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TETRAHEDRITE FROM THE DUBRAVE – DUGI DOL BARITE DEPOSITS, KREŠEVO, BOSNIA AND HERZEGOVINA

Ivan JURKOVIĆ¹ and Slobodan MIKO²

¹ Faculty of Mining, Geology and Petroleum Engineering, Univ. of Zagreb, Pierottijeva 6, HR-10000 Zagreb, Croatia

² Institute of Geology, Sachsova 2, HR-10000 Zagreb, Croatia

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The chemical composition of the tetrahedrite from the barite deposit Dubrave – Dugi Dol, south of the town Kreševo, Bosnia, hosted by Devonian Dolomite is as follows: $(\text{Cu}_{10.65}\text{Ag}_{0.073}\text{Hg}_{0.03})_{10.75}(\text{Fe}_{0.81}\text{Zn}_{0.94})_{1.75}(\text{Sb}_{3.57}\text{As}_{0.34}\text{Bi}_{0.004})_{3.91}\text{S}_{13.00}$. Cation proportions are: $\text{Me}^{2+}:\text{Me}^{3+}:\text{S} = 3.20:1.00:3.33$. The $\delta^{34}\text{S} = -10.91\%$. The calculated formula is in good correspondence with the compositions of other analyzed tetrahedrites from the whole Middle Bosnian Schist Mountains. These results as those of the fluid inclusion studies of gangue minerals suggests the generation of tetrahedrites from highly homogenized, hot (190–310 °C), high saline (24.2–26.3 wt% NaCl equiv.) mineralizing solutions generated by mixing of ascending deep seated upper mantle (Hg, F, Cu-bearing) and metamorphogenic lower crust (Zn, Fe, Ag, Au-bearing) fluids with descending highly evolved Upper Permian saline formation waters. The obtained data exclude the applicability of the fractional crystallization generation model.

Ključne riječi: živin-antimonski tetraedrit, kemijski i izotopski sastav, baritsko ležište Dubrave, Kreševo, Bosna.

Kemijski sastav tetraedrita iz baritnog ležišta Dubrave – Dugi Dol, Kreševo, Bosna je sljedeći: $(\text{Cu}_{10.65}\text{Ag}_{0.073}\text{Hg}_{0.03})_{10.75}(\text{Fe}_{0.81}\text{Zn}_{0.94})_{1.75}(\text{Sb}_{3.57}\text{As}_{0.34}\text{Bi}_{0.004})_{3.91}\text{S}_{13.00}$. Odnos kationa je: $\text{Me}^{2+}:\text{Me}^{3+}:\text{S} = 3.20:1.00:3.33$. The $\delta^{34}\text{S} = -10.91\%$. Formula tetraedrita se vrlo dobro uklapa u opći raspon sastava analiziranih tetraedrita iz cijelog područja Srednjobosanskih škriljavih planina. Navedeni rezultati kao i rezultati studije fluidnih inkluzija minerala jakovine upućuju na postanak tetraedrita iz vrlo homogeniziranih, vrućih (190–310 °C), jako slanah (24.2–26.3 wt% NaCl equ.) mineralizirajućih otopina generiranih miješanjem uzlaznih, duboko smještenih u gornjem omotaču (nosilaca Hg, F, Cu) i metamorfenih iz donjeg dijela kore (nosilaca Zn, Fe, Ag, Au) sa silaznim, jako slanim formacijskim vodama gornjopermskog evaporitnog horizonta. Dobiveni analitički rezultati isključuju primjenu modela frakcione kristalizacije za genuzu tetraedrita.

Introduction

Barite deposits Dubrave and Dugi Dol are situated about 1 km south-east of the summit of Međuvršje (+1195 m) and 3 km south-east of the town of Kreševo in the Mid-Bosnian Schist Mountains (fig. 1).

The mineralization occurs in dolomites, and assigned with some reservation as Devonian by Jovanović et al., (1977/78) based on analogy with the identical fossiliferous rocks in the Prozor area (Živanović, 1972). Ore occurrences are in the form of irregular nests, lense-shaped veins, networks of veins and veinlets, locally they occur as ore-breccias in tectonically dislocated dolomites (Fig. 2a, 2b). The ore occurrences have an east west extension. There are also significant allochthonous, residual barite deposits on a karstified relief of dolomite (fig. 2c).

The Dubrave deposit is situated at an elevation from +970 m to +1000 m, in a belt 30 m to 50 m wide. The Dugi Dol deposit occurs at elevations from +880 m to +930 m.

The deposit paragenesis is described in detail by Jurković (1987). *Barite* is the main and preponderant ore mineral. Its platy habit is relatively frequent, and granular structure (polygonal, allotriomorphic, hypidiomorphic, recrystallized, locally breccia-like) is most frequent (Plate I, phot. 1 and 3) *Tetrahedrite* is the most abundant sulphide, but it participates only with <1 wt% to 5 wt%. *Pyrite* (Plate I, phot. 4), *chalcopyrite I and II*, two unidentified *copper sulphosalts*, *colloform pyrite* and *ascendent goethite* are present only as accessories in these deposits.

Tetrahedrite forms nests, veinlets, incrustations, small rounded masses, dispersed idiomorphic grains replacing barite along the margins of grains or along fine cracks

and fissures (Plate I phot. 1 and 2). It is, in places, very strongly cataclased.

Subordinate gangue minerals *quartz I and II* occur as small nests, small irregular masses, or system consisting of more or less dense arrays of veinlets. Transparent *fluorite*, violet in colour or colourless, noticeably corroded (Barić, 1952), scarce leafs of *sericite* and *rutile* crystals are accessories. Ferrodolomitization, and simultaneous recrystallization of dolomite is stronger in the more silicified parts of the deposit.

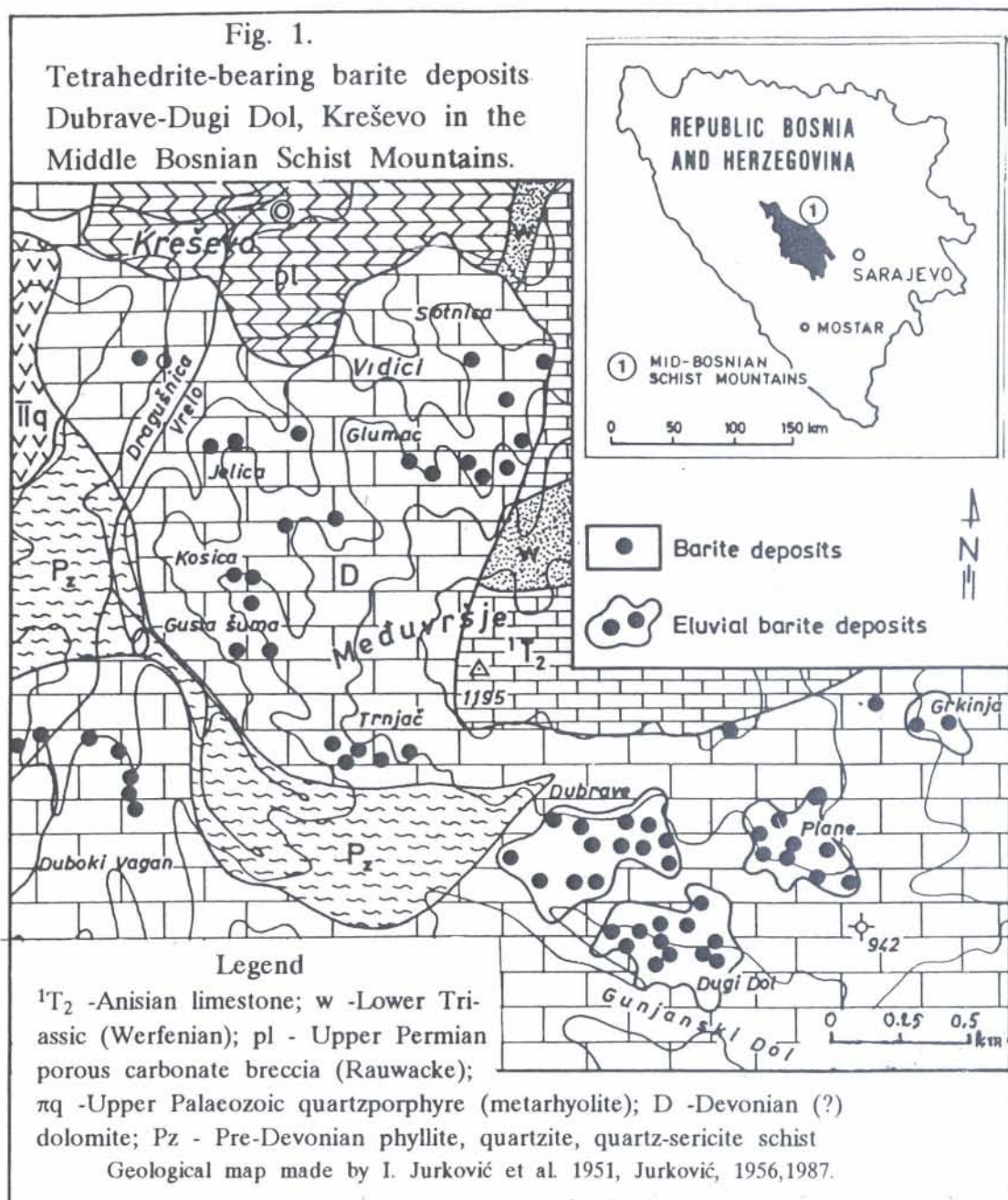
Recently, Radanović-Gužvica (1995) investigated two barite crystals from the deposit Dubrave. The habit of these crystals corresponds to the rectangular-platy type according to Braun (1932). They are thick-platy or columnar crystals, elongated parallelly with one of the crystallographic axis [100] with forms: (001), (010), (210), (011), (101), (213). Chemical analyses made by the ICP-AES method (Jobin-Yvon 50 Polyscan instrument) gave: crystal K₁ 2.93 wt% SrSO₄, crystal K₂ 0.38 wt% SrSO₄.

Analytical methods and results

I Strontium contents in barites

The two barite samples were investigated: 215/1 (thick platy aggregates) and 215/2 (recrystallized masses) by dr. D. Siftar who utilized his own spectrographic method (Siftar, 1974), and was controlled with a method by Gordon et al., (1959).

The obtained values of 7.32 wt% SrSO₄ for sample 215/1, and of 5.1 wt% SrSO₄ for sample 215/2 are relatively high and characteristic for barites from the Kreševo area (Siftar, 1988).



II Sulphate sulphur isotopic composition of barite

The barite sample 215/2 prepared and measured in the Institute Jože Štefan (Ljubljana) gave: $\delta^{34}S = +11.35\text{‰}$.

III Sulphide sulphur isotopic composition of tetrahedrite

The tetrahedrite sample T1 was analysed in the Institute Jože Štefan (Ljubljana). The $\delta^{34}S$ of this tetrahedrite sample is -10.91‰ .

IV Chemical analyses of tetrahedrites

For the chemical analyses we collected small (1 to 3 mm in diameter) individual dispersed tetrahedrite crystals found within the dolomite fragments inside the barite bodies. The grains were more or less rounded or with the terminal planes. All collected material was carefully purified under the binocular microscope. The tetra-

drite crystals and grains prepared for analyses were fresh, not weathered.

Sample preparation for AAS and ICP-AES analysis

Powdered tetrahedrite samples weighing 0.1 g were digested with a mixture consisting of 2 ml Br_2 and 15 ml conc. HCl on a water bath at $65^\circ C$. The resulting solutions were diluted to 100 ml with 10% HCl.

The solutions were analyzed by flame atomic absorption spectrophotometry for the following elements: Ag, Au, Sb, As, Bi, Pb, Cd, Mn, Fe, Ni and Cr. Mercury was analyzed by cold vapor AAS after Hg reduction with $SnCl_2 \times H_2O$ (10% wt. solution) and H_4CINO (hydroxylammonium chloride 20% wt. solution). All analysis were performed on a Pye Unicam SP9 AAS instrument. Copper was analyzed by a Jobin Yvon 50P simultaneous ICP-AES spectrometer with RSD of 2%. Accuracy was controlled with USGS geochemical standards GXR-1

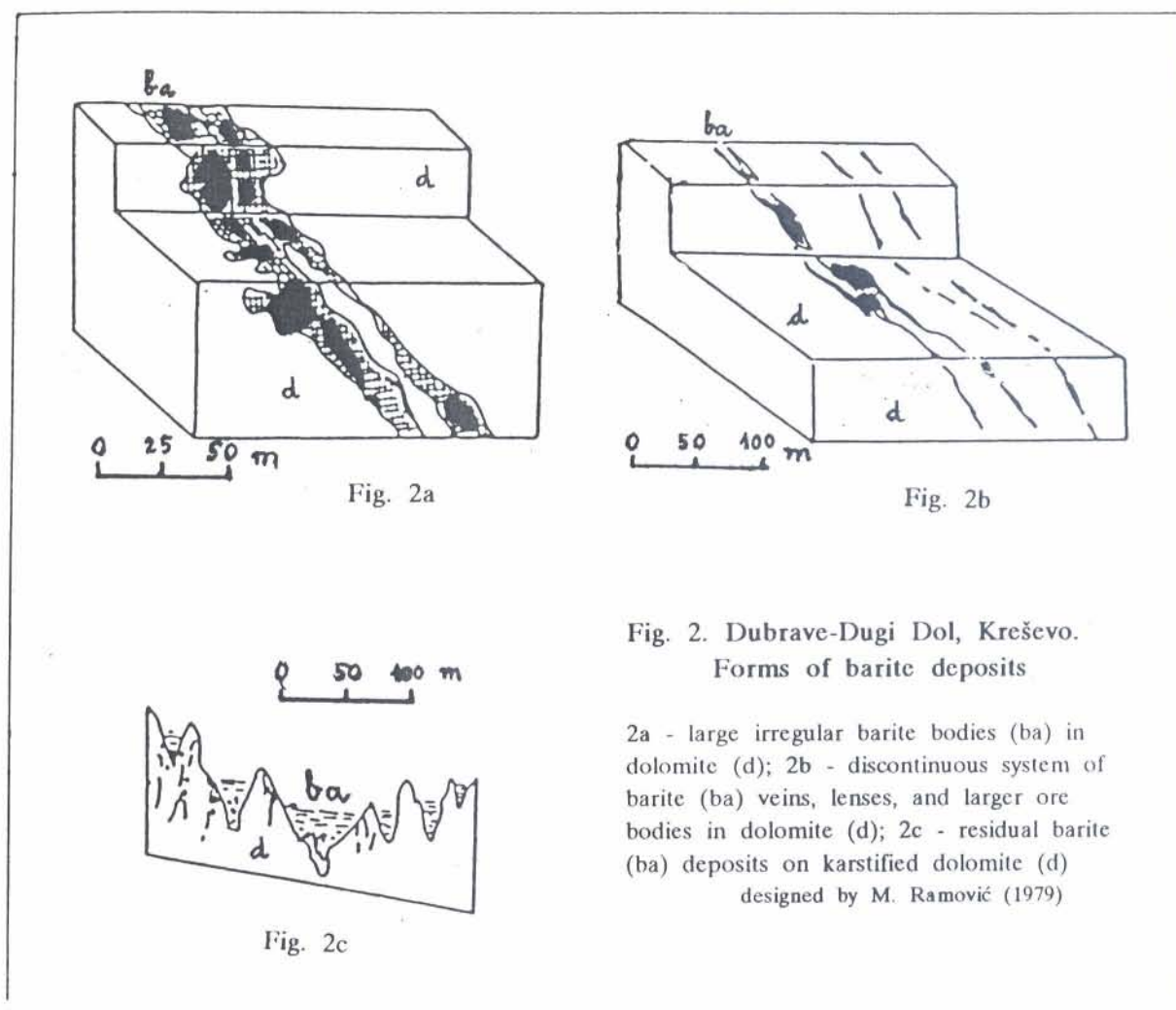


Fig. 2. Dubrave-Dugi Dol, Kreševo.
Forms of barite deposits

2a - large irregular barite bodies (ba) in dolomite (d); 2b - discontinuous system of barite (ba) veins, lenses, and larger ore bodies in dolomite (d); 2c - residual barite (ba) deposits on karstified dolomite (d)
designed by M. Ramović (1979)

Table 1. Chemical composition and cation proportions of the tetrahedrite sample 215, Dubrave – Dugi Dol barite deposit

Elements		Wt%	Cation proportions	
			Elements	Me ²⁺ +Me ³⁺ +S
Me ²⁺	Fe	2.67	0.047809	0.740756
	Cu	40.09	0.630882	
	Zn	3.65	0.055836	
	Hg	0.383	0.001909	
	Ag	0.466	0.004320	
Me ³⁺	Sb	25.73	0.211335	0.231437
	As	1.49	0.019887	
	Bi	0.045	0.000215	
S	S	24.70	0.770334	0.770334
	Total	99.224	1.742527	1.742527
Me ²⁺ : Me ³⁺ : S = 3.20 : 1.00 : 3.33				

$X_{Ag} = 0.68$ $X_{Sb} = 91.4$ $X_{Fe} = 46.1$

Table 2. Four partial chemical analyses of tetrahedrites from Dubrave – Dugi Dol deposit (samples T₁-T₄) as compared with the complete chemical analysis of the sample 215 (analyst S. Miko).

Components	Tet-1	Tet-2	Tet-3	Tet-4	215
Fe	2.68	2.60	2.71	2.60	2.67
Cu	41.25	37.59	38.45	41.45	40.09
Zn	3.98	3.86	3.65	3.89	3.65
Hg		0.32	0.29	0.31	0.383
Ag	0.26	0.27	0.26	0.20	0.466
Sb	23.81			24.24	25.73
As	2.69				1.49
Bi	0.067			0.063	0.045
S	22.14			24.54	24.70

Utilizing ICP-AES analysis the authors obtained the following additional very similar values for copper: 41.62; 40.54; 41.17 and 39.78 wt%.

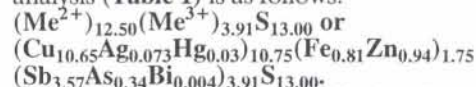
Table 3. Trace elements (in ppm) in tetrahedrite from the Dubrave – Dugi Dol barite deposit

Element	Tet-1	Tet-2	Tet-3	Tet-4	215
Co	8.5			8.7	
Ni				6.7	40.0
Au	9.3			7.2	
Cd	154.0	179.0	134.0	157.0	130.0
Mn	16.6		19.6	18.9	90.0
Pb	94.0	88.0		90.0	110.0
Cr	18.5			22.0	
Mo ⁺	< 40	< 40	< 40	< 40	
Se ⁺	< 30	< 30	< 30	< 30	

⁺ The values for molybdenum and selenium are below detection limit in the tetrahedrite

and GXR-4. Sulphur was analysed by »wet« chemical method.

The calculated formula on the basis of the performed analysis (Table 1) is as follows:



The tetrahedrite from the barite deposit Dubrave – Dugi Dol (Kreševo) belongs to the group of typical **antimonian tetrahedrites** with small partition of silver and mercury.

Authors performed four additional partial chemical analyses (AAS-technique) of tetrahedrites from the same deposit (Table 2) with an intention to define the composition span of the main tetrahedrite components (in wt%). The values for iron, zinc, mercury, silver and bismuth show relatively narrow ranges, whereas the values for copper, antimony, arsenic and sulphur display a wide range.

V Trace elements

The following trace elements have been analyzed by AAS: copper, nickel, gold, cadmium, manganese, lead, chromium, molybdenum and selenium (Table 3).

Table 4. Formulae of tetrahedrites from the Middle Bosnian Schist Mountains (MBSM) ore deposits

No	Locality	Formulae, cation proportions, analysts
1	Mačkara Gornji Vakuf	$(\text{Cu}_{11.65}\text{Ag}_{0.03}\text{Hg}_{0.73})_{12.41}(\text{Fe}_{0.97}\text{Zn}_{0.21})_{1.18}(\text{Sb}_{4.34}\text{As}_{4.34}\text{S}_{13.00})$ $\text{Me}^{2+} : \text{Me}^{3+} : \text{S} = 3.13 : 1.00 : 2.90$ Analyst: Patera in F. Pocchi (1900)
2	Mačkara Gornji Vakuf	$(\text{Cu}_{8.83}\text{Hg}_{0.48}\text{Ag}_{0.31})_{9.31}(\text{Fe}_{1.02}\text{Zn}_{1.02})_{2.04}(\text{Sb}_{3.48}\text{As}_{0.52}\text{Bi}_{3.80}\text{S}_{13.00})$ $\text{Me}^{2+} : \text{Me}^{3+} : \text{S} = 2.72 : 1.00 : 3.42$ Analyst: V. Vesely (1921)
3	Mačkara Gornji Vakuf	$(\text{Cu}_{9.79}\text{Ag}_{0.02}\text{Hg}_{0.63})_{10.41}(\text{Fe}_{1.14}\text{Zn}_{1.14})_{2.28}(\text{Sb}_{2.76}\text{As}_{1.04}\text{Bi}_{0.03}\text{S}_{13.00})$ $\text{Me}^{2+} : \text{Me}^{3+} : \text{S} = 3.03 : 1.00 : 3.32$ Analyst: I. Jurković (1960)
4	Vidici Kreševo	$(\text{Cu}_{10.90}\text{Ag}_{0.03}\text{Hg}_{0.16})_{11.09}(\text{Fe}_{1.28}\text{Zn}_{0.31})_{1.60}(\text{Sb}_{3.52}\text{As}_{0.63}\text{Bi}_{4.15}\text{S}_{13.00})$ $\text{Me}^{2+} : \text{Me}^{3+} : \text{S} = 3.06 : 1.00 : 3.14$ Analyst: I. Jurković (1986)
5	Trošnik Fojnica	$(\text{Cu}_{10.13}\text{Ag}_{0.03}\text{Hg}_{0.02})_{10.18}(\text{Fe}_{1.86}\text{Zn}_{0.49})_{2.35}(\text{Sb}_{3.62}\text{As}_{0.25}\text{Bi}_{0.02}\text{S}_{13.00})$ $\text{Me}^{2+} : \text{Me}^{3+} : \text{S} = 3.22 : 1.00 : 3.34$ Analyst: I. Jurković (1959)
6	Dubravec Kreševo	$(\text{Cu}_{10.65}\text{Ag}_{0.073}\text{Hg}_{0.03})_{10.75}(\text{Fe}_{0.81}\text{Zn}_{0.94})_{1.74}(\text{Sb}_{3.57}\text{As}_{0.34}\text{Bi}_{0.004})_{3.91}\text{S}_{13.00}$ $\text{Me}^{2+} : \text{Me}^{3+} : \text{S} = 3.20 : 1.00 : 3.33$ Analyst: S. Miko, this paper

Plate I

- Phot. 1. Dubrave – Dugi Dol, Kreševo. Polished section, parallel nicols. Tetrahedrite (td, light gray) replaces coarse grained barite (ba, dark gray).
 Phot. 2. Dubrave – Dugi Dol, Kreševo. Thin section, parallel nicols. Tetrahedrite (td, black) replaces barite (ba, light gray). Inside tetrahedrite sieved relics of replaced barite. Barite crystals are strongly cataclased.
 Phot. 3. Dubrave – Dugi dol, Kreševo. Thin section, crossed nicols. Allotriomorphic texture of barite (ba).
 Phot. 4. Dubrave – Dugi Dol, Kreševo. Polished section, parallel nicols. Pyrite grains (py, light gray) zonary replaced by tetrahedrite (td, dark gray). Barite (ba, almost black) is the main gangue mineral.

Discussion

In Table 4 are presented the formulae of six tetrahedrite samples from the Middle Bosnian Schist Mountains (MBSM) ore deposits calculated on the basis of performed chemical analyses. Five of them (1–5) are already published, the sixth is the result of analysis presented in this paper. Ten additional complete chemical analyses of MBSM tetrahedrites are printed recently (Jurković et al., 1997).

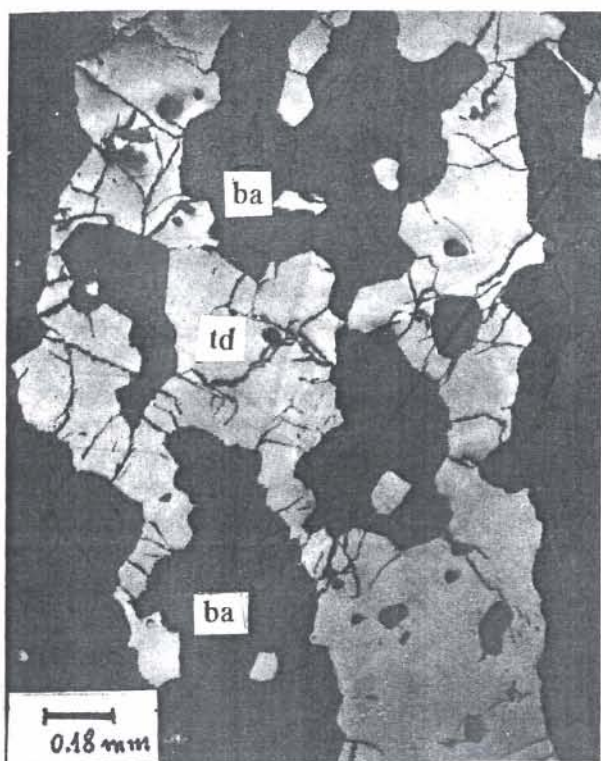
The obtained most characteristic results are as follows:

(1) Tetrahedrites from the *Gornji Vakuf area*: (a) those in the Mačkara siderite-barite vein hosted by pre-Devonian metamorphic rocks, and (b) those in the Sabiljine Pećine, big irregular barite bodies, hosted by Devonian carbonate rocks, are distinguished by high contents of mercury (5 wt% in average, but locally up to 16 wt% Hg). This area is particularly suitable (permeable) for inflow of mercury-bearing fluids deriving from deep parts of the degassed upper mantle along the first-order Voljevac fault which stretches NNW-SSE as do all ore veins in the same area.

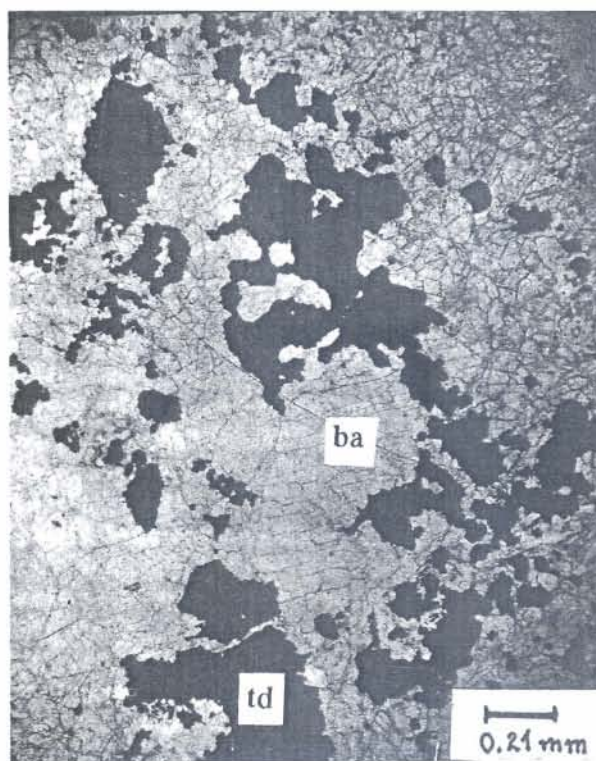
The tetrahedrites from the Gornji Vakuf area are typical **mercurian-antimonian tetrahedrites or schwartzites**. The zinc content of these tetrahedrites is very low.

(2) Tetrahedrites from the *Kreševo area* barite deposits, hosted by Devonian dolomites, which are distant to the major first-order Busovača fault on the eastern margin of the MBSM, are confined to second-order faults which were evidently less permeable for ascending fluids. This is reflected in the lowering of the mercury content, which averages 2.7 wt% Hg. The content of zinc is elevated and that of iron lowered. These tetrahedrites also belong to the group of **mercurian-tetrahedrites** with relatively lower mercury content.

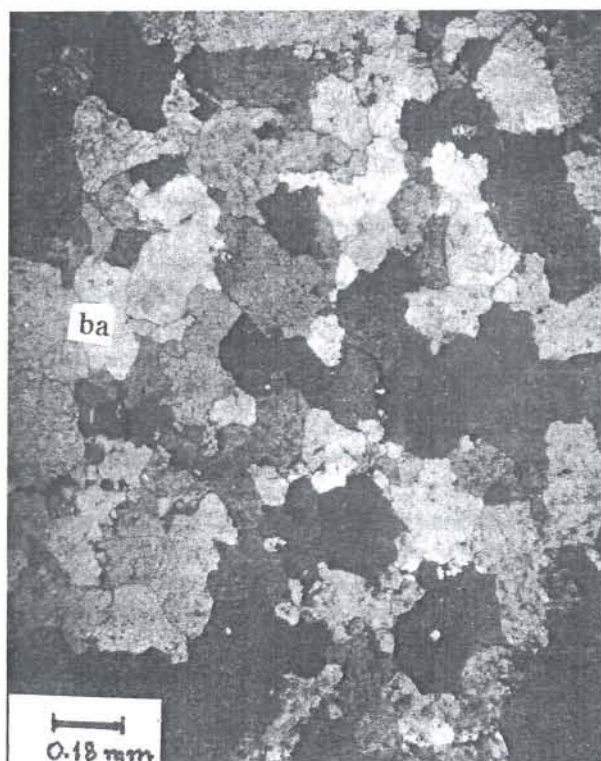
(3) Tetrahedrites from the *Fojnica area* (the Trošnik deposit) hosted by carbonate rocks intercalated in the highest horizon of pre-Devonian metamorphic rock complex, represent typical **ferroan, antimonian tetrahedrites** with only traces of mercury, and low arsenic content.



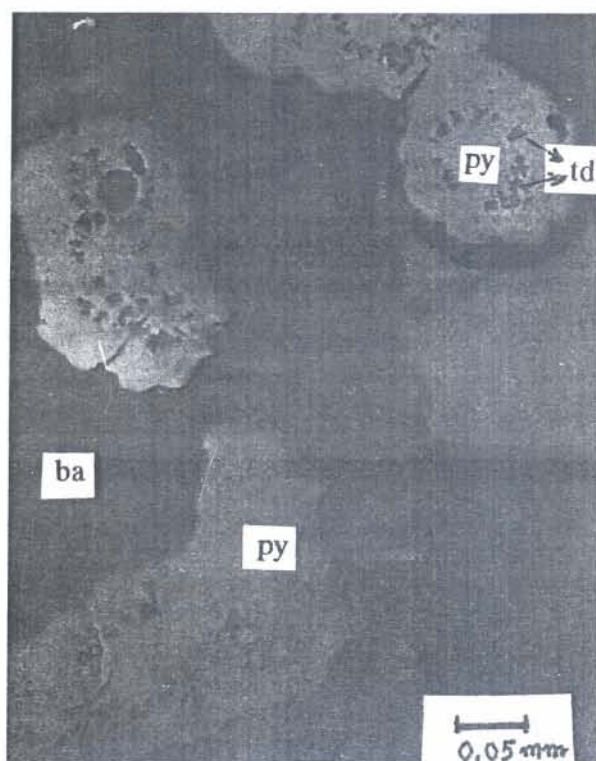
phot. 1



phot. 2



phot. 3



phot. 4

(4) All tetrahedrites from the MBSM are gold-bearing and contain (in ppm): 50; 30; 21.8; 27.3; 62; 154.5; 8.3; 13.4; 11.6 (Jurković et al., 1997) – an average 42.1 = 42.1 g/t. Mercury-bearing tetrahedrites from Brixlegg, Austria (Lukas, 1971; Gstrein, 1983) and those from the Rudnany deposit, Slovakia (Cambel-Jurkovski et al., 1985) are not gold-bearing.

(5) Tetrahedrites from the MBSM deposits contain (from 14 analyzed tetrahedrite samples) only 0.20 wt% Ag varying from 0.083 to 0.62 wt%. Since the range is relatively narrow, the model of fractional crystallization proposed for the argentinian tetrahedrites from the Orcopampa deposit (Peru) by Hackbarth & Petersen (1984) and Petersen et al., (1990), can not be applied for the MBSM tetrahedrite deposits.

(6) The fluid inclusion study (Palinkaš & Jurković, 1994) of (a) quartz, fluorite, and barite crystals from the Kreševo tetrahedrite-bearing barite deposits; (b) of barite crystals from the Mačkara tetrahedrite-bearing siderite-barite deposits, and (c) of quartz crystals from the Busovača monomineral quartz deposits, implies a simultaneous and uniform generation of tetrahedrite bearing, ore deposits from the uniformly extended mineralizing fluids through the whole Middle Bosnian Schist Mountains (MBSM). These fluids were highly homogenized, hot (190^o–310 °C), and saline (24.2 to wt% NaCl equiv.).

They were formed by mixing of the ascending hydrothermal deep seated magmatic and/or metamorphic fluids with descending highly evolved Upper Permian formation (evaporite) waters.

This homogeneity of fluids is also supported by the fact that all 18 analyzed tetrahedrites showed pronounced negative $\delta^{34}\text{S}$ values (ranging from -5.5‰ to -15.4‰, on the average -10.48‰) suggesting the uniform generative process.

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