

STABLE ISOTOPIC STUDY OF THE RUDABÁNYA IRON MINE. A CARBONATE-HOSTED SIDERITE, BARITE, BASE-METAL SULFIDE REPLACEMENT DEPOSIT

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ABSTRACT

The Rudabánya iron mine is one of the oldest mines in Hungary. The deposit is hosted in Lower to Middle Triassic dolomites and siliciclastics. The stratabound replacement deposit is localized by faults produced during Middle Triassic rifting and is deformed by Late Cretaceous folds. The present day distribution of the ore is controlled by a large asymmetric and locally overturned dome. The primary massive replacement ores formed in 3 stages (1) hematite (\pm siderite) and (2) siderite, cut by (3) stockworks and veins of barite, galena, and chalcopyrite with anomalous Ag and Au. Mineral zoning in the ores from the surface to a depth of about 300 m consists of a zone of secondary limonite (40–50 m) that locally contains native copper and silver, followed by a zone of primary siderite (150 m) with chalcopyrite and galena and then hematite (>100 m) with lesser sulfides. The presence of marcasite in the ores is indicative of temperatures less than 240 °C and pH < 5. Results of stable isotope studies suggest (1) the host dolomite is diagenetic in origin, (2) the hydrothermal siderite has isotopic compositions similar to dolomite in MVT Zn-Pb deposits, (3) the SO₄ in barite was derived from Lower Paleozoic black shales underlying the deposit, (4) the sulfur in sulfides was derived from thermochemical reduction of Upper Permian sedimentary sulfate, and (5) sphalerite-galena geothermometry yields a temperature of 115 °C. The ore deposit formed where migrating, acidic, basinal brines containing Fe, base metals, and sulfate moved up faults produced by Middle Triassic rifting and encountered reactive carbonate rocks and indigenous ground waters containing Ba and H₂S.

1. INTRODUCTION

One of the oldest mines in Hungary, the Rudabánya iron mine is located in the Northeastern Range near the town Miskolc (Fig. 1). The deposit is hosted in Lower to Middle Triassic dolomites and siliciclastics where they are cut by a major NE-striking fault system called the Darnó Fault. Mining began about 2000 years ago for native copper. In the Middle Ages, Rudabánya was a free mining town and became one of the centers of copper and silver mining. Although mining declined during the 16th and 17th centuries, there was a resurgence with commencement of the first open pit iron mine in 1878. Intensive exploitation of the limonitic iron ores resulted in the opening of seven pits (Polyánka, Bruinmann, Andrásy I, II and III, Vilmos, Ruda-hegy and Deák pit) that together form a 5 km long by 1.5 km wide continuous mining belt. About 1,000 limonitic ore bodies were mined to depths of 50 to 60 m with maximum sizes of 500 × 40 × 10 m (PANTÓ 1956). Over 21 Mt of limonite ore with an iron content of 33–35% was produced. Surface operations were abandoned in 1953 but underground workings continued to extract siderite ore with 24% iron. About 50,000 t/year of copper ore was processed as a by-product. Iron mining ceased in 1985 after producing 11 Mt of siderite ore. Present day proven ore reserves according to BARTÓK and NAGY (1992) are as follows: 37.2 Mt of siderite ore (24.29% iron), 1.54 Mt of chalcopyrite ore (0.56% Cu and 1.2 ppm Ag) and 0.56 Mt of galena ore (1.42% Pb). At the current prices the average concentrations of the ores are insufficient to be mined economically. Gold concentrations are also low and generally <50 ppb (KÖRÖS et al. 1999).

Rudabánya shares many features with carbonate-hosted siderite replacement deposits that form from basinal brines and are akin to MVT Pb-Zn deposits [e.g. Bilbao, Spain; Quenza, Algeria] (BOUZENOUNE and LECOLLE 1997).

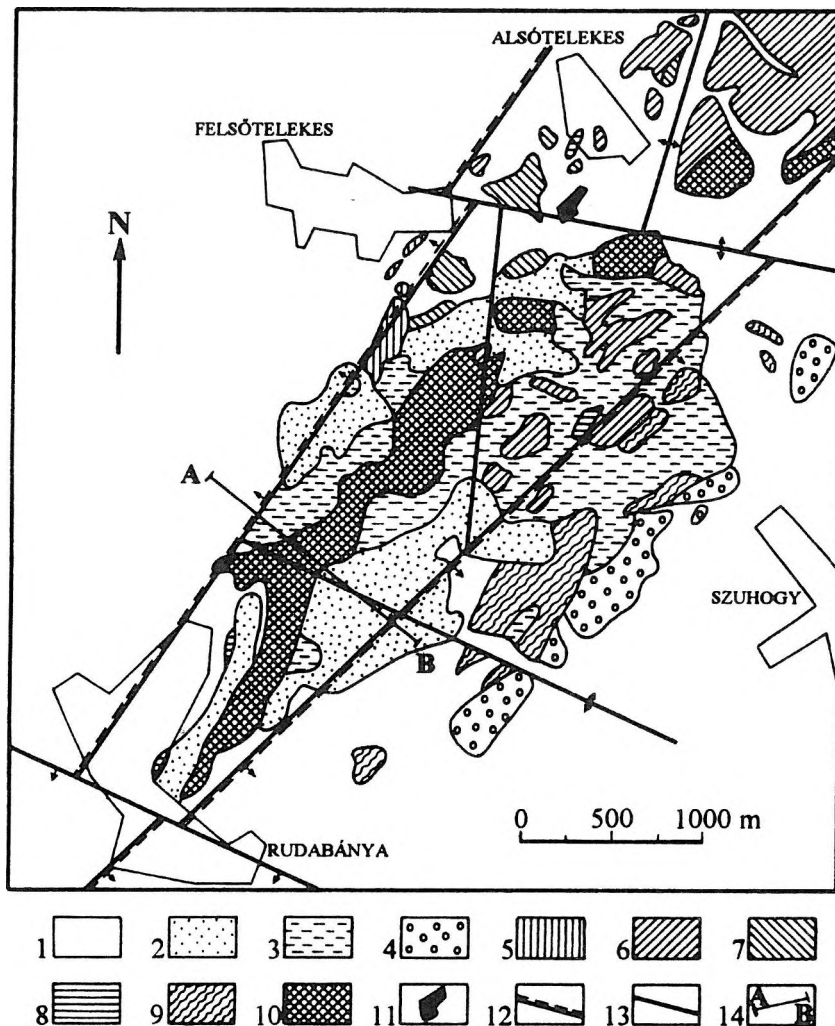


Fig. 1: Geologic map of the Rudabánya iron mine (after HERNYÁK 1977)

1. Quaternary sediments, 2. Mine waste, 3. Late Miocene clay, sand and gravel, 4. Early Miocene Szuhogy Conglomerate, 5. Jurassic shales, cherty limestone and radiolarite, 6. Middle Triassic Gutenstein Dolomite, 7. Lower Triassic Szin Marl and Szipetri Limestone, 8. Lower Triassic Bódvaszilás Sandstone, 9. Paleozoic Tapolcsány Formation, 10. Limonite and siderite ore, 11. Hematite ore, 12. Reverse fault, 13. Normal fault, 14. Cross section.

Rudabánya also shares some features with polymetallic-siderite-quartz vein deposits hosted in clastic metasedimentary rocks that form from deep formation or metamorphic waters that ascend along major faults [e.g. Příbram, Czech Republic; Rudnany, Nizna Slana, and Dobsina in the Gemer Mountains, Slovakia; Bakalsk in the Ural Mountains, Russia; Kremikovci and Ciprovci in Sredna Gora, Bulgaria; Vares and Liubia, Bosnia; and Eiesenerz, Austria] (NAGY 1982; BEAUDOIN and SANGSTER 1992).

Previous investigations at Rudabánya by PANTÓ (1956), CSALAGOVITS (1973a,b) and NAGY (1982) resulted in the following conclusions regarding its origin. According to PANTÓ, the mineralization formed at shallow depths of up to 200 m, at temperatures of 100–150 °C, without any direct influence from magmatic activity. A Laramian age of mineralization was postulated by him based on evidence that the deposit was deformed during Late Cretaceous regional compression. Based on a systematic geochemical study of the mineralization and its footwall and hanging wall formations, CSALAGOVITS (1973a, b) concluded that the deposit formed from the mixing of two contrasting fluids. He thought that there were two major paleoaquifers in the region and that the replacement mineralization resulted from the mixing of a low temperature, reduced, descending water (rich in Ca, Mg, H₂S and HCO₃ with Fe, Mg, Ba) with a high temperature ascending thermal water (rich in Mg, Fe, Cl and SO₄ with Cu, Pb, Zn, Ag, As, Sb, Hg, Tl, Bi, Cd, In, and Ge). The source of the metals was considered by him to be the evaporites (Perkupa Anhydrite) and Paleozoic black shales below it. Moderate to deep burial subsidence, pre-ore tensional tectonics, and porosity controlled the localization of ore, which was sealed and trapped mainly by the Szin Marl. The mineralization predates compressional deformation. NAGY (1982) utilized fluid inclusion decrepitation temperatures from chalcopyrite (430 °C), galena (360 °C) and barite (350 °C and 260 °C) to estimate the temperature of ore formation and concluded that, in the absence of magmatic activity, these data require a long period of deep burial and/or elevated heat flow during ore formation.

During a field trip to Rudabánya in 1996, we collected nineteen representative samples from four sites in the Andrásy I. open pit. Samples were obtained from the dolomite host rock, siderite ores containing base metal sulfides and barite, and limonite ores containing supergene gypsum. Another sample was collected from the Alsótelekes gypsum-anhydrite open pit, which is in rocks representative of the foot wall of the Rudabánya Iron Mine. Polished pucks and thin sections were made of samples from the Andrásy I. pit and examined to document the paragenetic sequence. Minerals representative of the major stages of mineralization were separated and analyzed using standard stable isotopic methods. The goal of this study was to utilize new stable isotopic data along with previous geologic and geochemical information to improve understanding of the origin of the deposit.

2. GEOLOGIC SETTING AND TECTONIC EVOLUTION

The geologic setting and tectonic evolution of the area are described below based on information in GRILL et al. (1984), LESS and SZENTPÉTERY (1987), GRILL (1988), RÉTI (1988), KOVÁCS et al. (1989), HIPS (1996) and LESS (1998). The Late Permian to Middle Triassic pre-rift sedimentary sequences are representative of a continental to shallow ramp depositional system. Early evaporites (Perkupa Anhydrite) were followed by peritidal siliciclastics (Bodvaszilás Sandstone*), subtidal carbonates (Szin Marl*, Szinpetri Limestone*) and capped by dolomites of restricted lagoon (Gutenstein Formation*) or limestones of open lagoon (Steinalm Formation) environments. Rifting, accompanied by synchronous volcanism, was completed by Ladinian time and resulted in disintegration of the ramp system and opening of an oceanic basin between two continental units to the south and north. This oceanic basin was consumed partly during Middle Jurassic north-oriented subduction and closed during the Late Jurassic to Early Cretaceous collision of the two continental units. Coeval volcanism and emplacement of Gemer Granites far to the north can be related to the subduction and collision. A new depositional cycle started in the Middle Oligocene, after a long period of subaerial exposure, characterized by progressive nappe-formation to the south during the Late Cretaceous. Middle Oligocene to Early Miocene clastics were deposited in a narrow pull-apart basin oriented NNE-SSW controlled by sinistral movement along the Darnó Fault. Since then, the 25 km long iron ore belt (including the Rudabánya, Martonyi and Esztramos deposits) along the border east of the Rudabánya Mts. has been in its current position. Late Miocene to Quaternary alluvial, lake and eolian clastic sedimentation was the last depositional event, and was followed by subsequent uplift and erosion, yielding the present day picture.

3. DEPOSIT DESCRIPTION

The stratabound siderite replacement mineralization is hosted in the Bodvaszilás Sandstone, Szin Marl, Szinpetri Limestone and Gutenstein Dolomite (Fig. 2). The overlying Steinalm Limestone is unmineralized. The total thickness of these formations ranges from 800 to 1000 m. Siderite mineralization is known to a depth of 300 m, with mined ore to a depth of about 130 m (underground level +180 m, HERNYÁK 1977). Distribution of the ore is controlled by a large asymmetric and overturned dome that strikes NNE (PÁLFY 1924, BALOGH and PANTÓ 1952). This dome exhibits minor "roof structures" and is dissected by internal folds that verge to both the NW and SE (PANTÓ 1956).

Present day vertical zoning is outlined below after HERNYÁK (1977). The total thickness of mineralized rock is about 300 m and is divided from top to bottom in the following order:

Zone of secondary limonite: 40-50 m with variable basemetals,

Zone of primary siderite: 100 m of siderite that is rich in base metals (Cu, Pb) underlain by 50 m of silica-rich siderite that is poor in base metals (Cu),

Zone of primary hematite: >100 m of hematite that is poor in base metals (Cu).

Although siderite and Cu mineralization only extend to depths of about 300 to 400m, hematite mineralization has been encountered at the deepest levels explored by drilling (800m).

Three stages of mineralization were recognized by PANTÓ (1956), consisting of primary massive replacement ores with (1) hematite (\pm siderite) and (2) siderite, cut by (3) base metal stockworks and veins of barite, galena and chalcopyrite. The simple paragenesis of the primary iron ores, with ankerite, calcite and pyrite, was overprinted by a base metal sulfide-barite paragenesis accompanied by siderite, pyrite, pyrrhotite, bornite, chalcocite, tetrahedrite, stibnite, schwazite, pyrrargyrite, bourmonite, jamesonite and sphalerite with quartz, fluorite, sericite and other clay minerals (CSALAGOVITS 1973a, b and NAGY 1982). The secondary iron ore, consisting of massive

* Host rocks of the Rudabánya Iron Ore.

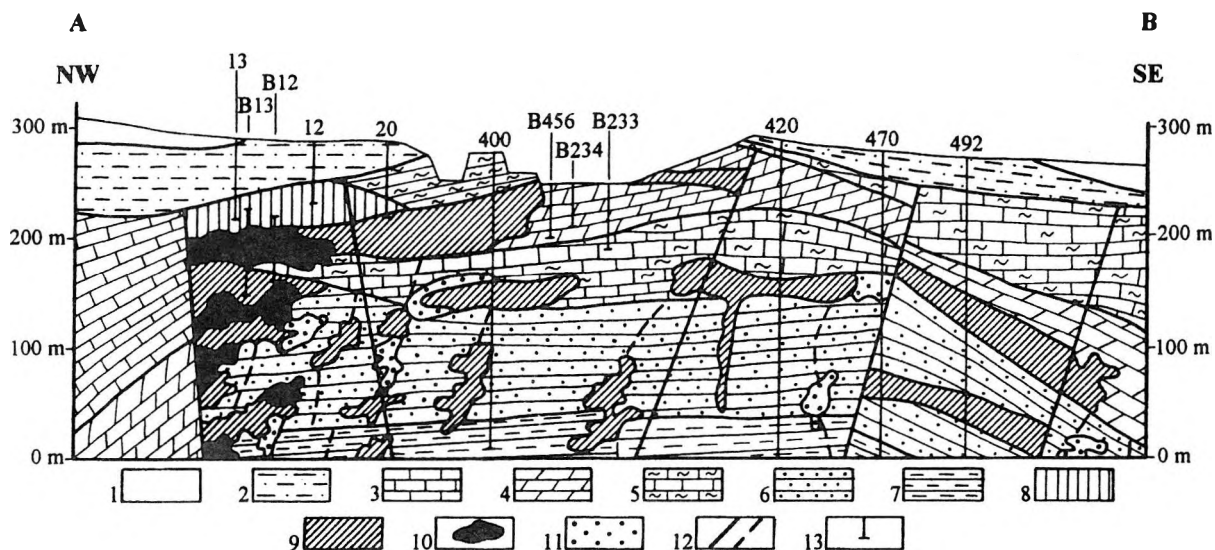


Fig. 2: Cross section of the Rudabánya iron mine (after HERNYÁK)

1. Mine waste, 2. Late Miocene clay, sand and gravel, 3. Jurassic shale, cherty limestone and radiolarite, 4. Middle Triassic Gutenstein Dolomite, 5. Lower Triassic Szin Marl and Szinpetri Limestone, 6. Lower Triassic Bódvaszilás Sandstone, 7. Black shales, 8. Limonite ore, 9. Siderite ore, 10. Polymetallic ore, 11. Barite (±sulfides), 12. Faults, 13. Drill holes.

limonite, goethite, hematite, sphaerosiderite, marcasite, and relict primary minerals, is rich in supergene Mn-(pirolusite, psilomelane, waad), Cu-(covellite, tenorite, cuprite, azurite, malachite, native copper), Ag-(native silver), Pb-(cerussite, anglesite), and Hg-minerals (cinabar, mercury), and also contains gypsum, epsomite, and melanterite. Traces of native gold were reported by PANTÓ (1956) and NAGY (1982).

Many of the primary hypogene minerals mentioned above were also observed in our samples. However, we observed textural features in the pyrite which indicate that it formed from a metastable marcasite precursor (MUROWCHICK 1992). We also think that the "secondary marcasite" described above may actually be primary. This is important because marcasite only forms at temperatures <240 °C from fluids with a pH <5 (MUROWCHICK 1992). The presence of marcasite in the ores suggests that the temperature of mineralization was much lower than that inferred from decrepitation temperatures of fluid inclusions (NAGY, 1982). The decrepitation temperatures are also unreliable because similar decrepitation temperatures are obtained from MVT Pb-Zn deposits in the United States that formed at temperatures <150 °C (D. LEACH and A. HOFSTRA unpublished data). The paragenetic sequence portrayed on Fig. 3 is based on crustification sequences and cross cutting relationships observed in polished pucks, thin sections, and field observations. It is similar to previous ones with precipitation of siderite followed by basemetal sulfides and barite. Episodes of brecciation or faulting are also indicated. As above, weathering and oxidation of the hypogene mineral assemblage resulted in the formation of limonite, gypsum, and a variety of supergene Cu, Pb, and Ag minerals.

Geochemical studies (CSALAGOVITS 1973a, b) have shown that the replacement mineralization is characterized by the introduction of Fe, Mg, Mn, Ba, Cu, Pb, Zn, Ag, As, Sb, Hg, Bi, Cd, In, Ge, and Tl. Previous Au and Ag analyses, conducted between 1971 and 1975, on 160 underground, surface and drillhole samples yield sporadic low level Au anomalies (traces to 0.2 ppm with a maximum of 1 ppm) and widespread Ag anomalies ranging from a few ppm to 20 ppm with a maximum of 451 ppm (RUDABÁNYA). New gold analyses of 50 grab samples from the mine ranged from <2 to 630 ppb, but only 5 samples contained more than 50 ppb Au (KORPÁS et al. 1999). Silver content ranges between 0.2 and 425 ppm with 11 samples containing more than 50 ppm Ag (KORPÁS et al. 1999).

4. STABLE ISOTOPES

The stable isotopic composition of brecciated host rock dolomite, evaporite gypsum, hydrothermal siderite, sulfides, and barite, and supergene gypsum are shown on Figs. 4 and 5. The brecciated dolomite host rock has $\delta^{13}\text{C}$ values (-0.8 to -1.7‰) typical of marine limestone and $\delta^{18}\text{O}$ values (18.2 to 18.8‰) that are at the low end for marine limestone. The isotopic data suggest that the dolomite is diagenetic in origin and was not substantially recrystallized by the hydrothermal fluids, although there may have been minor ^{18}O depletions. The isotopic com-

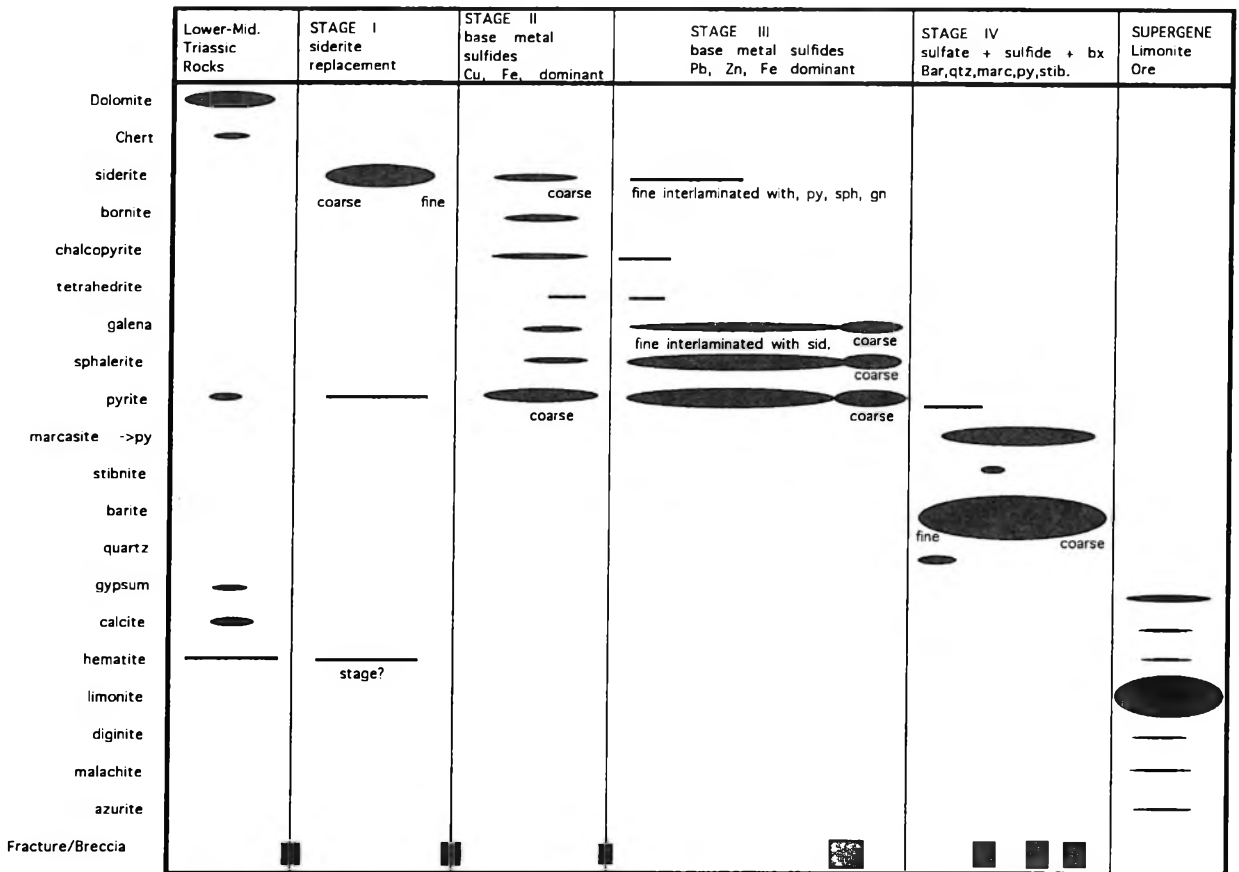
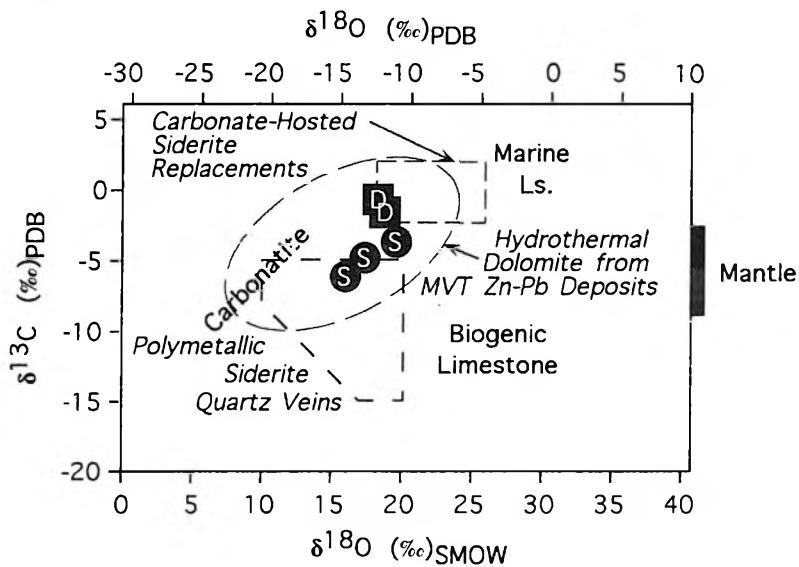


Fig. 3: Paragenetic sequence constructed from field observations and petrographic examination of polished pucks and thin sections
Fracturing and brecciation are also indicated

Fig. 4: Carbon and oxygen isotope composition of host rock dolomite (D) and hydrothermal siderite (S)
Results are shown relative to common ranges for marine limestone, biogenic limestone, carbonatite, and mantle carbon. The ellipse covers the common range of values for hydrothermal dolomite from MVT Zn-Pb deposits. The isotopic compositions of siderite in carbonate-hosted siderite replacement deposits (rectangle, BOUZENOUNE and LECOLLE 1997) and polymetallic siderite quartz veins (polygon, BEAUDOIN and SANGSTER 1992) are also shown



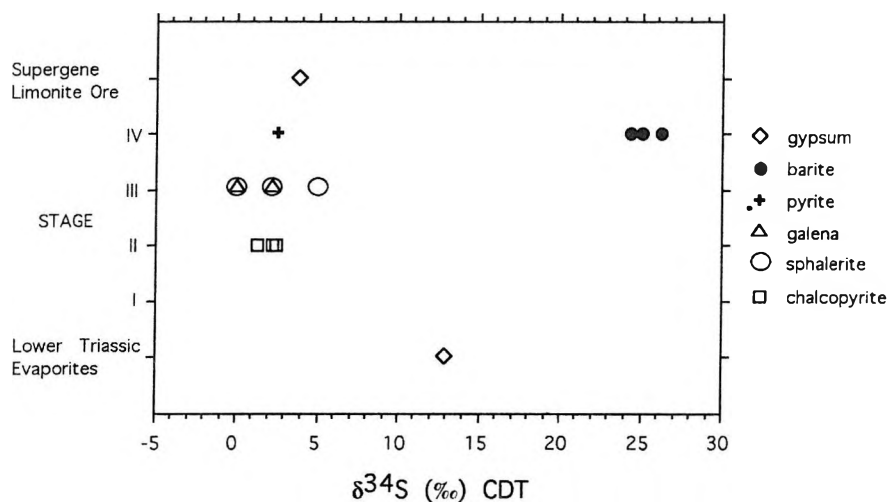


Fig. 5: Sulfur isotopic composition of sulfides and sulfates through the paragenetic sequence.

position of the siderite ($\delta^{13}\text{C} = -3.7$ to -6.1‰ , $\delta^{18}\text{O} = 16.0$ to 19.6‰) is lower than host rock dolomite, lower than carbonate-hosted siderite replacements, within the range of hydrothermal dolomite from MVT Zn-Pb deposits, and one value is within the range of polymetallic siderite quartz veins (Fig. 4). Although the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ data do not permit a unique interpretation, the geology of the deposit is more similar to carbonate-hosted siderite replacements and MVT Zn-Pb deposits.

To determine whether the sulfate in ore fluids was derived from the Late Permian Perkupa Anhydrite, the sulfur isotopic composition of hydrothermal barite from Rudabánya was compared with evaporite gypsum from Alsótelekes. The $\delta^{34}\text{S}$ values of barite (24.3 to 26.2‰) and gypsum (13.0‰) are distinctly different (Figs. 3 and 5). The $\delta^{34}\text{S}$ composition of the gypsum is typical of Upper Permian and Lower Triassic marine evaporites while the barite is similar to marine sulfate in Late Devonian-Early Mississippian and Silurian rocks (CLAYPOOLE et al., 1980). The sulfate in the fluids that deposited barite may therefore have been derived from unexposed Lower Paleozoic rocks at deeper levels in the system. Rocks correlative with the Paleozoic black shales exposed in the Uppony and Szendrő Mts. are inferred to underlie the deposit.

Chalcopyrite, sphalerite, galena, and pyrite have $\delta^{34}\text{S}$ values that range between 0.3 and 5.1‰ (Figs. 3 and 5). The equilibrium sulfur isotopic fractionations between sphalerite and galena were used to estimate the temperature of ore formation (OHMOTO and RYE 1979). Sphalerite and galena from sample RB8-5 ($\Delta_{\text{sph-gn}} = 4.8\text{‰}$) yield a temperature of 115 °C that is well below the $<240\text{ °C}$ temperature constraint provided by the presence of marcasite in the ores. This temperature estimate is also similar to those from other siderite replacement deposits and MVT Pb-Zn deposits world wide. The temperature of 115 °C was used to calculate the isotopic composition of H_2S in ore fluids. The $\delta^{34}\text{S}$ values calculated for H_2S range from -0.3 to 6.4‰ . H_2S produced by thermochemical sulfate reduction has $\delta^{34}\text{S}$ values 0 to 20‰ less than the sulfate source (OHMOTO and RYE 1979). The calculated range of $\delta^{34}\text{S}$ values for H_2S are 13.3 to 6.6‰ less than the sulfate in gypsum from the Alsótelekes evaporite deposit suggesting that H_2S was in fact derived from thermochemical reduction of Late Permian sedimentary sulfate. Although the calculated $\delta^{34}\text{S}$ values for H_2S are similar to those commonly observed in porphyry copper deposits (OHMOTO and RYE 1979), the lack of igneous intrusions in the area suggests that H_2S was derived from a local sedimentary source.

The sulfur isotopic composition of gypsum in the limonite ores was analyzed to determine whether sulfate was derived from oxidation of sulfides or from sulfate in evaporites. Because the isotopic fractionation associated with the oxidation of sulfides is very small, sulfate produced by oxidation of sulfides will have $\delta^{34}\text{S}$ values that are the same as the sulfides (OHMOTO and RYE, 1979). The gypsum analyzed has a $\delta^{34}\text{S}$ value of 3.8‰ that is similar to the sulfides but not the evaporites (Fig. 5). The $\delta^{34}\text{S}$ value of the gypsum in the limonite ores is therefore consistent with a supergene origin and is representative of the average isotopic composition of preexisting sulfides in siderite ore.

5. PROCESSES OF ORE FORMATION

Ore depositional processes were evaluated based on the geology of the deposit, temperature estimates, isotopic results, and solubilities of ore and gangue minerals. The introduction of Fe, basemetals, and SO_4 suggests

that the ore fluid was a relatively oxidized, acidic, saline, SO₄-rich fluid that transported Fe and basemetals as chloride complexes. The red bed sequence of the Perkupa Anhydrite and the Bódvaszilás Sandstone are the most obvious source for the Fe and basemetals. Surprisingly, the sulfur isotopic composition of barite suggests that SO₄ was derived from the Paleozoic black shales and not from Permian evaporites and siliciclastic rocks. Since siderite and most of the basemetal sulfides precipitated prior to barite, it is likely that Fe and basemetals were initially derived from fluids moving through red beds and that later on fluids containing SO₄ were derived from the black shales. The isotopic composition of the sulfides suggest that H₂S was generated locally by thermochemical reduction of Late Permian sedimentary sulfate by reaction with organic matter in the carbonate host rocks. Since Ba is relatively insoluble in SO₄-rich fluids, it must have been transported by the H₂S-rich fluid in the carbonates. Because quartz precipitates during cooling, the relative lack of hydrothermal quartz in the ores suggests that the ascending ore fluids were not appreciably hotter than the host rocks. The abundance of hematite, rather than siderite, in the deeper levels of the system suggests that the ore fluids lacked appreciable CO₂. Rather, the CO₂ in siderite was derived from the carbonate host rocks. These considerations suggest that Rudabánya formed along tensional faults where ascending ore fluids, derived from the Perkupa Anhydrite, Bódvaszilás Sandstone and Paleozoic black shales, reacted with dolomites and indigenous ground water. This model is essentially the same as that originally proposed by CSALAGOVITS (1973a, b) but is inconsistent with models for polymetallic-siderite-quartz veins (BEAUDOIN and SANGSTER 1992).

6. SUMMARY

The mineral paragenesis and stable isotopic data provide the following constraints on genetic models for Rudabánya:

1. The carbon and oxygen isotopic composition of the host dolomite is similar to normal marine limestones suggesting that the dolomite is diagenetic in origin and not a hydrothermal alteration product.
2. The carbon and oxygen isotopic composition of the siderite is consistent with results from MVT Zn-Pb deposits.
3. The sulfur isotopic composition of barite suggests that the SO₄ in ore fluids was derived from Lower Paleozoic sedimentary rocks correlative with the black shales of the Uppony and Szendrő Mts. and not from the Late Permian Perkupa Anhydrite.
4. The sulfur isotopic composition of the sulfides suggests that H₂S was derived from thermochemical reduction of Late Permian sedimentary sulfate.
5. The isotopic composition of sulfur in coeval sphalerite and galena yield a temperature of 115 °C.
6. The presence of marcasite in the ores is indicative of temperatures < 240 °C and pH < 5.
7. Ore precipitation took place along faults where ascending acidic, saline, sulfate-, Fe-, basemetal-rich fluids reacted with dolomite host rocks and indigenous Ba- and H₂S-rich ground waters.
8. The isotopic data confirm the mixing model of CSALAGOVITS (1973a, b) and suggest that Rudabánya formed where migrating basinal brines moved up faults produced by Middle Triassic rifting and encountered reactive carbonate rocks and indigenous ground waters.

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