

Sulphur Isotope Distribution in the Pb-Zn-Deposit Bleiberg (Carinthia, Austria)

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Von der Blei-Zinklagerstätte Bleiberg (Kärnten, Österreich) wurden 233 Sulfide und 40 Sulfate auf ihre Schwefelisotopenzusammensetzung untersucht und zusammen mit 29 bereits bekannten Daten ausgewertet. Die $\delta^{34}\text{S}$ -Werte der Sulfide liegen im Bereich von -40 bis -1 ‰. Für die einzelnen Mineralarten wurde gefunden (jeweilige Probenanzahl in Klammern): Galenit (96) -32 bis -2 ‰, Sphalerit (141) -30 bis -4 ‰, Markasit (16) -27 bis -1 ‰, Pyrit (10) -26 bis -13 ‰, Molybdänit (3) -40 bis -29 ‰, Anhydrit und Gips (8) +15 bis +20 ‰, Cölestin (1) +19 ‰ und Baryt (33) +7 bis +18 ‰. Die Häufigkeitsverteilung der $\delta^{34}\text{S}$ -Werte charakterisiert die Komplexität des Vererzungsvorganges. Sechs schichtgebundene Vererzungsabläufe vom Unterkarn bis zur Basis des Nor zeigen bei den Sulfaten nur geringe Variation. Die Herkunft des Sulfatschwefels aus dem Meerwasser ist daraus ableitbar. Die Sulfide sind durch bakteriogene Prozesse abgeschieden worden und ihre Schwefelisotopenzusammensetzung erscheint sowohl von der Lithofazies der Karbonatsedimente als auch von späteren diagenetischen Prozessen abhängig.

The sulphur isotope composition of 233 sulphides and 40 sulphates has been investigated and evaluated in combination with 29 earlier published data. The total variation of $\delta^{34}\text{S}$ values for the sulphides and the sulphates ranges from -40 up to -1 ‰ and from +7 up to +20 ‰, respectively. For the mineral species the variations are (with number of samples in brackets): galena (96) -32 up to -2 ‰, sphalerite (141) -30 up to -4 ‰, marcasite (16) -27 up to -1 ‰, pyrite (10) -26 up to -13 ‰, molybdenite (3) -40 up to -29 ‰, anhydrite and gypsum (8) +15 up to +20 ‰, coelestine (1) +19 ‰, and barite (33) +7 up to +18 ‰. The frequency distribution of the $\delta^{34}\text{S}$ values corresponds with the complexity of the ore forming processes which resulted in six strata-bound ore mineralizations. The sulphate values clearly show that the sulphate sulphur originates from sea water sulphate. The sulphides are formed by bacteriogenic processes from seawater sulphate, and their sulphur isotope composition depends on the lithofacies of the sediments as well as on the following diagenetic processes.

1 INTRODUCTORY REMARKS

1.1 *Position of the Deposit*

Bleiberg (often called Bleiberg-Kreuth, recently officially named Bad Bleiberg, 13°41' E - 46°37'N) is one of the economically most important Pb-Zn ore deposits in the Alpine Mid Triassic province, besides Mežica (Jugoslavia), Raibl/Cave di Predil and Salafossa (Italy). It is run by the Bleiberger Bergwerks-Union (BBU). The mine is located within the Gailtaler Alpen (Drauzug) mountain range, is about 10 × 2 square km in size and has extensions to the N (Rubland) and W. Review articles on the geology, mineralogy, and geochemistry of the deposit include Schroll (1953), Schulz and Schroll (1977), Brigo et al. (1977), Schroll (1978), and Schulz (1978).

1.2 *Stratigraphy of the Orebodies*

According to recent research at least six ore bearing stratigraphic levels can be distinguished:

1. Maxer Bank ore layers within the Wetterstein Dolomite sequence (Cordevolian), located about 220 m below the Raibl schists (Schulz, 1960b).

2. Kalkscholle ore breccia, a special development in the Cordevolian (Schulz, 1973 and 1975).

3. Erzkalk ore mineralizations in the uppermost 120 m of the Wetterstein Limestone sequence (Upper Cordevolian). The conformable layers appear in a sequence of marly and dolomitic banks with lagoonal and partly sabkha facies. They gave rise to the early mining activity and to the most thorough scientific research. Besides, discordant ore veins appear, which have been formed synsedimentarily as well. (Schulz, 1966 and 1968).

4. 1st Cardita ore layers (also called Unionserzzug) in the dolomites between 1st and 2nd Raibl schist (Lower Julian) (Schulz, 1960a).

5. 2nd Cardita ore layers in the dolomites between 2nd and 3rd Raibl schist (Lower Julian) (Cerny, 1981).

6. 3rd Cardita ore layers (also called Rublandstollen ores) above the 3rd Raibl schist (Upper Julian) (Siegl, 1974).

The transition from the Wetterstein Limestone to the Raibl beds (Cardita layers) is marked by the pyrite bearing Oolithbank which contains Pb-Zn ores only to a small extent. Pyrite nodules are found in the 1st Raibl schist.

The Anisian sequence contains Pb-Zn ores only in N Drauzug but not in the Bleiberg area.

1.3 *Sulphur Bearing Minerals*

In the mining area of Bleiberg the following minerals are available for sulphur isotope analysis:

a) Sulphate Minerals

Anhydrite: grey (concordant) or blue (discordant)

Gypsum: together with blue anhydrite secondarily formed in Raibl schist and in the oxidation zone.

Barite: primarily formed in concordant or discordant occurrence, secondarily in the oxidation zone.

b) Sulphide Minerals

Sphalerite (scarcely wurtzite): synsedimentary fine-grained types, colloform types (schalenblende), synsedimentary and diagenetic, recrystallized types and single crystals.

Galena: mostly recrystallized, concentrated in discordant veins.

Marcasite: in Oolithbank and in Raibl schists, further in synsedimentary ores of all types (also in so-called Bodenerz), and especially in late-diagenetic paragenesis.

Pyrite: in marly beds, in the Oolithbank, and mostly in sphaerolites in the Raibl schist.

Molybdenite (jordisite): only secondarily formed without connection to the Pb-Zn mineralization.

Within the Erzkalk ores the different sphalerite species are indicative of a three-stage succession which can be compared to that of Raibl (Schroll,

1953; di Colbertaldo, 1948): fine-grained sphalerite ("blenda cristallina") - yellow colloform sphalerite ("blenda gialla") - red colloform sphalerite ("blenda rossa") in Raibl and brown colloform sphalerite in Bleiberg.

2. SULPHUR ISOTOPE INVESTIGATION METHOD

The sample material for which the sulphur isotope data are presented in this article derive mainly from specimens collected by Schulz between 1978 and 1980, to some extent also from material collected by Schroll between 1949 und 1952, and from present mining activities.

Minerals have been picked out visually to separate the different phases as carefully as possible. Several samples have been enriched by flotation at the BBU. Barites have been cleaned from sulphides by chemical treatment.

Sulphur isotope measurements were carried out at the Institut für Radiumforschung und Kernphysik using the equipment and technique described by Pak and Felber (1974). Data are presented here in histogram form, but the precise values will be given together with sample descriptions and locations by Pak (1983).

3. SULPHUR ISOTOPE DISTRIBUTION

3.1 Previous Sulphur Isotope Investigations

The first sulphur isotope measurements of samples of Erzkalk ore minerals have been published by Schroll and Wedepohl (1972). Further values have been available from Grinenko et al. (1974). These earlier data are shown in Fig. 1 with special marks and are included in Fig. 2 and its interpretation as far as sample descriptions allow the classification of the different orebodies.

Part of this work has been included in earlier papers by Schroll (1978), Pak (1978), and Schroll and Pak (1980).

3.2 General Review on the Data

Fig. 1 shows all results obtained hitherto by sulphur isotope investigations.

Sulphate minerals, at least those of primary origin, e.g. syngenetic barite (cf. Schroll and Pak, 1980) and grey concordant anhydrite, show $\delta^{34}\text{S}$ values around +15 ‰ which are similar to those of sea water sulphate in the Mid Triassic (cf. e.g. Nielsen, 1979) and lead to the conclusion that they have been precipitated from sea water. A significantly lower value of +9 ‰ possibly indicates specific local conditions in the ore muds. A barite from the oxidation zone is still lighter (+7 ‰). Coelestine, blue anhydrite, and white gypsum (formed from anhydrite) show somewhat higher values than contemporary sea water.

Sulphide minerals vary considerably between -40 to -1 ‰. The frequency distribution of the $\delta^{34}\text{S}$ values is indicative of several different mineralization processes.

Galenas show a distinct frequency peak at -7 ‰ and statistically less significant accumulations about -13 and between -32 and -23 ‰.

Sphalerites give two distinct peaks at -26 and -6 ‰ with intermediate accumulations near -17 and perhaps near -12 ‰. The dominating peak at -26 ‰ is not obvious in Fig. 2 although Fig. 1 is a mere compilation of Fig. 2.

Pyrites and marcasites vary only between -27 and -12 ‰. The values of marcasites which are coparagenetic with the Pb-Zn mineralization (cf. Fig. 2) are similar to those of the lead and zinc sulphides. The pyrites and marcasites of Oolithbank and Raiblschist vary between -23 and -17 ‰. A quite different marcasite (-1 ‰) originates from an occurrence without Pb-Zn sulphides in a fissure on the S slope of the Dobratsch mountain (S Bleiberg) described by Schroll (1953), just above a layer of volcanic rocks. This value near zero may possibly indicate that the sulphur is of igneous origin, but could also be explained by bacterial reduction of sea water sulphate in Skythian or Anisian time.

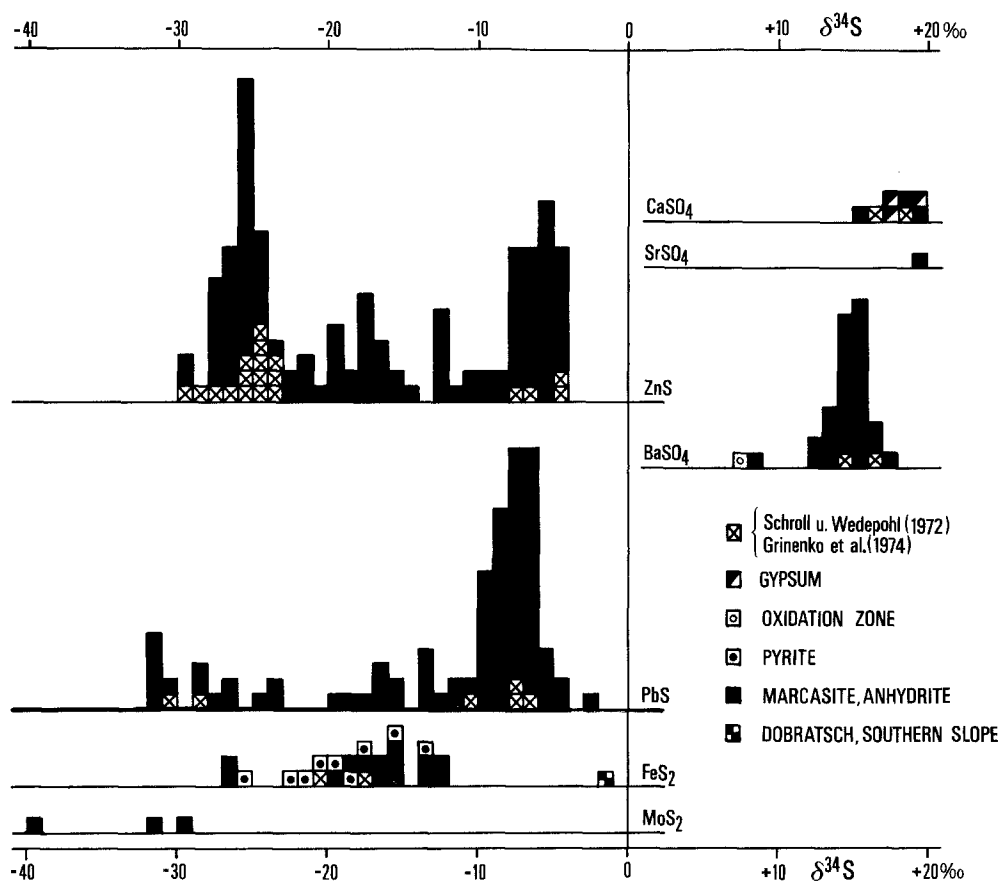


Fig. 1. Sulphur isotope distribution in the minerals of Bleiberg

Molybdenite (jordisite) contains extremely light sulphur down to -40 ‰ and must have been formed by bacteriogenic processes independently of the Pb-Zn mineralization in a late stage.

This is a confirmation of the genetic interpretation given by Schroll (1949) and Kostelka (1956).

3.3 The Different Ore Mineralizations

The sulphur isotope distribution shown in Fig. 1 must not give rise to the conception that the whole ore mineralization was the result of a single process with a definite ore succession. On the contrary, several independent phases of sedimentation at different times between the Ladinian and the Carnian must be assumed. It is possible

that some late diagenetic processes in areas of tectonic faults caused a mobilization of ores in the Erzkalk horizon. The Kalkscholle ores are influenced by dolomitization processes and found on a swell of the lagoonal Erzkalk ore mineralization. Ores of the Maxer Bank and of the 1st Cardita ore layers seem to be tectonically linked.

Fig. 2 shows the sulphur isotope distribution for the different ore mineralizations.

The Maxer Bank and Kalkscholle ores, although the numbers of samples are statistically insufficient, have similarity with each other and with the Cardita ores, with mostly very light galenas and a bimodal distribution intimated for the sphalerites.

For the Erzkalk ores the attempt has been made to classify the concordant and discordant ores separately but

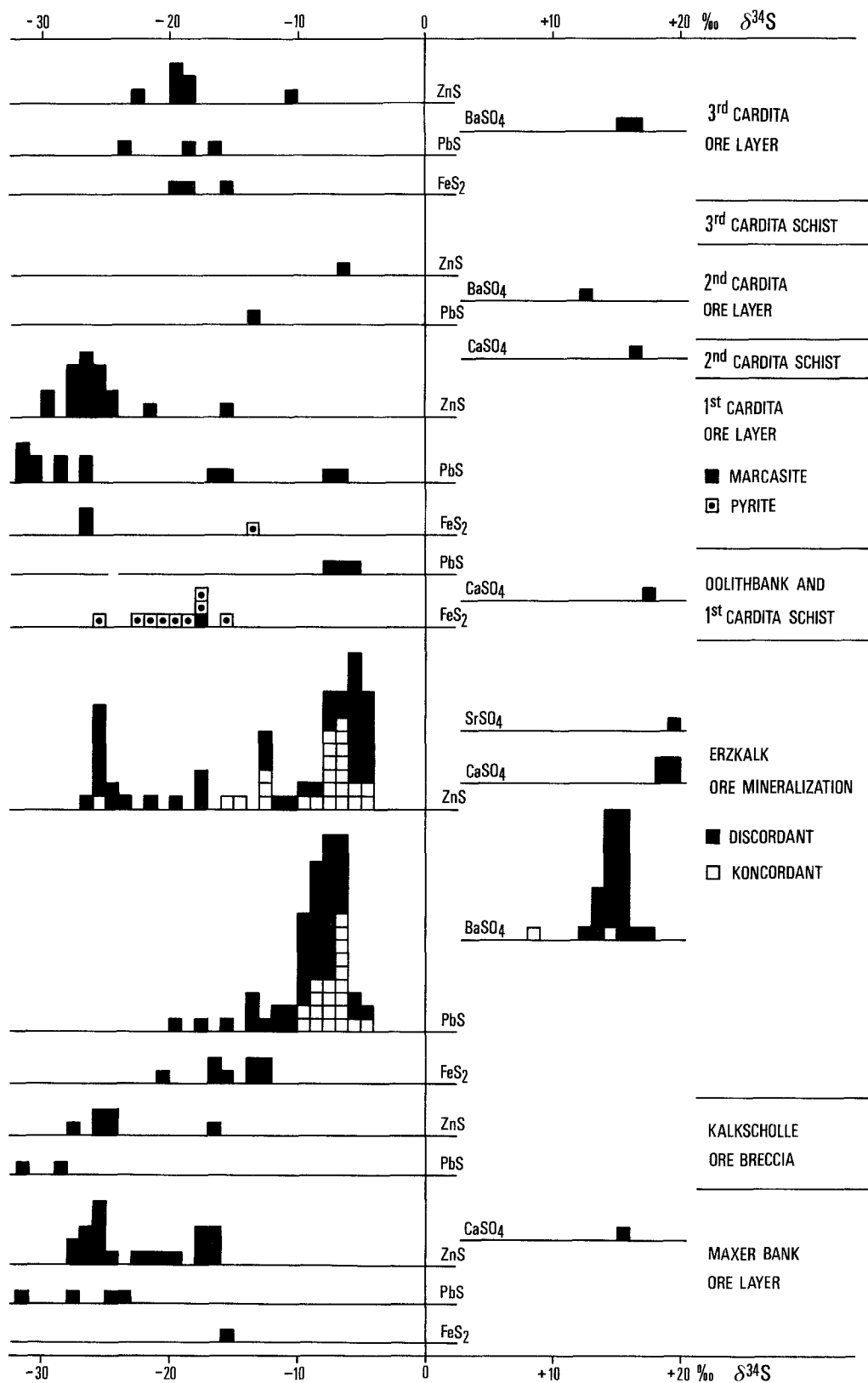


Fig. 2. Sulphur isotope distribution in the individual orebodies

it was not possible in all cases to distinguish definitely between the two types (e.g. some samples classified as discordant possibly contain syndiagenetic or recrystallized mineralizations too). It must be admitted that the number of samples from the Erzkalk ores are somewhat preponderant over those of the other ore levels. The striking peaks at about -6 ‰ for the sphalerites and about -8 ‰ for the galenas are composed of concordant as well as discordant samples whereas the discordant ones also extend to much lighter values. For the sphalerites a moderate accumulation is seen around -25 ‰ , these values belonging to colloform sphalerites (schalenblende) and breccia mineralizations. Marcasites from the Erzkalk ores are somewhat heavier than the iron sulphides from Oolithbank and Raibl schists.

From the 1st Raibl clay-schist a few galenas, which can be found only very scarcely, were remarkably heavier than the iron sulphides and lay in the range of the distinct peak of the Erzkalk galenas.

The 1st Cardita ore layers contain mostly light sulphur in the rather narrow range between -32 and -24 ‰ . Similarly light sulphur about -19 ‰ has been found in other Cardita ores of the Drauzug region (Schroll and Pak, 1983).

In the 2nd and 3rd Cardita ore layers the sulphides contain heavier sulphur than in the 1st one and are accompanied by barite. Still heavier sulphur (-3 ‰ for galena and -6 ‰ for fine-grained sphalerite) was found in the 3rd Cardita mineralization of Mitterberg approximately 20 km W Bleiberg (Schroll and Pak, 1983).

3.4 Observations in Detail

Recrystallization of galena and sphalerite alters the sulphur isotope composition only slightly. At Jauken (some 40 km W Bleiberg), fine-grained sphalerite gave $+2.4\text{ ‰}$ and coarsely recrystallized sphalerite $+1.6\text{ ‰}$. At Bleiberg, in a colloform sphalerite (schalenblende) the dark zone was 1.3 ‰ lighter than the fair zone. In

a 1.7 cm large galena octahedron the core was 1.7 ‰ lighter than the marginal zone. From this example it can be seen that even single crystals need not have a homogeneous sulphur isotope composition.

Open systems seem to have prevailed during the ore succession of the Erzkalk ore mineralization. In so-called cockade ores three generations of sphalerite with distinctly different sulphur isotope composition can be found: I, fine-grained fair sphalerite, older than the breccia formation, with layered structure; II, yellow or orange sphalerite, mostly with colloform structure; III, enveloping dark-brown colloform sphalerite (schalenblende). In a single specimen of such an assemblage e.g. $\delta^{34}\text{S}$ of -4.1 , -12.8 and -23.7 ‰ were found for the three generations respectively. Further progress in such investigations can be expected in the near future from secondary ion mass spectrometry (SIMS) which will render possible isotope micro analysis.

4. INTERPRETATION OF THE RESULTS

Considering the complexity of the deposit, a still much higher number of analyzed samples would be necessary for statistically well secured conclusions. In addition, the numbers of the data give some preference to the Erzkalk mineralization over the others.

4.1 Sulphur Isotope Correspondence Between Sulphate Minerals and Sea Water Sulphate in the Triassic

Upper Skythian or Anisian gypsum samples from the S slope of Dobratsch (S Bleiberg) gave an average of $+27\text{ ‰}$ (Strehl et al., 1980), and similar values were measured for other Anisian gypsums of the Alps (Pak, 1974).

Barites from the Erzkalk (Cordevolian) have an arithmetic mean of $+15\text{ ‰}$. For the corresponding barites of Windisch-Bleiberg, Raibl and Gorno values around $+14\text{ ‰}$ were found (Schroll

and Pak, 1980 and unpublished). Therefore sea water sulphate in the Cordevolian can be supposed to have had a $\delta^{34}\text{S}$ in the range of +14 ‰ to +15 ‰. The values of blue anhydrite and coelestine are somewhat higher. Investigations of present sabkha evaporites (Butler et al., 1973) also gave increased values for calcium sulphate minerals.

In the 3rd Cardita ore layers barites (yet very few) lie near +16 ‰, which seems to be the $\delta^{34}\text{S}$ for Julian sea water sulphate (cf. e.g. Nielsen, 1979).

4.2 Sulphide Minerals

The sulphur isotope distribution of all sulphide minerals is in accordance with the assumption that their sulphur derives from bacterial reduction of sea water sulphate.

Schwarzc and Burnie (1973) stated that the sulphur isotope composition of sedimentary sulphides depends on the conditions for sedimentation: in euxinic environment with slow sedimentation, much clay and organic material, and an open system for sulphates, sulphides are 40 to 50 ‰ lighter than sea water sulphate, with modest variation. On the other hand, in marginal shallow basins with high sedimentation rates, less organic material, and a rather closed system for sulphate, the fractionation between sulphate and sulphides is less by approx. 20 ‰ with more variation. Fruth and Maucher (1966) found at Gorno differences of the sulphur isotope composition between sphalerites of swell and basin facies.

Also at Bleiberg, sulphides of calcareous, less bituminous lagoonal facies of the Erzkalk are isotopically heavier than those of bituminous, dolomitic facies which is characteristic for the Cardita layers. The difference between the $\delta^{34}\text{S}$ values of the sulphides of the 1st Cardita ore layer and the Upper Triassic sea water is similar to the corresponding difference between the Kupferschiefer sulphides and the Upper Permian sea water (Marowsky, 1969). A similar dependence on lithofacies has been obtained for

many other ore occurrences of the Drauzug and Karawanken region (Schroll and Pak, 1982). A detailed investigation of the relation between the lithofacies and the sulphur isotope distribution is still to be done.

During early diagenesis of the ore mud closed systems must be assumed with little if any alteration of the sulphur isotope composition. Recrystallization and other early diagenetic processes seem to have taken place without considerable biological activity. However, for the sphalerite succession of Erzkalk with its multi-peak sulphur isotope distribution also during diagenesis bacteriogenic influences in open systems can be supposed. It may also be that isotopically light sulphur from iron sulphides of Oolithbank and Raibl layers has been introduced to the Erzkalk sulphide minerals.

Bacteriogenic formation of the ores implies low temperatures about 30°C, and later warming up by superposition and tectonic events did not exceed 100°C (Kappel and Schroll, 1982). Therefore equilibrium isotope fractionation between coexisting sulphide minerals cannot be expected (cf. Ohmoto and Rye, 1979).

5. CONCLUSIONS

1. Sulphate minerals of Bleiberg were formed from sea water sulphate, as well as the sulphide minerals by bacteriogenic reduction.

2. The sulphur isotope distribution of the syndiagenetic sulphides depends primarily on the facies conditions of the sedimentation: Dolomitic bituminous sediments contain lighter sulphides than calcareous, less bituminous ones. The 1st Cardita ore layer belongs preferably to the euxinic type, the Erzkalk ore layers to the other one.

3. Diagenesis and redeposition of minerals scarcely alter the sulphur isotope composition. Multi-stage processes, mostly in an open system with bacterial precipitation, generate increasingly light sulphides, the youngest molyb-

denites becoming the lightest. Late-stage evaporites (blue anhydrite and coelestine) are somewhat heavier than the barites.

4. It is in accordance with the assumption of low-temperature genesis that sulphur isotope equilibria do not occur during ore mineralization.

5. Further progress can be expected from sulphur isotope measurements in micro areas by SIMS and from correlative investigations of geochemical, lithofacial and isotopic data.

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