Iron ores of Southern Belgium: much more than hematite

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1. INTRODUCTION

Wallonia is particularly rich in iron ores. About 3000 occurrences and extraction sites of iron ores were identified after analyzing historical sources, mining maps, geological studies and toponymy. The latter preserved more traces of mining activities than the landscapes themselves. Most of the iron ores were mined since the Antiquity. In its chronicles De Bello Gallico, Julius Caesar (58-50 BC) already noticed the mining skills of the Nervian and Ebu-rons peoples. During the High Middle Age, the Entre-Sambre-et-Meuse area is thought to be one of the most important mining and iron industry center of Western Europe (Pleiner, 1964; Gillard, 1971). Legends tell that the cast iron was discovered in Liège at the beginning of the 14th century, where the blast furnace was also invented (Pasquasy, 2005). Nowadays, the blast furnace is known to have been invented in England by A. Darby who first introduced the use of coke.
2. GEOLOGY OF THE IRON ORES

The Walloon iron ores can be grouped into three categories: (1) sedimentary ores, (2) weathering deposits and (3) lead-zinc sulfides-related iron ores. With a few exceptions, all of the iron ores are hosted in Palaeozoic sedimentary rocks (Devonian and Carboniferous) and, to a lesser extent, in Jurassic and Paleogene rocks (Fig. 2).

The iron oxides and hydroxides are the dominant components of the ores. The iron oxide (hematite, Fe₂O₃) is a dense, black, grey or red mineral occurring in massive or granular (oolitic) form or, more rarely in Belgium, as crystals. Theoretically, it contains up to 70 % of iron but due to frequent impurities, the iron content of the ore is usually less than 50 %. The goethite (FeO(OH)) is an iron hydroxide occurring in dark or yellowish massive, concretionary or pulverulent forms. Goethite is often associated with limonite, a general term used for minerals composed of hydrated iron hydroxides (FeO(OH),nH₂O). Limonite results often from the weathering of other iron minerals, such as pyrite. Pyrite and marcasite are two common iron sulfides (FeS₂) not used as an iron ore but mined for the production of vitriol (iron sulfates) and sulfur. Iron silicates (glauconite, chlorite, berthierine) are common iron minerals occurring as grains (oolids) within sedimentary rocks but of no industrial utility if not weathered into limonite.

2.1. Sedimentary iron ores

Because the overall geological characteristics and the petrographical description of the Belgian hematitic oolitic ironstones aim the papers of Goe maere et al., 2016: volume 1 and Dreesen et al., 2016: volume 1), we will rather focus on the other sedimentary iron deposits. Nevertheless, a short description of these hematitic oolitic sedimentary iron ores is given below, because of their geological and industrial importance and their large spatial distribution within Southern Belgium and surroundings areas. They are described in an ascending stratigraphical order.

2.1.1. The Middle Devonian (Eifelian) hematitic oolitic ironstone (‘couche d’oligiste oolithique du Couvinien’)

This first hematitic oolitic ironstone is composed of 2 to 6 individual centimetric oolitic ore layers separated by shales and dolomitic sandstones. The ferruginous ooids are rare but hematitized bioclasts are frequent (‘fossil ore facies’ according to Dreesen et al., 2016: volume 1). Red-coloured facies are dominant but some layers richer in siderite (iron carbonate) are grey to green in colour (Cayeux, 1911). The mean iron content varies from 35 to 42 % (Delmer, 1913). The oolitic ironstone horizon stratigraphicaly forms the top of the Emsian Hierges Formation (formerly called the
‘Assise de Bure’) but locally it grades into younger strata (Eifelian, Saint-Joseph Formation). It extends from the Wignehies (France) to Couvin areas and reaches a maximum development south of Chimay. A second outcrop area is situated eastward, between Wellin and Champlon-Famenne (Fig. 1).

2.1.2. The Frasnian hematitic oolitic ironstone (‘couche d’oligiste oolithique du Frasnien’)

A hematitic oolitic ironstone horizon occurs at the extreme base of the Frasnian: it is a 0.2-1 m-

Fig. 2 – Stratigraphic position of the main sedimentary rocks-related iron ores of Wallonia. Illustrated ores (boxes) were the most important from an industrial and economical point of view. Fragments of ores c. 10 cm. Abbreviations: Loch. Lochkovian, Prag.: Pragian, Ems.: Emsian, Eifel.: Eifelian, Givet.: Givetian, Fras.: Frasnian, Famen.: Famennian, Tourn.: Tournaisian, IV: Quaternary.
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2.1.3. Hematitic oolitic ironstones from the basal Famennian (‘couche d’oligiste oolithique du Famennien’)

The Clinton-type hematitic oolitic ironstone horizon in the basal Famennian is the most important of all Walloon oolitic ironstones (Fig. 2). It is composed of true ooids and pisoids (microconcoïds), enclosed in a hematitic, sideritic or carbonated cement. The ooids are usually ellipsoidal and flattened (forming the so-called ‘flaxseed ore’, Dreesen et al., 2016: volume 1). This points to the fact that the original material was not entirely hardened when deposited (Dreesen, 1982a-b). The iron content varies from locality to locality and from layer to layer, but a mean value of 35 % is acceptable, the richest layer reaching up to 55 % of iron (Delmer, 1912). The horizon crops out along two inliers parallel to the Sambre and Meuse valleys (Fig. 1). The southern limb ranges from Aisemont to Haltinne and eastward from Ben-Ahin to Engis. The ore is here composed of one to four layers, the thickest reaching 45 cm and the beds are almost vertically disposed in all these localities. The northern limb forms three segments separated by faults, the westernmost ranging from Les Isnes to Marche-les-Dames, the middle one occurring between Houssoy and Vezin and the easternmost occurrence located near Couthuin (Fig. 1). The ore is also present in the Theux area (‘la Heid de Fer’ outcrop, Marion et al., 2013) and in the Vesdre valley (Vaux-sous-Chevremont, Chaudfontaine, Fraipont) but it rapidly thins eastwards, showing ferruginous ooids scattered in shales only, in the Verviers area (Macar & Calembert, 1938). The ore is composed of two to ten individual layers of oolitic ironstones, the thickness of each layer varying from several centimetres to 125 cm (in the Couthuin mine, Delmer, 1912). More to the South, in the Hamoir, Durbuy and Somme-Leuze areas (yellow stars on Fig. 1), the horizon crops out discontinuously and presents a green facies (distal facies), with scarce ooids composed of berthierine, chamosite and chlorites (Anthoine, 1912).

2.1.4. Other hematitic oolitic ironstone horizons within the Famennian succession

Dreesen (1982) recognized five stratigraphically distinct layers of hematitic oolitic ironstones in the Famennian strata. The lower one, identified as level I, is the main ore deposit as described above. The other stratigraphical levels, labeled II, IIIa, IIIb and IV are rather lenticular and scattered within the lower and middle Famennian rocks. They crop out punctually, e.g. in the Hoyoux, Lesse and Hermeton valleys (orange stars on Fig. 1). These ironstones are very thin (a few centimetres at the most). The composition of these horizons is highly variable, mostly composed of hematitized bioclasts (‘fossil ore facies’; mainly crinoids). These horizons were rarely mined because of their lesser geographical extent, thickness and quality.

2.1.6. Other sedimentary iron ores within the Palaeozoic rocks

The ‘couche d’oligiste oolithique du Gedinien’ - a hematitic oolitic ironstone horizon in the Lochkhovien and the ‘couche d’oligiste oolithique du Givetien’ - a hematitic oolitic ironstone horizon in the Givetien - are cited in the literature but only poorly described (Ancion & van Leckwijck, 1947; Fourmarier, 1954). Both probably represent rocks that are locally enriched in hematite but they do not represent true sedimentary ores (Dimanche & Toussaints, 1977). The manganesiferous iron ore of the Lienne valley is a sedimentary deposit transformed and enriched by low-grade metamorphism, but does not display any oolitic facies. It comprises two to six layers of dark or reddish iron and manganese oxides, often laminated (Libert, 1906). The ores have a mean iron content of 20 % and about 5-25 % of manganese (Firket, 1884) and were mined to produce a high-
manganese cast iron in the first half of the last century (Dusart & Dusart, 1991). Several sideritic horizons (nodular iron carbonates) are known throughout the thick sequence of coal measures in the Mons and Liège areas (Kaisin, 1943) and in the Lower Devonian of the eastern Ardennes (Legrand, 1965). Once weathered, these rocks yielded an ore of bad quality, siliceous and argillaceous, that was locally extracted only.

2.1.5. The Minette ore of Lorraine ('Minette de Lorraine')

The goethitic oolitic ironstone horizon of Lorraine, in the southernmost part of Wallonia, corresponds to the northern edge of the large iron ore basin of the Grand Duchy of Luxembourg and Eastern France. In our region, the ore is composed of two thick individual layers (50 cm and 110 cm respectively) interstratified with shales and marls, forming the Toarcian-Aalenian boundary succession (Cayeux, 1922). The ferruginous are composed of goethite – not of hematite – and are enclosed within a carbonate matrix or cement (‘Minette-type’ oolitic ironstone). Its phosphorous content long restricted its use in blast furnaces. The so-called Thomas-Gilchrist de-phosphorization process that allowed the use of minette in furnaces was finalized only in 1878, but mining activities are recorded at least from the beginning of the 17th century (Delhez, 2004). In Southern Belgium, the Musson-Halanzy mine was the last active iron mine in Belgium that finally closed down in 1978.

2.2. Weathering deposits

The weathering-related ores – secondary ores, replacement ores or impregnation ores after Routhier’s classification (1963) – are the dominant ones, both in term of quantity and variety in Southern Belgium. All these weathering ores originated through the same three-step process:
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Infiltration of meteoric waters through siliciclastic sedimentary rocks (sand, sandstones, siltstones);

Leaching of the iron ions (Fe$^{2+}$) released by the weathering of iron-rich minerals (glauconite, chlorite, pyrite, etc.);

Deposition of the iron ions by modification of the pH-Eh values of the water where it touches carbonate rocks (limestones), as iron oxides and hydroxides.

This category of ores is rather restricted to the geological contact between carbonate and siliciclastic sedimentary rocks. In Southern Belgium, such contacts are frequent throughout the stratigraphic column (see below and Fig. 2), and iron ores are also well developed. The subsequent subcategories are detailed below in ascending stratigraphic order.

2.2.1. Replacement ores along geological contact (‘amas couchés’)

This old denomination designates stratiform ore bodies chiefly composed of goethite and limonite, occurring along the contact of two different lithologies of sedimentary rocks (e.g. between limestones and shales or between limestones and shales; Fig. 3A). The contact between Eifelian shale and Givetian limestone bears iron mineralization in the Entre-Sambre-et-Meuse area (e.g. Erquelinnes, Merbes-le-Château, Berzée, Gourdinne, Gerpinne and near Fosse-la-Ville; Fig. 1) and in the Condroz area (around Esneux, Plainevaux, Dollembreux, Trooz, Sougné, Aywaille, Harzé, Ferrière and Weris). The ores form long (up to several hundreds of meters) and narrow (no more than 40 m) bodies stretched parallel to the strata, funnel-like in section and up to 80 m thick (Fig. 3B). This type of ore yields limonite and goethite concretions often carbonated or siliceous, that reach a content of 25-50% of iron. The deepest parts of the ores are usually made of siderite and/or pyrite. In the Entre-Sambre-et-Meuse area, the contact between the middle Frasnian limestone and upper Frasnian shale occasionally contains such ores (e.g. Gerpinnes and Saint-Gérard areas). They form ore bodies similar to the previous ones but smaller in dimension. The most important ‘amas couché’ in Southern Belgium occurs along the contact between the Famennian sandstone and the Tournaisian limestone (Fig. 1, Fig. 2). These ore bodies reach several hundred metres in length and reach up to 100 m in width and thickness. The mineralization is dominated by limonite and goethite, siderite and rare hematite and pyrite (Delmer, 1913). The Famennian-Tournaisian ore is omnipresent in the central part of the Entre-Sambre-et-Meuse area: Saint-Gérard, Biesmes, Scry, Mettet, Furnaux, Oret, Denée, Graux, Ermeton-sur-Biert, Sosoye, Biesmerée, Stave, Hanzinne, Yves-Gomezée, Jamiolle, Falaën, Serval, Onhaye, etc. The ores were mined discontinuously between the Antiquity and the 19th or 20th century in all these localities (Gillard, 1971) sometimes at a true industrial scale. In the Condroz area, the same geological contact is less mineralized and this for unclear reasons (hydrological context? enhanced erosion?). Some mines also operated in the Dinant, Hanoes, Gesves, Anthisnes and Comblain-au-Pont vicinities and in the Vesdre area. Several ores, mainly those of the Vesdre valley, are developed along the contact between Viséan limestone and overlying Namurian shale. Some rare ores are known to occur between the Jurassic marl and sandstone in the Belgian Lorraine (Souchez-Lemmens, 1968) but they were not mined in situ. The same phenomenon is observed if the two lithologies are in tectonic contact (e.g. along the Theux Fault in eastern Belgium). This first subcategory of ores was abundantly mined in Southern Belgium and is at the origin of the Gallic, Roman and medieval industries and lately of Walloon cast iron and steel industry (Denayer et al., 2011).

2.2.2. Cryptokarstic ores

The abannets-type or cryptokarstic ores formed in open karsts (developed in Devonian and Carboniferous limestone, Quinif, 1993) filled with Paleogene glauconitic sand. The glauconite being unstable under atmospheric conditions, it released its iron, subsequently leached through the sand by infiltrated meteoric waters and precipitated along the contact with the basement limestone. This category forms irregular bulks of limonite that are usually sandy (Van den Broeck et al., 1910) and contain up to 45% of iron (Delmer, 1913). The renowned ‘Abannets’ and ‘Fondry des Chiens’ sites in the Caléstienne area, are the most emblematic of this category of ores. They were mined since the late Antiquity and the sand initially filling the pockets was excavated. The large ore deposits of Fraire, Saint-Aubain and Morialmé in the Entre-Sambre-et-Meuse area, also belong to this cryptokarstic category (Fig. 3A), together
with the smaller ore deposits of the Fleurus and Oret vicinities (Rucloux, 1849; Dejaer, 1970).

2.2.3. Ferruginous gravels

Ferruginous gravels in the Belgian Lorraine area, consist of reworked fragments of goethitic and limonitic crusts. These crusts occur along lithological contacts within Jurassic rocks (see paragraph 2.2.1). The dismantling of the ferruginous crusts by superficial erosion and the sedimentation of the fragments in the valley, produced superficial deposits (so-called ‘fer d’alluvion’, alluvial iron ore) mined in the Tintigny and Châtillon areas (Delhez, 2004). This ore is mainly composed of dark goethite, reaching up to 25-50 % of iron (Delmer, 1913). Similar ores are observed in the Hainaut Province, where the Paleogene sands contain iron concretions reworked as gravel in some valleys (e.g. in the Quevy area, Delmer, 1913).

2.3. Lead-zinc sulfides-related iron ores

The lead-zinc ores are usually developed as sulfide veins composed of galena (lead sulfide) and sphalerite (zinc sulfide) associated with pyrite/marcasite (iron sulfide). The latter being unstable, it often degrades into iron oxides and hydroxides. The uppermost part of the vein, the so-called ‘chapeau de fer’ (iron cap or ‘gossan’) corresponds to the weathered, iron-enriched ore, capping the underlying lead-zinc ore (Fig. 3C). The lead-zinc veins are developed inside the Lower Carboniferous limestone and dolostone in the eastern part of Wallonia (Moresnet, Plombière, Kelmis), the Verviers and Theux areas (Dejonghe et al., 1993). In the Sambre and Meuse valleys, from Rhisnes (west of Namur) to Liège, several iron cap ores were mined for iron long before the discovery of zinc and lead. The Vedrin mine, where the iron cap was mined at least since the 14th century, while the galena was discovered only in 1612 (Évard, 1943). In other deposits (e.g. Haies Monets, Héron, Engis, etc.) the iron cap is less developed and the limonite was less exploited (Balcon, 1981; Dargent, 1949). The same veins are observed throughout the Devonian limestone and dolostone along the Calestienne area (from Chimay to Hotton) and in the Philippeville and Durbuy areas. Occasionally, barite, fluorite and copper minerals are associated with the lead-zinc veins (Dejonghe, 1985).

Remarkable hematite-rich veins occur near Porcheresse (Bièvre) in the Ardenne (Denayer et al., 2011). It is poorly known, but consists obviously of quartz-hematite veins set in Lower Devonian sandstone.

3. CONCLUSIONS

The hematitic oolitic ironstones (‘oligiste oolithique’) is probably the most renowned iron ore of Southern Belgium and it is the most studied both from an archaeological and historical point of view (see Dreesen et al., 2016: volume 1; Goemaere et al., 2016: volume 1). Nevertheless, it represents only a small part of the total volume of Southern Belgium iron ores, the first place being occupied by iron hydroxides (limonite and goethite) extracted from weathering deposits. Contrary to the hematitic oolitic ironstones, the weathering ores are very poorly documented, both from a geological point of view (except Delmer, 1913a) or from a historical and archeological point of view (e.g. Bonenfant et al., 1986). Iron hydroxides initially produced the source materials for prehistoric ochre industries. Occurrence of iron ochre associated with burial has been documented in Mesolithic and Neolithic sites throughout Belgium (e.g. Van den Broeck et al., 1910; Cauwe, 2001; Polet & Cauwe, 2002; Steevens et al., 2009; Miller et al., 2011 and references within). Nevertheless, the origin of the material (geographical and geological) has not been answered. The source of iron hydroxide ochre may be local considering the wide distribution of ore bodies in Southern Belgium, but long-distance dispersion cannot be excluded since such dispersal patterns are documented for hematitic oolitic pigment (Roebroeks et al., 2012). Analytic methods – such as Raman spectroscopy, Froment et al. (2008) – are interesting for the characterization of pigments but a database of natural ores is yet to be established for comparison purpose.

The iron hydroxides were the source of an important steel industry in Belgium reaching acmes during the Middle Ages and in the 19th century. The palaeometallurgy is an important human activity of our history but the source and process of ores are still poorly understood in Belgium. Again, analytic methods (e.g. Blakelock et al., 2009; Fluzin et al., 2000) applied to iron artifact or slags are promising but require a prerequisite knowledge of the iron ores.
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