



A mixed hydrogenous and hydrothermal origin of the Bahariya iron ores, Egypt: Evidences from the trace and rare earth element geochemistry



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ARTICLE INFO

Article history:

Received 13 November 2012

Accepted 21 August 2014

Available online 29 August 2014

Keywords:

Iron ores

Bahariya

Egypt

Trace elements

Rare earth elements

Origin

ABSTRACT

This study utilizes the trace and rare earth element geochemistry of bulk ores and in situ LA-ICP-MS analyses of mineral grains to clarify the controversy in the origin of the Bahariya sedimentary iron ores, Egypt. Different types of iron ores were identified in the Bahariya Oasis including high grade, high-Mn, high-Ba, oolitic, and ochreous iron ores. Hematite and goethite are the main constituents of the studied ores with some manganese oxide and hydroxide minerals (pyrolusite, bixbyite, cryptomelane, aurorite, romanechite, manjiroite, and pyrochroite). Barite is common in the high-Ba ore, while some quartz, calcite, and halite are detected in the oolitic iron ore. High-Mn iron ores are characterized by higher contents of SiO₂, Al₂O₃, MgO, CaO, Na₂O, and K₂O, while high-Ba iron ore has higher TiO₂ contents. Analyses of bulk ores and in situ mineral grains are characterized by substantially high contents of Ba (average of 9497 ppm); also Zn and Sr occur in considerably high concentrations (averages of 4263 and 429 ppm, respectively). Mn-rich ores and Mn-bearing minerals show relative enrichments of trace metals compared to the Mn-poor ores probably due to the ability of Mn-bearing minerals to fix such trace metals by adsorption, absorption, and/or replacement. The Σ REE ranges between 2.6 and 80 ppm with an average of 23 ppm. High-Mn ores show higher Σ REE (average of 61 ppm) compared with the low Mn ore (average of 13 ppm). The oolitic iron ore shows very low Σ REE content (7 ppm). Red and yellow ochers from El Gedida mine have similar Σ REE values (38 and 39 ppm, respectively), while red ochre from the Ghorabi area has relatively higher Σ REE (57 ppm). Chondrite-normalized REE patterns of all types of iron ores, whether in the form of bulk ores or Fe- and Mn-bearing grains, have LREE enrichment relative to HREE as shown by (La/Yb)_N ratios that vary from 1.7 to 29.4. Majority of the bulk samples and mineral grains have negative Eu anomalies with Eu_N/Eu* ranges from 0.68 to 0.8. However, bulk samples of one high grade and high-Ba ores as well as some of the Mn-bearing grains from El Gedida ores show positive Eu anomalies with Eu/Eu* ranges from 1.1 to 17.7. With few exceptions, all bulk ores and mineral grains show negative Ce anomalies with Ce/Ce* range from 0.28 to 0.96. A seawater precipitation (hydrogenous to hydrothermal exhalite) is proposed for the Bahariya iron ores. The hydrogenous origin is suggested based on the occurrence of high-Mn iron ores in the base of the iron ore succession, oolitic texture of some of these ores, Si–Al plot, low La/Ce ratios, high Y/Ho ratios, and LREE-enriched patterns with negative Eu anomalies of most of the bulk ores and mineral grains. The hydrothermal contribution to the source of these iron ores can be evident from the high contents of some trace elements such as Ba, Zn, and Mo, and plots the analyzed samples in the hydrothermal fields of the Fe–M–(Ni + Co + Cu) * 10 ternary diagram, (Co + Ni)–(As + Cu + Mo + Pb + V + Zn) and (Co + Ni + Cu–Co/Zn) binary plot, low Σ REE concentrations and positive Eu anomalies and high La/Ce ratios in some of the analyzed samples. The hydrothermal contribution looks for local effect and is restricted to El Gedida area.

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1. Introduction

Economic iron ores (270 million metric tons of iron ore deposits) with an average of 47.6% Fe occur in the Bahariya Oasis (Said, 1990). The iron ores are exploited now as raw materials for the Egyptian Iron

and Steel Company, Helwan, Egypt. Due to their geological and economic significance, such deposits have been subjected to several investigations. However, the genesis of the ore has been a matter of scientific discussions for a long time. Attia (1950) and El Shazly (1962) suggested an epigenetic–supergene origin in which the deposits have been formed later than the Eocene limestone by supergene solutions. Both authors favored a deposition of leached iron in a shallow water lacustrine environment with subsequent replacement of the underlying Middle Eocene

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limestone. Akkad and Issawi (1963) suggested a syngenetic–supergene origin where the ore deposits have been formed contemporaneously with the Eocene limestone by surface marine or fresh water in either shallow marine or lagoonal lacustrine environments. Gheith (1955), Basta and Amer (1969), and Nakhla (1961) postulated an epigenetic–hypogene origin where the ore minerals were formed by metasomatic replacement by hydrothermal solutions. Tosson and Saad (1974) discussed the actual source of the iron-bearing fluids and found it to be of two types, one related to a volcanogenic source and the other derived from weathering processes. El Sharkawi et al. (1984) distinguished three genetic types for El Gedida iron ores: (1) hydrothermal–metasomatic, of a massive nature; (2) mobilized iron and manganese re-deposited in fresh water lakes, possibly through biogenic activity, cavernous, ochreous or massive in nature; and (3) a product of the weathering of glauconitic clays and sands, usually oolitic or pisolitic in texture. El Aref and Lotfy (1985) discussed the karstic significance of the iron ore deposits of Bahariya Oasis. They concluded that the ores are confined to karst features and suggested that they were formed through lateritization processes. Helba et al. (2001) recognized four depositional stages of ore development based on ironstone paragenesis suggesting alternating humid and dry conditions. Based on U isotopes, Dabous (2002) pointed out that the iron ore of Bahariya Oasis is neither lateritic nor volcanogenic. It is epigenetic, of fresh groundwater origin, and formed in two different stages. First, during dry climatic conditions, the iron has been oxidized and leached from the sandstone of the Nubia Aquifer by the upward-movement of groundwater and deposited in the overlying pre-existing Lower–Middle Eocene karstic limestone. Second, during wet pluvial periods, the iron has been leached from the glauconitic clayey beds, infiltrated downwards and deposited on the underlying primary ore. During the two stages, initial structures, variations in pH–Eh, and biological activities were the main factors controlling the deposition and character of the ore. Baioumy and Hassan (2004) suggest that the weathering of overlying glauconites of the Hamra Formation can be the source of part of iron in the Bahariya iron ore.

Although many geochemical investigations have been performed on the iron ores from the Bahariya Oasis including major and trace elements as well as U isotopes, nothing was reported on the distributions and geochemistry of rare earth elements in these studies. In addition, the source and origin of these iron ores are still controversial as shown above. The current study shows, for the first time, the geochemistry of rare earth elements along with the trace and major elements of both bulk ores and Fe- and Mn-bearing minerals of samples represent the different iron ores including high grade, high-Ba, oolitic, high-Mn, and ochreous iron ores from El Gedida and Ghorabi areas to examine the source(s) of these deposits and clarify their origin.

2. Geology and stratigraphy of the iron ores

The Bahariya Oasis is a large depression in the Western Desert of Egypt with hot, arid climatic conditions. It is located about 270 km SW of Cairo and 180 km west of the Nile Valley (Fig. 1). Economic iron ore deposits with an average of 47.6 wt.% of Fe as Fe₂O₃ (Said, 1990) occur in the Bahariya Oasis. The ores are situated at the northern part of the depression and extend over an area of 11.7 km² with a thickness varying from 2 to 35 m, averaging 9 m (Said, 1990). The iron ores in the northern Bahariya Oasis are confined to the lower part of the Middle Eocene limestone of the Naqb Formation. The Naqb Formation ranges in thickness between 8–11.6 m and is composed of hard yellowish brown crystalline limestone with marl intercalations (El Bassyony, 2005). The iron ores occur in three areas which include the Ghorabi–Nasser (3.5 km²), El Harra (2.9 km²), and El Gedida (15 km²) areas. Since the current study focuses on the iron ores in the Ghorabi and El Gedida areas due to their relatively large reserves of iron ores, detailed geology and stratigraphy of these two areas will be addressed in this section.

The El Gedida iron mine is an oval shaped depression up to 15 km² in area, situated within the degraded cone hills of the Naqb Formation. The

central part of the depression is characterized by a high relief (up to 254 m above sea level). The low Wadi area, up to 198 m above sea level, surrounds the high central area, which comprises the Cenomanian sandstone and sandy clays of the Bahariya Formation at the base, overstepped by the main Lutetian iron ore successions of Naqb Qazzun Sequence. In the Eastern and Western Wadi areas, the ore successions are truncated unconformably by late Lutetian–Bartonian glauconitic sediments with lateritic ironstone interbeds of the Hamra Formation. The iron ore sequence attains its maximum thickness, up to 35 m, in the Western and Eastern Wadi areas, and it is much reduced in the high Central area where it attains a thickness of only 11 m. This iron ore sequence consists of a pisolitic–oolitic ironstone unit followed by bedded karst iron ores intercalated with ferruginous mudstones (Fig. 2A) (e.g. El Aref et al., 1999). The thickness of the overlying glauconitic sandstone of the Hamra Formation varies in thickness from ≤25 m in the Western and Eastern Wadis areas to <1 m in the high central area. According to Mesaed and Surour (1999), the glauconite facies of El Gedida mine area represents deposition in a shallowing-upward regime.

The exposed sedimentary sequence at the Ghorabi area (Fig. 2B) starts at its bottom with the Bahariya Formation that is composed of moderately hard gray to yellowish gray clayey sediments and is overlain directly by a number of glauconitic sandstone beds range in thickness from 20 cm to 1 m and are separated by iron-rich bands and concretions. Glauconite of this horizon occurs as oval, rounded, fine to medium-grained (100 to 200 μm), moderately- to well-sorted, pale green to yellowish green pellets in smectite and iron oxides matrix (Baioumy and Bulis, 2012). This glauconite is considered by Baioumy and Bulis (2012) as parautochthonous glauconite formed from detrital smectite precursor through diffusion of K from the seawater–sediment interface in a Transgressive System Tract (TST). The total thickness of glauconite-bearing unit is approximately 4 m and is overlain by the Naqb Qazzun Sequence starting with yellowish white soft to moderately hard, laminated clayey sediments of about 3 m thick and intercalated with iron-rich bands and concretions. Overlying these yellowish white sediments, the iron ores ranging in thickness from 6 to 10 m occur as massive and brown iron ore with some reddish bands of iron ocher. Salama et al. (2012) subdivided this succession into lower and upper sequences. The lower sequence is a lagoonal manganiferous and fossiliferous ironstone facies association. The upper sequence is a peritidal microbially mediated stromatolitic and nummulitic–oidal–oncoidal ironstone facies association. The iron ore beds are generally capped by an alluvial cover and/or quartzites of approximately 50 cm in thickness.

3. Materials and methods

Selected samples represented the different types of iron ores in the Ghorabi and El Gedida areas including high grade (Fig. 3A), oolitic (Fig. 3B), high-Mn (Fig. 3C), high-Ba (Fig. 3D), red ocher (Fig. 3E), and yellow ocher (Fig. 3F) iron ores were subjected to detailed petrographical, mineralogical, and geochemical investigations. Polished sections were prepared and investigated under the Olympus optical microscope (Model: BX 51). Sample powders were analyzed for their mineralogical composition using a Philips PW 1730 X-ray diffraction (XRD) technique. Operating conditions were 40 kV and 25 mA. Mineralogical and petrographical investigations were conducted at the Central Metallurgical R & D Institute (CMRDI), Cairo, Egypt.

Fused disks prepared from three bulk ore samples were analyzed for their major oxides using Philips PW 2400 X-ray fluorescence (XRF) spectrometer at Tohoku University, Japan. Tube voltage and current for W target were 40 kV and 60 mA, respectively. Loss on ignition (L.O.I.) was obtained by heating sample powders to 1000 °C for 6 h. Trace and rare earth elements of three bulk ore samples were determined by inductively coupled plasma mass spectrometry (ICP-MS) at the ACME Lab., Canada. The sample powders were digested with 2 ml conc. HF in capped Teflon bombs on an electrical hot plate (~150 °C)

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