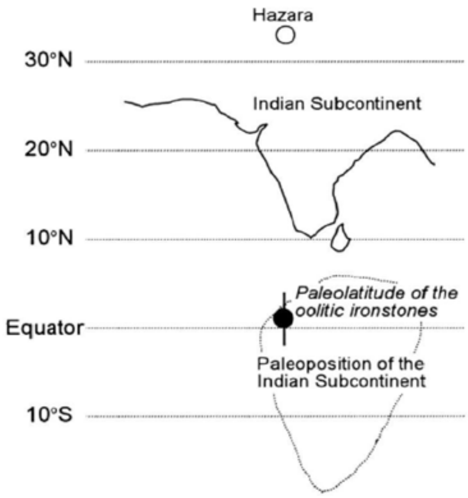

Figure 7. Fluctuating environmental conditions of Hazara area**.**

6. Conclusion

Langrial Iron Ore occurs in the basal part of Hangu Formation of Paleocene age mainly of hematitic nature with gangue minerals. The deposits are low grade Iron ore mostly consisting of hematite and chamosite in nature with an average Fe_2O_3 content of 52.37%. The mineralogical and petrological interpretation indicated that the deposit was formed under warm humid climate. This low grade iron ore has silica aluminum ratio of less than 1, which is good for beneficiation. Langrial Iron Ore has economic significance proved by combined petroghraphical, geochemical and mineralogical data and the value can be further increased by the application of modern metallurgical processing technologies. The oolitic layers vary in numbers and will lead to a high difficulty to separate iron from gangue minerals in the oolitic structure. It is necessary to carry out a very fine grinding to achieve the mineral separation. These deposits show potential to be economically viable in the present market.

Conflict of interest

The authors declare that they have no conflict of interest.

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