DIAGENESIS AND ORE FORMING PROCESSES IN THE BÓDVASZILAS SANDSTONE OF THE RUDABÁNYA ORE DEPOSIT, NE HUNGARY

Sarolta BODOR, János FÖLDESSY, Ferenc KRISTÁLY & Norbert ZAJZON

Institute of Mineralogy and Geology, University of Miskolc, H-3515 Miskolc, Egyetemváros, Hungary, (bodors@gmail.com, foldfj@uni-miskolc.hu, askkf@uni-miskolc.hu, nzajzon@uni-miskolc.hu)

Abstract: We present the petrographical and geochemical study of the Bódvaszilas Sandstone Formation from the Rudabánya iron ore open pit. The work is linked to the current iron and base metal exploration in Rudabánya. According to our examinations the pore- and vein-filling carbonate cement of the Bódvaszilas Sandstone Formation is presumably early diagenetic. According to the concavo-convex grain contacts and the synthaxial quartz overgrowths on monocrystalline quartz grains, the formation underwent deep burial diagenesis. Base metal sulphides dominantly occur in the cover formations of Bódvaszilas Sandstone, appear only in the fracture zones of the Bódvaszilas Sandstone, as the permeability of the formation was low, therefore the metal bringing solutions were able to move mainly along the fractures. The chemical composition and crystal structure related properties of the "acidic sparry iron ore/cream-spar" from Bódvaszilas Sandstone is described for the first time. These carbonates are intermediate solid solutions between siderite and magnesite. The "cream-spar" was formed simultaneously with the metasomatic "sparry iron ore" by the partial or complete Fe replacement of the diagenetic dolomite cement and carbonate layers of the Bódvaszilas Sandstone Formation. Cassiterite has been detected first time in Rudabánya. According to its crystal habit is not detrital, but formed here, and it is present in a "cream-spar" section in the Bódvaszilas Sandstone.

Keywords: siliciclastic rocks, diagenesis, siderite-magnesite solid solution, iron ore, cassiterite, Rudabánya ore bearing complex, NE Hungary

1. INTRODUCTION

After centuries of base metal and silver mining, the open pit and underground iron ore mining of Rudabánya (NE Hungary) went on from 1872 until 1985. The ore deposit is located in the Lower-Middle Triassic siliciclastic and carbonatic succession. The majority of the ore is metasomatic siderite and its "limonitic" supergene oxidation.

In 2006 a new ore exploration project started, intending to define the Pb, Zn, Ag and Cu enrichments which occur within and near to the iron ore in Rudabánya. This paper describes the petrographical features of Lower Triassic Bódvaszilas Sandstone Formation and observes its role in the base metal mineralization, as part of the recent exploration project.

2. GEOLOGICAL SETTING

The Rudabánya ore deposit is located in the Aggtelek-Rudabánya Mts. and in a major Oligocene-

Miocene shear zone, the Darnó Zone.



Figure 1. Location of Rudabánya in the Alp-Carpathian region (after Fodor et al., 1999)

The Aggtelek-Rudabánya Mts. is part of the Silicicum stratigraphic superunit which consists of Upper Permian-Lower Triassic evaporites (Perkupa Evaporite Formation), Lower Triassic marine, siliciclastic and carbonate rocks (Bódvaszilas Sandstone, Szin Marl, Szinpetri Limestone Formations) and Middle-Upper Triassic carbonates (Gutenstein Formation, Steinalm Formation, Dunnatető Limestone, Bódvalenke Limestone, Hallstatt Limestone Formations) (Less et al., 2006).

In the Rudabánya ore deposit numerous drillholes penetrated the Bódvaszilas Sandstone Formation and reached the underlying Perkupa Evaporite Formation. The Bódvaszilas Sandstone is a tidal-subtidal succession and consists mainly of greenish-grey or red slate, siltstone and sandstone. The carbonate beds are more frequent in the upper part of the formation (Pantó, 1956, Kovács et al., 2004).

According to the latest studies (Földessy et al., 2010, Németh et al., 2013) a synsediment, stratiform, sedimentary-exhalative Pb-Zn-barite accumulation was formed in the Szin Marl. A later metasomatism has introduced the siderite ores hosted by the Lower-Middle Triassic Szinpetri Limestone, the Gutenstein Dolomite and the Bódvaszilas Sandstone.

According to Pantó (1956) the mined siderite ore ("sparry iron ore") is mainly derived from Lower Anisian dolomite. Although it is called siderite, its main mineral constituents (besides siderite) are ankerite, calcite, barite, quartz, pyrite, chalcopyrite, hematite, galenite and tetrahedrite. The fractured dolomite blocks, which placed in the marl, were metasomatised along the fractures and the differences in the chemical composition derive from the variable intensity of the metasomatism - cca. the 1/3dolomite of the and limestone were metasomatised. The original composition was pure dolomite and calcite with dispersed mica, clay minerals and minor quartz. In the amoeboid-like

texture euhedral siderite is almost absent. Late barite (as the accompanying mineral of sparry iron ore) is in cca. 8 wt% present in the border of the blocks and in veins, veinlets.

These carbonatic formations, which are strongly tectonized, can be found only in separate blocks. In the next ore forming phase (Paleogene) mainly pyrite-bearing massive sulphide filled some veins, and later Pb-, Zn-, Ag- and Cu-enrichments with barite were generated. The faults of the Darnó Zone promoted the latest sulphidic ore formation (As-, Sb-, Hg- and Ag-enrichments).

The cover of the ore-bearing formation is of Miocene sediments, in which the ore-bearing blocks are observed (Fig. 2) (Földessy et al., 2010).

3. ANALYTICAL METHODS

85 samples were investigated during this study, which were selected from 9 drillcores, drilled between 2008 and 2010 in the Rudabánya open pit. Detailed macroscopic and microscopic petrographic investigations were done before other analyses.

X-ray powder diffraction (XRD) of bulk rock samples was performed on a Bruker D8 Advance diffractometer (CuK α source, 40 kV, 40 mA, secondary graphite crystal monocromator, scintillation detector) at the Institute of Mineralogy and Geology, University of Miskolc, Hungary. Electron microprobe analyses by the means backscattered electron imaging (BSE) and energy dispersive spectrometry (EDS) was done on a JEOL JXA-8600 Superprobe (15 kV, 20 mA, carbon coating, 60 s acquisition time) at the same institute.



Figure 2. Geological section of the Rudabánya ore deposit (after Németh et al., 2013, modified)

The ${}^{13}C_{carb}$ and ${}^{18}O_{carb}$ stable isotopic analyses were carried out on 10 samples in the Institute for Geological and Geochemical Research, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences (Budapest, Hungary).

4. RESULTS

4.1. Petrology of Bódvaszilas Sandstone Formation in the Rudabánya open pit

The studied Bódvaszilas Sandstone samples are predominantly bluish green, greyish green, grain-supported, well sorted, oligomict, very fine or rarely fine grained quartzarenites, and matrixsupported, well sorted, oligomict siltstones and claystones, slates. The grains are roundedsubrounded. The material sometimes locally red colored in the drillcores.

The Bódvaszilas Sandstone is moderately brecciated. Although the samples are crumbly, the sedimentological characteristics are well-defined, such as parallel lamination, less frequently crosslamination, lenticular bedding, with claystone and siltstone intraclasts.

The most abundant constituent in the formation is quartz. This is only monocrystalline in anhedral grains, subhedral grains are subordinate. Resorbed grains also occur rarely.

Besides the monocrystalline quartz detrital muscovite is present, nevertheless in more or less amount. Some altered biotite were observed as well.

Feldspars are very rare in the Bódvaszilas Formation in Rudabánya, only plagioclase (albite) is present in minor amount, K-feldspar was not observed. Feldspars are in higher quantity in the Bódvaszilas Sandstone in the non-mineralised areas (Aggtelek Mts.), which are mostly plagioclase (albite) and more or less altered K-feldspar (microcline).

Rarely a few altered amfibole and piroxene grains were found. Accessory minerals are rounded zircone, subhedral or well rounded apatite, rutile, monazite, less frequently xenotime, ilmenite and rounded tourmaline (schorl). The size of accessory minerals is usually small (10–30 μ m).

There are no lithic fragments in the formation. Among opaque minerals only disseminated or framboidal pyrite is present which occurs along fractures and the laminae. Minerals with enrichment of Pb, Zn, Cu, Ag, Sb and As were observed only in the fracture zones.

There is a great amount of pore-filling, layered and fracture-filling carbonate cement in the

formation which composition is Mg- and Mnbearing siderite, Fe-bearing magnesite and ankerite.

The smaller proportion of the cement of Bódvaszilas Sandstone is quartz, chlorite, illite and rarely kaolinite and hematite.

The formation is strongly compacted and the line, concavo-convex (Fig. 3) and sutured grain contacts and the quartz overgrowths on the surface of monocrystalline quartz grains (Fig. 4) are typical. Frequently small carbonate inclusions are present in the authigene quartz phase.



Figure 3. Concavo-convex grain contacts between monocrystalline quartz grains in Bódvaszilas Sandstone



Figure 4. Synthaxial quartz overgrowth (white arrow) on monocrystalline quartz grain

Coarse grained sparry carbonate and accompanying anhedral quartz occurs in fractures, and in the carbonate there are euhedral quartz crystals of several millimeters. Detrital, disseminated or vein-filling barite occurs rarely.

4.2. "Cream-spar" in the Bódvaszilas Sandstone

The "acidic sparry iron ore" or "cream-spar" in the Bódvaszilas Sandstone in Rudabánya was also

known previously. The mining of this ore-type was not economic because of its relatively high SiO_2 content. Nevertheless the estimated resources of "cream-spar" were 20 million tons in 1987 after the closure of the mine (Balla et al., 1987). The creamspar's composition was usually published as siderite. It occurs mainly along the tectonic lines with the most intense mineralization (Hernyák, 1967).

In the boreholes located in Rudabánya, "creamspar" is present commonly in 1–2 cm thick veins or in disseminated zones of several meters of thickness. It occurs as a pale whitish yellow, sparry carbonate with scattered hematite flakes. In the fine-grained sandstone fraction it occurs as pore-filling cement also, therefore it closely follows the bedding and where the formation originally was more compact, the carbonate content decreases.

According to our examinations the "creamspar" showed no luminescence due to its high Fe content (Boggs & Krinsley, 2006).

By XRD we observed peaks of the R-3c spacegroup, with d(Å) values as intermediary between siderite and magnesite structure. The $d_{(104)}$ varies between 2.76 Å and 2.78 Å, indicating a larger range of substitutions. Intensities were affected by strong preferred-orientation, but applying the March-Dollase correction, we obtained relative intensities characteristic for the siderite lattice.

By BSE and EDS we could observe the relations between composition and texture of the "cream-spar". According to our observations it often on shows chemical zonation patterns, the rhombohedron faces. By EDS the "cream-spar" is always inhomogeneous in elemental composition, but it is always Fe and Mg carbonate with low Mn content (never pure siderite or pure magnesite, it is always with excess substitution of Mg and Fe). The Fe and Mg ratio changes in steps of several micrometers, it is independent of textural characteristics. In the analysed samples the Fe content varies between 0.11 and 0.84, and the Mg content between 0.25 and 0.65 atoms in the RCO₃ theoretical formula (R: Fe, Mg, Mn, Ca).

In a thick "cream-spar" section of a drillcore there are some brownish red patches or fragments frequently with disseminated pyrite. They consist of euhedral-subhedral quartz and hematite flakes (frequent as inclusions in quartz, Fig. 5). Small (~1–5 μ m), disseminated cassiterite occurs also in a fragment and very rarely in the "cream-spar" in thin (0.5 mm wide) quartz veins of a lineation. Locally small (~1–10 μ m) xenotimes are also present besides cassiterite. Due to the small amount of the patches it is not sure wheter these patches are part of the quartz veins or the fragments of Bódvaszilas Sandstone.



Figure 5. Euhedral quartz (Q) and hematite (Hem, white arrow) beside creamspar with variable Fe and Mg content (lighter and darker zones, BSE image)

Although cassiterite is a common accessory mineral in siliciclastic rocks, in our case according to its habit it is not of detrital origin. It is located inside or on the border of quartz or pyrite (Fig. 6) grains. In some cases pyrite was overgrown on cassiterite. In the Bódvaszilas Sandstone samples xenotime occurs as detrital grains, and its fragments also present in the "cream-spar" section too, in the carbonate or in quartz grains.





Figure 6. Disseminated cassiterite (white phase shown by white arrows) inside, or on the margin of pyrite (Py) in a fragment enclosed in "cream-spar" (Cp)

In a 1.0 m long "cream-spar" section of a drillcore 1.28 ppm and 0.77 ppm Au was also detected. In the separated cassiterite and xenotime bearing "cream-spar" sample 0.74 ppm Au has been detected. Besides the fire-assay we could not observe the gold as mineral phase by any microscopy technique.

The aim of our ${}^{13}C_{carb}$ and ${}^{18}O_{carb}$ stable isotopic examination was to determine the source of the "cream-spar" and to compare it with the sparry iron ore.



Figure 7. The analysed "cream-spar" and sparry iron ore samples on the $\delta^{18}O-\delta^{13}C$ diagram, some comparative samples of Hofstra et al., (1999) are also plotted

Sample		δ ¹³ C (‰) _{PDB}	δ ¹⁸ Ο (‰) _{PDB}
PV/I/1	sparry iron ore	-4.69	-13.88
PV/I/2	sparry iron ore	-3.67	-11.50
PV/II/1	sparry iron ore	-3.45	-11.77
T12A/4	"cream-spar"	-5.60	-13.55
T12A/5	"cream-spar"	-4.77	-13.44
T12A/7	"cream-spar"	-3.91	-12.21
T14A/11	"cream-spar"	-5.18	-14.39
T14A/8	"cream-spar"	-6.18	-14.66
T15/8	"cream-spar"	-5.23	-12.96
T15/KP2	"cream-spar"	-5.09	-13.96

Table 1. δ^{18} O and δ^{13} C data of the analysed sparry iron ore and "cream-spar" samples

On the $\delta^{13}C/\delta^{18}O$ diagram (Fig. 7) the "creamspar" and sparry iron ore samples appear as grouped on the MVT-type material, but constitute two distinct fields. The ¹³C_{carb} and ¹⁸O_{carb} values show positive correlation with each other. Our ¹³C_{carb} and ¹⁸O_{carb} values (Table 1) are compared to the brecciated dolomite host rock and siderite from Rudabánya of Hofstra et al. (1999). Our "cream-spar" and sparry iron ore and the siderite samples of Hofstra et al., (1999) resemble to each other and they are located in the field of the MVT-type hydrothermal dolomite. The two dolomite samples of Hofstra et al., (1999) differ from the other samples.

5. DISCUSSION AND CONCLUSIONS

In the Bódvaszilas Sandstone in Rudabánya

(and in Aggtelek Mts. as well) we have observed textural characteristics of diagenetic origin. The formation is rather compacted which is particulalry visible in the fine-grained sandstone. The grain contacts are mainly straight and concavo-convex. Where the pore- and vein-filling carbonate cement is present, the grains form clusters. On the surface of dominately monocrystalline quartz grains synthaxial quartz overgrowths are frequent. Here, only a thin borderline shows that these are two different quartz generations. In the carbonate we could observe larger (0.1–1 mm), euhedral-subhedral quartz grains. These are detrital monocrystalline quartz grains with quartz overgrowths. In fractures euhedral quartz crystals with few inclusions occur rarely, indicating the existence of voids/pores during diagenesis.

Mechanical compaction is considered to

happen during early and intermediate burial diagenesis (0-2.5 km depth), and involves the rearrangement of the grains (Berner, 1980).Chemical compaction begins during deep burial and leads to the solubilization of grains at their contacts due to pressure and the reprecipitation of dissolved material on the grain surfaces. Chemical compaction gains importance at temperatures above 80-90°C, and the formation of quartz overgrowths and illite usually occurs between 80 and 130°C (Fischer et al., 1999, Worden & Morad, 2000, Peltonen et al., 2009). Morad et al. (2000) used two burial ranges according to the burial depth and the temperature. Theses are eodiagenesis (0-2 km burial depth, <70°C) and mesodiagenesis (> 2 km burial depth, >70°C), where the latter is the range of chemical compaction.

With increasing intensity of diagenesis the number of compound grains rises. Compound grains consisting of 4–5 pieces constitute a cluster (Ahmad & Bhat, 2006). Compaction leads to the increase of contact strength, with more straight and concavo-convex grain contacts than point contacts or isolated grains (Bernet et al., 2007).

Considering the presence of the straight and concavo-convex grain contacts and the quartz overgrowths in the Bódvaszilas Sandstone, the formation underwent deep burial diagenesis. The euhedral-subhedral quartz grains in the massive carbonate evidenced that in the upper, carbonatic part of the formation monocrystalline quartz grains existed which served as nucleation sites for authigenic quartz precipitation.

The space between the grain clusters filled by the large amount of carbonate cement indicates that carbonate cementation happened before the compaction in the sandstone with high primary porosity. According to the ¹³C_{carb} and ¹⁸O_{carb} stable isotope values Hofstra et al., (1999) pointed out that the dolomite (Gutenstein Formation) in Rudabánya is of diagenetic origin and only slightly altered by hydrothermal fluids. Our opinion is that the Bódvaszilas Sandstone's pore- and vein-filling carbonate is an early diagenetic cement which presumably later dolomitized during the diagenesis as the Gutenstein Formation.

We were able to define the composition and the crystal structure of the "cream-spar" in Bódvaszilas Sandstone. The "cream-spar" is a solid solution between siderite and magnesite in which the siderite/magnesite (Fe/Mg) ratio shows large scattering and variability on micron scale. The earlier opinion about the origin of "cream-spar" was that it is a metasomatic ore such as the "sparry iron ore", according to our view it might be sedimentary cement as well.

The ¹³C_{carb} and ¹⁸O_{carb} values of siderite, ankerite and Fe-bearing dolomite cement of deep burial diagenetic sandstones examined by Mansurbeg et al., (2008) resemble our stable isotope values indicating the replacement of shallow diagenetic calcite and dolomite cement by porewaters. In line with this, our conclusion is that the "acidic sparry iron ore" was generated during the metasomatism of the shallow diagenetic pore- and vein-filling cement and carbonate beds in the upper parts of the formation. The metasomatism was the same which created the mineable "sparry iron ore", and their protolith may have had the same composition. The frequent zonation which occurs occasionally in rhombohedral shape indicates that the metasomatism proceeded parallel to the rhombohedron crystal faces of the dolomite - it corresponds with the similar observations in "sparry iron ore" (Pantó, 1956, Szakáll, 2001). The experiments of Kiss (1981) pointed out that the cation exchange along the rhombohedron faces is intensified. The Fe-bearing dolomite which occurs occasionally besides sideritemagnesite in "cream-spar" indicates also that presumably a dolomite suffered metasomatism and and Fe replacement occured in more or less proportion.

Taken into consideration the former genetic models of the Rudabánya ore deposit the presence of non-detrital cassiterite is peculiarly new recognition. However, due to the small number of studied cassiterite bearing samples, additional examinations will needed to clarify its formation.

The base metal sulphides, which occur in the overlying formations of the Bódvaszilas Sandstone, were enriched only in the fracture zones as the permeability of the formation was low, therefore the solutions were able to move only along the fractures. This statement corresponds with former observations, that in massive sandstones the fluids move in fractures and the permeability of the sandstone blocks is irrelevant (Martos et al., 1975).

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