

THE CISMA DEPOSIT, BAIUT DISTRICT, EASTERN CARPATHIANS, ROMANIA: SPHALERITE COMPOSITION AND FORMATION CONDITIONS

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Abstract: Sphalerite at the Cisma deposit (Baiut ore district, Eastern Carpathians, Romania) occurs in two generations formed in different environments. Sphalerite-1 is characterized by a negative correlation between Fe and Mn and precipitated under equilibrium conditions. Sphalerite-2 was deposited in a non-equilibrium environment; it contains chalcopyrite inclusions and shows positive correlation of Fe and Mn. Fluid inclusion data on quartz identify boiling and point to depositional temperatures of 270 to 316°C in the Bandurita vein and 143 to 344°C in the Cisma vein at a paleo-depth of 500-800 m. Salinity (Na- and Mg-chloride) is less than 10 wt.% eq. NaCl for both veins. The various zoning patterns in sphalerite derive from a combination of boiling, cooling, and dilution of the ore-forming fluid.

Key words: epithermal, base metal, Baia Mare, Baiut, sphalerite, fluid inclusions

1. INTRODUCTION

Although geology, mineralogy, and fluid regime of most base metal deposits of the Baia Mare region have been studied in detail (e.g., Grancea et al., 2002; Ciobanu et al., 2006, etc.), little has been published on the Baiut district, which occupies the easternmost part of the Gutai Mountains of the Inner Carpathians. A few exceptions e.g. (Costin, 2003; Costin & Vlad, 2005, etc.) concerned the Varatec and Breiner deposits only. This paper is aimed to provide novel data on the chemical composition of sphalerite which is one of the main ore minerals and to derive conclusions on the fluid regime for the Cisma deposit.

2. GENERAL GEOLOGY AND MINERALOGY

The Baiut district is part of the Baia Mare region and is situated in the easternmost part of the Gutai Mountains with a surface area of approximately

40 km² (Fig. 1). The stratigraphic sequence is represented by Paleogene sedimentary rocks which belong to Tocila – Secu Trans Carpathian Flysch (Bombiță, 1972), overlain by Lower Neogene sediments and Upper Neogene (Pontian) andesites. The sedimentary groups of the Trans Carpathian Flysch are disposed in the two structural units: the Botiza nappes and the Lapus paraautochthonous domain. The Upper Cretaceous (Turonian to Senonian) layered marlstones occur only in the Botiza Nappe. Upper Jurassic pelagic facies rocks and breccias occur as tectonic klippen. Paleogene sediments (Eocene marls, clays, and grits) dominate in the area and are represented by three horizons: lower, middle and upper ones (Fig. 2).

The sedimentary rocks are intruded by Late-Neogene quartz micromonzodiorite and microgranodiorite porphyries. The intrusive magmatic rocks are affected by propylitic and phyllic alteration, and have hornfels aureoles in albitic and chloritic facies.

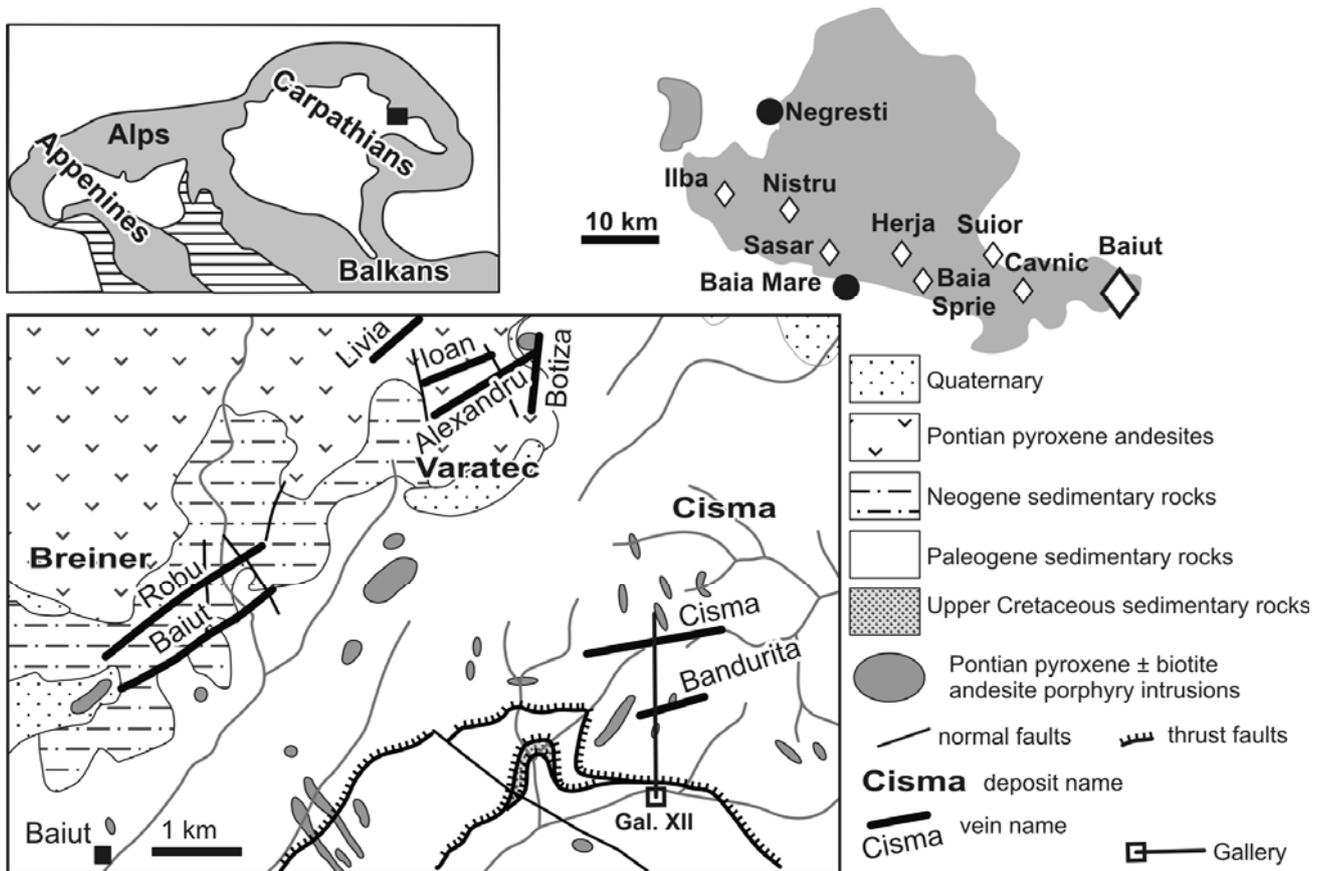


Figure 1. Geological sketch map of the Baiut area, after (Borcos & Gheorghita, 1976) with minor changes

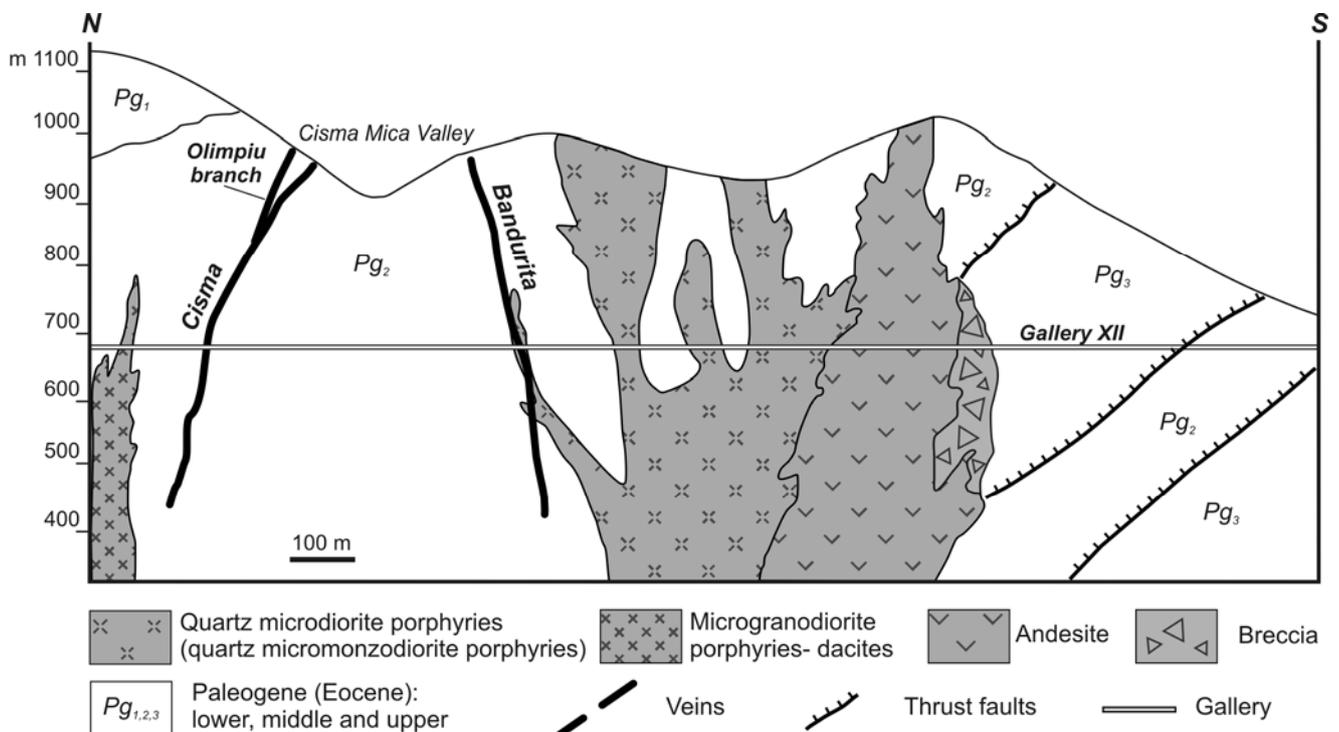


Figure 2. Cross section through the Cisma (Cisma Poiana Botizei) deposit

The Baiut district includes three Pb-Zn-Cu-Au deposits: Cisma (Cisma Poiana Botizei), Breiner and Varatec (Borcos & Gheorghita, 1976).

The Cisma Poiana Botizei deposit is hosted by Paleogene sedimentary rocks intruded by Neogene quartz microdiorite and microgranodiorite

porphyries (Fig. 2). It consists of two main NEE striking veins: Cisma and Bandurita. The deposit was explored from mid-1900s to 2006; the total production was approximately 3 Mt of ore at 1% Cu, 0.7% Zn and 0.5% Pb.

Damian & Damian (2004) identified two mineralization stages at the Cisma deposit. The early stage minerals are: hematite, pyrite, chalcopyrite, tennantite, tetrahedrite and quartz with minor sphalerite, galena, wolframite, pyrrhotite, bismuthinite, emplectite, lillianite-gustavite series, galenobismutite, native bismuth. Mineralization of the second (or base-metal) stage contains galena, sphalerite, pyrite, chalcopyrite, marcasite, native gold, stibnite, realgar, orpiment, bournonite, semseyite, boulangerite, jamesonite. Native gold forms small inclusions in pyrite and occurs mostly in the upper part of the Cisma vein.

The Cisma vein is situated in the middle horizon of the Eocene Tocila–Secu flysch. It dips NNW and is 2 km long and 600 m deep; in the central zone its thickness is up to 5–6 m and more. Intervals of maximum thickness (up to 10 m) alternate with wedging-out zones with a thickness of about 1 m. The main vein has several branches.

In the Cisma vein the early stage mineralization dominates. The vein contains bismuth minerals (Damian et al., 2010), similar to copper mineralization in other deposits of the Baia Mare area (Damian, 2000). The base-metal stage mineralization is confined to marginal and apical parts of the Cisma vein. Stibnite is abundant in the Olimpiu branch of the vein. The vein is zoned inward from massive ore to impregnations in gangue minerals. Those are represented by quartz, and small amounts of carbonates, chlorite and clay minerals. In the central zone of the vein bands of massive ore alternate with bands of quartz with intercalations of the argillized and silicified host rocks.

The Bandurita vein is located in the contact area of the micromonzodiorite porphyry intrusion and in the Eocene flysch. The vein dips SSE and has a length of about 1500 m and the maximum thickness is 1.5 m. In the Bandurita vein base-metal stage mineralization is more wide-spread than in Cisma and thus, Pb and Zn contents are higher. The mineralization consists of pyrite, galena, sphalerite, chalcopyrite, and arsenic minerals. Gangue minerals are represented by quartz and abundant carbonates. There are appreciable quantities of orpiment and realgar associated with marcasite. The vein is zoned inward from pyrite-rich to galena and sphalerite rich. The central part of the vein is predominantly composed of carbonates.

3. SPHALERITE COMPOSITION

The chemical composition of ore and gangue minerals was studied with a VEGA-II Tescan scanning electron microscope with Oxford Instruments EDX detector at IEM RAS, Chernogolovka, Moscow region (A. Virus, analyst) and with a Cameca-SX-100 electron microprobe with four WDX detectors, TU Clausthal, Germany (A.R. Cabral, analyst).

For the Cameca SX-100 the following conditions were used: accelerating voltage 20 keV, sample current 20 nA, beam diameter 1 μm . For sphalerite samples: $K\alpha$ for S, Cu, Fe, Zn, and Mn, $L\alpha$ for Cd; standards: ZnS for S and Zn, CuFeS_2 for Cu and Fe, CdS for Cd, and pure Mn for Mn. Peak time was 10 s for S and Zn, 20 s for Cu, 60 s for Mn and 100 s for Cd. Detection limits (wt.%) were: 0.08 for S, 0.09 for Cu, 0.07 for Cd, 0.15 for Zn, 0.03 for Fe and 0.02 for Mn.

The base-metal mineralization in the investigated samples is represented by pyrite, chalcopyrite, galena, abundant low-Fe sphalerite, and minor tennantite. Sphalerite is represented by two generations (Figs. 3 and 4). Sphalerite-1 overgrows earlier pyrite and is free of any mineral inclusions. It has Fe contents up to 4.6 wt.% at Cisma and up to 2.8 wt.% at Bandurita, and variable admixtures of Mn (from 0.48 to 0.71 wt.%) and relatively constant Cd contents (0.21 to 0.25 wt.%). As shown on Fig. 3, contents of Fe and Mn establish a negative correlation.

Sphalerite-2 overgrows sphalerite-1 and is characterized by abundant chalcopyrite inclusions (Fig. 4). It has variable admixtures of Fe (from 1.9 to 3.2 wt% at Cisma and to 4.5 wt% at Bandurita), Mn (0.1 to 0.2 at Cisma and 0.04 to 0.24 at Bandurita) and Cd (0.21 to 0.26 and 0.27 to 0.45). Contents of Fe and Mn show remarkable positive correlation (Figs. 4 and 5a) with $r = 0.72$ ($n=28$, $r_{\min} = 0.38$).

4. FLUID INCLUSION STUDY

Microthermometric measurements were carried out on a THMSG 600 (Linkam) heating stage mounted on an Olympus microscope in the Institute of Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry of Russian Academy of Sciences (IGEM RAS), Moscow.

The temperature of phase transitions was measured within the range -196 to $+600^\circ\text{C}$ with an accuracy of $\pm 0.2^\circ\text{C}$ within the interval $+20$ to -20°C and about $\pm 1.0^\circ\text{C}$ beyond this interval. The salt composition of the fluids was determined from the eutectic temperature (T_{eut}) according to

Borisenko (1977); the salinity (C) was estimated from the temperature of ice melting (T_m) using the FLINCOR software (Brown, 1989). The measurements were carried out for groups of inclusions with similar phase relationships to avoid

errors related to the disintegration of vacuoles after fluid heterogenization (Roedder, 1984). Pressure (P) was calculated for coexisting vapour- and liquid-rich fluid inclusions using the FLINCOR software (Brown, 1989).

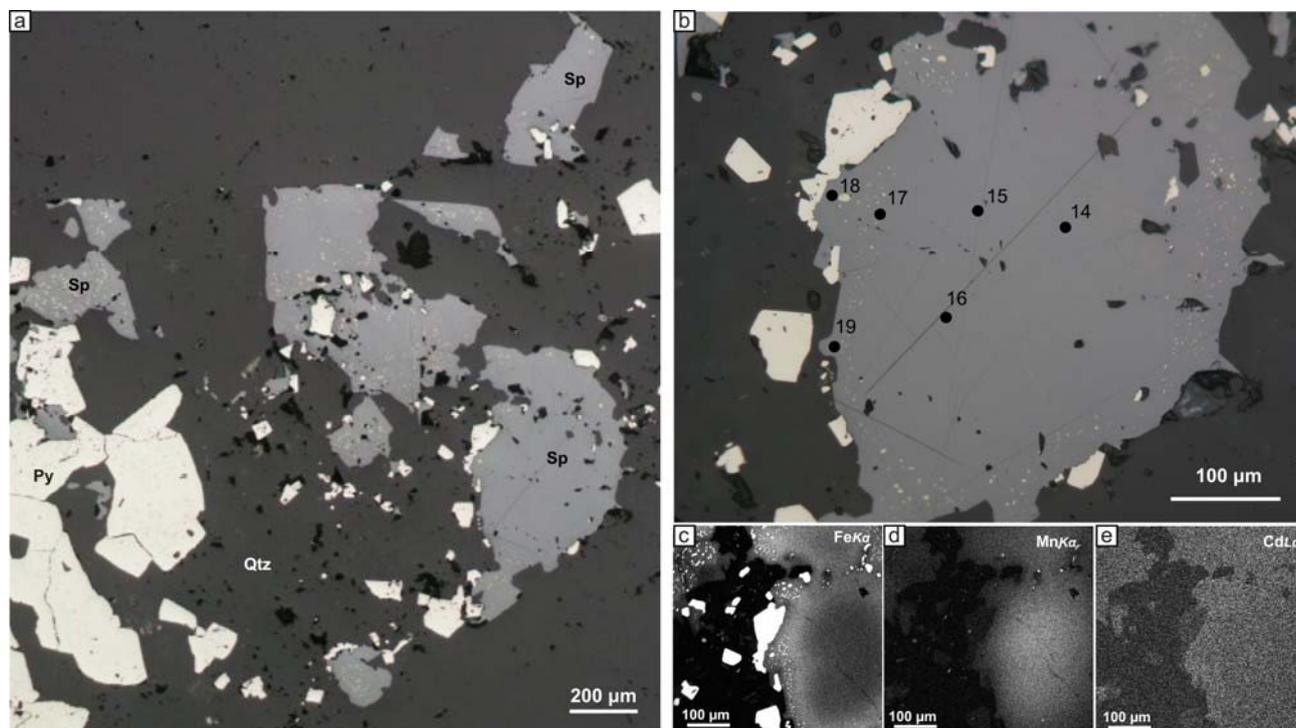


Figure 3. Zoned sphalerite 1 from the Bandurita vein. (a,b) – reflected light, (c) to (e) X-ray maps of FeK_{α} , MnK_{α} and CdL_{α} respectively. Note the remarkable negative correlation between Mn and Fe. Sp – sphalerite, Py – pyrite, Qtz – quartz.

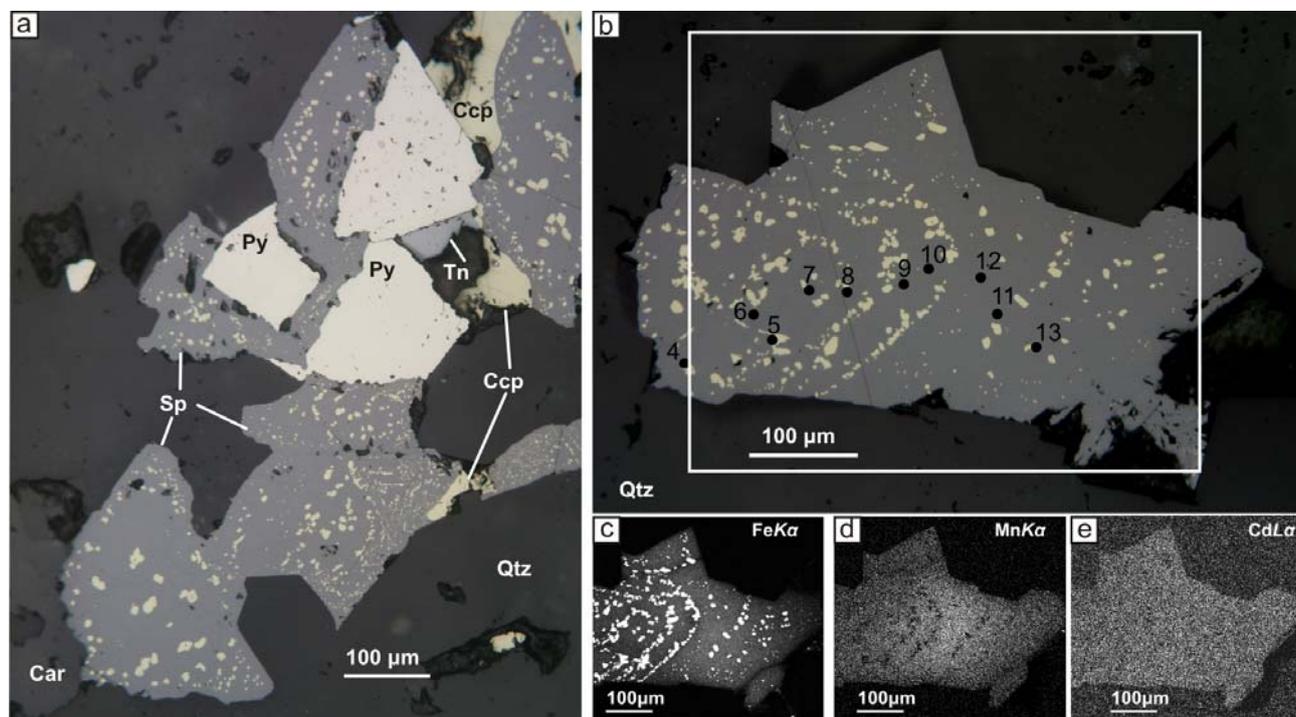


Figure 4. Zoned sphalerite-2 and associated minerals from the Cisma vein. (a, b) reflected light, (c) to (e) X-ray maps of FeK_{α} , MnK_{α} and CdL_{α} respectively. Note the positive correlation between Mn and Fe. Sp – sphalerite, Py – pyrite, Ccp – chalcopyrite, Tn – tennantite, Qtz – quartz, Car – carbonate.

Table 1. Chemical composition of sphalerite (wt.%) from Bandurita and Cisma veins

No	Zn	Fe	Mn	Cd	Cu	S	Total	XFeS	No	Zn	Fe	Mn	Cd	Cu	S	Total	XFeS
Cisma Cs-310 (level XII), Sphalerite 1									Bandurita Bd-127 (level XII), Sphalerite 1								
1*	59.23	4.62	0.70	bdl	bdl	32.38	96.93	8.22	14	64.30	1.52	0.62	0.25	bdl	33.25	99.93	2.64
2*	57.39	4.56	0.41	bdl	bdl	32.07	94.42	8.30	15	63.98	1.73	0.59	0.21	bdl	32.83	99.39	3.03
3*	59.20	4.38	0.54	0.63	bdl	31.89	96.64	7.88	16	64.40	1.56	0.71	0.23	bdl	32.81	99.74	2.72
Sphalerite 2									Sphalerite 2								
4	63.84	2.39	0.10	0.25	0.22	32.57	99.36	4.19	17	63.29	2.88	0.48	0.23	bdl	33.39	100.31	4.98
5	64.26	1.88	0.10	0.25	0.20	32.24	98.93	3.32	18	62.89	2.51	0.08	0.32	0.10	33.42	99.31	4.37
6	64.01	2.36	0.13	0.23	0.16	32.07	98.96	4.17	19	63.78	2.24	0.12	0.33	bdl	32.86	99.36	3.92
7	63.84	2.43	0.14	0.25	0.25	32.50	99.41	4.26	20	63.51	2.41	0.14	0.31	0.57	33.10	100.04	4.19
8	63.75	2.40	0.15	0.24	0.27	32.27	99.07	4.23	21	63.18	2.12	0.06	0.36	0.42	33.42	99.57	3.69
9	63.23	2.64	0.17	0.23	0.14	32.34	98.76	4.66	22	60.99	4.53	0.22	0.29	bdl	33.05	99.09	7.91
10	64.03	2.27	0.16	0.26	0.16	32.15	99.02	4.00	23	60.91	4.35	0.23	0.27	0.10	33.41	99.26	7.56
11	63.12	3.02	0.22	0.21	bdl	32.27	98.91	5.32	24	60.98	4.50	0.24	0.31	bdl	32.84	98.95	7.88
12	63.01	3.04	0.22	0.26	bdl	32.06	98.67	5.38	25	64.32	1.30	0.04	0.45	0.07	32.95	99.13	2.28
13	62.67	3.15	0.20	0.27	bdl	32.42	98.80	5.55	26	64.08	1.87	0.05	0.37	0.16	32.93	99.45	3.27

Note: * – EDX data, bdl – below detection limit. Low totals in EDX data are caused by matrix capture.

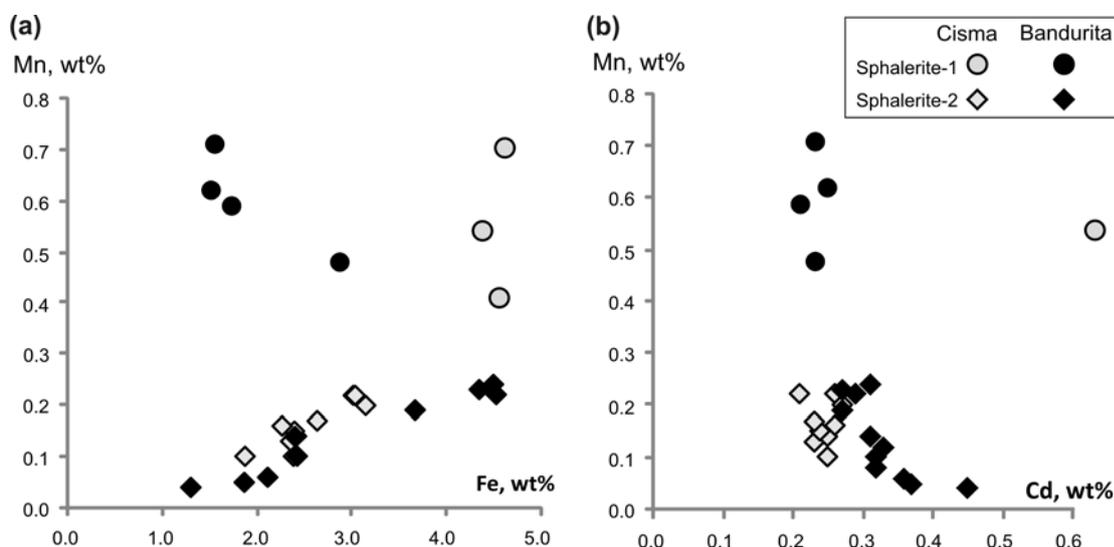


Figure 5. Fe vs Mn (a) and Mn vs Cd (b) variations in sphalerite from Cisma and Bandurita veins.

Fluid inclusions suitable for microthermometric investigations were found in 7 samples from the Cisma vein and in 3 samples from the Bandurita vein. In most samples fluid inclusions were abundant and were confined to growth zones of quartz crystals (Fig. 6a and b) and thus were considered as primary (Roedder, 1984). Microscopic observations at room temperature revealed two types of fluid inclusions: V type, vapor rich inclusions with minor liquid phase (Fig. 6c) and L-V type, liquid inclusions with vapor bubble as much as 25-30 vol.% of the inclusion volume (Fig. 6d and e). No solid phases or liquid CO₂ were observed. Fluid inclusions of V and L-V types are often confined to

the same growth zone pointing to fluid heterogenization (boiling).

The results obtained from 250 fluid inclusions are presented in Table 2 and Fig. 7. Fluid salinity (C) varies from 0.4 to 11.1 wt.% eq. NaCl. Eutectic temperatures of L-V type inclusions range from -21 to -40°C suggesting the presence of Na, K, and Mg chlorides (Borisenko, 1977). Homogenization temperature (T_{hom}) ranges from 143 to 344°C. Boiling is observed in most samples and suggests that the T_{hom} data obtained are the real temperatures of ore formation, and no pressure corrections are required (Roedder, 1984). Fluid density (d) of L-V type inclusions is 0.64–0.95 g/cm³, while V-type inclusions contain low-density H₂O vapor.

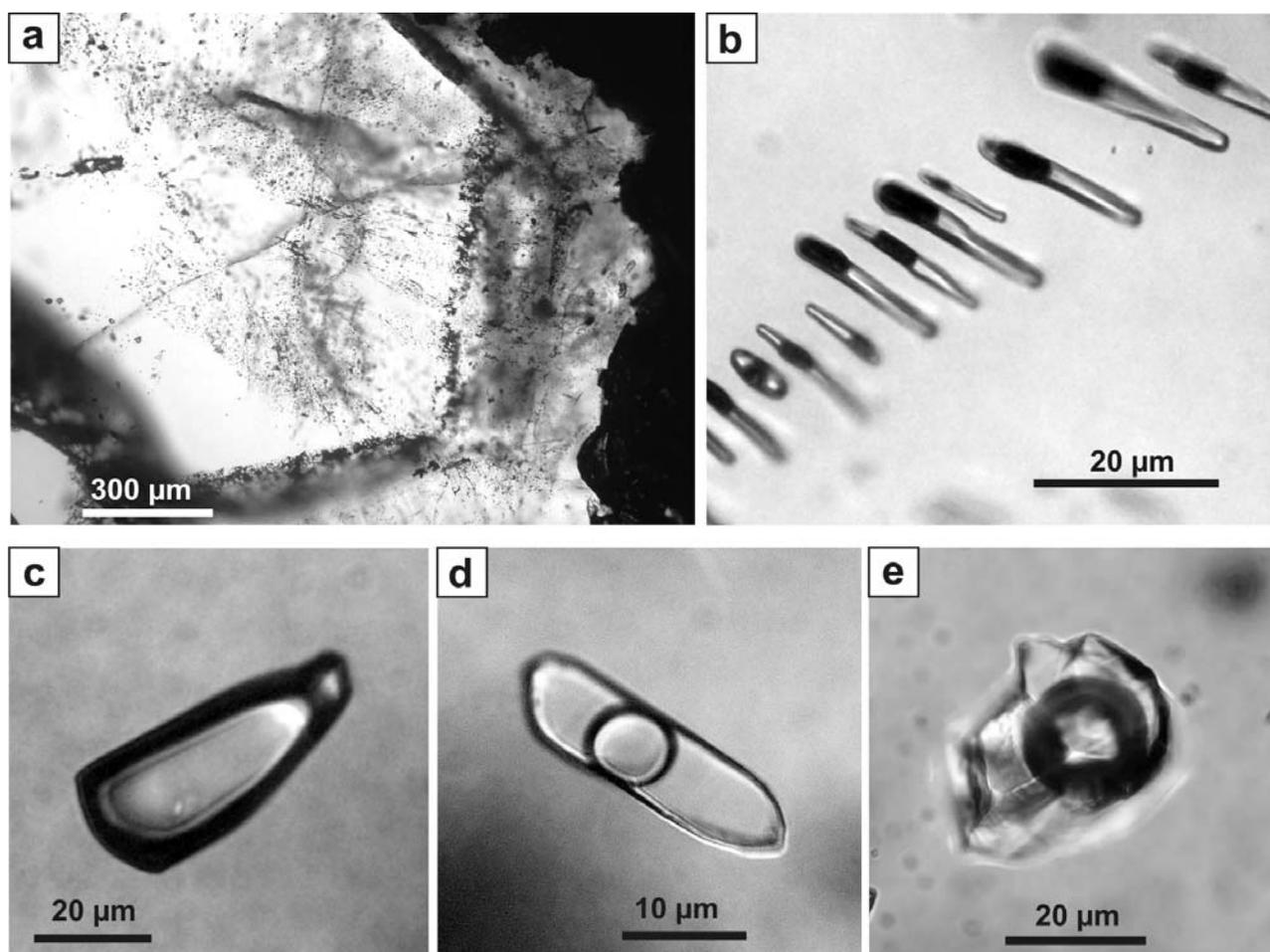


Figure 6. Fluid inclusions from Cisma and Bandurita veins. A – fluid inclusions in quartz crystal, Bandurita vein; B – fluid inclusions along growth zone of quartz crystal, Cisma vein; C – vapor rich fluid inclusion, Cisma vein; D and E – liquid rich fluid inclusions, Bandurita vein.

The results obtained are in a good agreement with fluid inclusion data observed by Costin and Vlad (2005) for the Varatec deposit of the same ore district: T_{hom} from 228 to 357°C and salinities 0.5 to 3.4 NaCl wt.% equiv.

Pressure (P) calculated for coexisting inclusions of V and L-V types defines 50 to 80 bars. This suggests formation depth of approximately 500 to 800 m for a hydrostatic regime. This depth roughly corresponds with the observation of (Maldarescu & Popescu, 1980) that maximum erosion interval in the area was 400-500m.

Fluid inclusions of the Cisma deposit form two main trends in T_{hom} – salinity space (Fig. 7): (i) wide salinity range (almost one order of magnitude) at relatively constant homogenization temperature was observed both at the Cisma and Bandurita veins, and (ii) narrow salinity range (2-4 wt.-%-NaCl eq) at variable T_{hom} (340 to 140°C) is a feature of some samples from the Cisma vein only. The first trend is typical for fluid boiling while the second may indicate cooling of the fluid perhaps due to dilution by meteoric waters.

5. DISCUSSION

Di Benedetto et al. (2005) noted a zonal partitioning between Fe and Mn admixtures in zoned sphalerite crystals, which was explained by an absorption process at the mineral-liquid interface controlled by a competition between Mn and Fe on crystal growth. Such a mechanism is possible only if sphalerite grows relatively slowly and in equilibrium with the fluid. However, in the Cisma deposit a negative correlation between Mn and Fe was noted in sphalerite of the first generation, i.e. sphalerite free of chalcopryrite inclusions (Figs. 3 and 5a). In sphalerite of the second generation (with chalcopryrite inclusions) higher contents of Mn and Fe are confined to the same growth zones (Figs. 4 and 5a). This might be a result of a rapid crystallization in non-equilibrium environment when sphalerite coprecipitated simultaneously with chalcopryrite and captured all possible chemical admixtures. The presence of non-equilibrium conditions is supported by wide-spread fluid boiling observed from fluid inclusion petrography. Cadmium in most cases demonstrates homogeneous distribution.

Table 2. Data for primary fluid inclusions in quartz from Bandurita and Cisma veins

n^*	T_{hom} , °C	C, wt. % eq, NaCl	T_{evt} , °C	$T_{\text{m. ice}}$, °C	d, g/cm ³	P, bar	n^*	T_{hom} , °C	C, wt. % eq, NaCl	T_{evt} , °C	$T_{\text{m. ice}}$, °C	d, g/cm ³	P, bar
Bd-127, Level XII, 320m East part of the Bandurita vein							Cs-771, Level XII, 500m East part of Cisma vein						
14*	270	1.2	-24	-0.7	0.77	50	16	297	3.1	-34	-1.8	0.74	-
4	272	0.9	-22	-0.5	0.76	-	5	267	2.9	-38	-1.7	0.80	-
16	316	8.3	-34	-5.3	0.78	-	3	301	4.0	-36	-2.4	0.75	-
3	274	7.7	-31	-4.9	0.84	-	3	298	2.4	-36	-1.4	0.73	-
7	295	6.0	-33	-3.7	0.79	-	4	283	3.2	-30	-1.9	0.77	-
8	299	0.7	-35	-0.4	0.70	-	Cs-773, Level XII						
4	273	0.9	-30	-0.5	0.76	-	4	286	2.4	-26	-1.4	0.75	-
Bd-835, Level XI, 1224m East part of the Bandurita vein							3	284	2.4	-24	-1.4	0.76	-
17*	297	3.2	-30	-1.9	0.74	80	Cs-369, Level XII						
5*	300	2.6	-31	-1.5	0.73	80	4	257	2.4	-22	-1.4	0.81	-
Bd-66, Level XII, 30m West part of the Bandurita vein							3	250	2.2	-25	-1.3	0.81	-
13*	281	4.5	-32	-2.7	0.79	60	5	262	3.1	-27	-1.8	0.81	-
23*	300	4.2	-30	-2.5	0.75	80	Cs-706, Level XI, 95m West part of II Branch Cisma						
4	283	2.6	-30	-1.5	0.76	-	3	344	4.2	-33	-2.6	0.66	-
Cs-155, Level XII, East part Cisma vein							3	296	3.7	-32	-2.2	0.75	-
12*	280	2.2	-30	-1.3	0.76	60	4	293	3.7	-34	-2.2	0.76	-
7*	273	2.1	-34	-1.2	0.77	50	3	198	3.9	-32	-2.3	0.90	-
Cs-313- Level XII, East part of Cisma vein							3	143	3.4	-31	-2.0	0.95	-
3	267	2.6	-21	-1.5	0.79	-	4	300	4.0	-34	-2.4	0.75	-
Cs-714 Level XII							5	295	3.4	-31	-2.0	0.75	-
3	327	5.6	-27	-3.4	0.72	-	3	301	3.2	-32	-1.9	0.74	-
4	272	11.1	-29	-7.5	0.88	-	6	299	4.0	-35	-2.4	0.75	-
3	322	0.4	-40	-0.2	0.64	-	3	190	2.2	-30	-1.3	0.90	-
3	326	7.2	-26	-4.5	0.75	-	3	283	2.2	-29	-1.3	0.76	-

n – number of inclusions;

* – fluid heterogenization (coexisting vapor- and liquid- rich fluid inclusions), data are given for liquid-rich fluid inclusions.

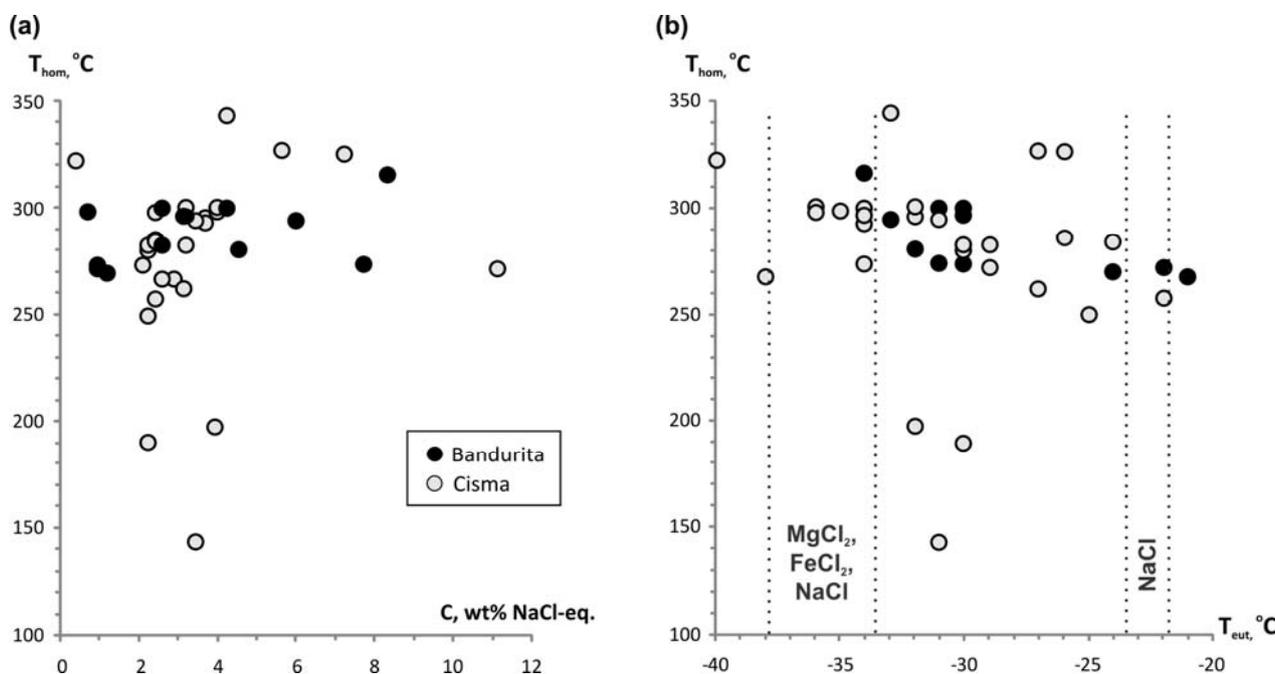


Figure 7. Homogenization temperature (T_{hom}) vs salinity (a) and T_{hom} vs eutectic temperature (T_{eut}) diagram (b) for Cisma and Bandurita veins.

Among all chemical impurities in sphalerite only Fe is known to be an important indicator of physical-chemical conditions (Vaughan & Craig, 1997,

etc). The FeS content in sphalerite (X_{FeS}) is known to be a function of sulfur fugacity, temperature and pressure (Vaughan & Craig, 1997). Pressure, however,

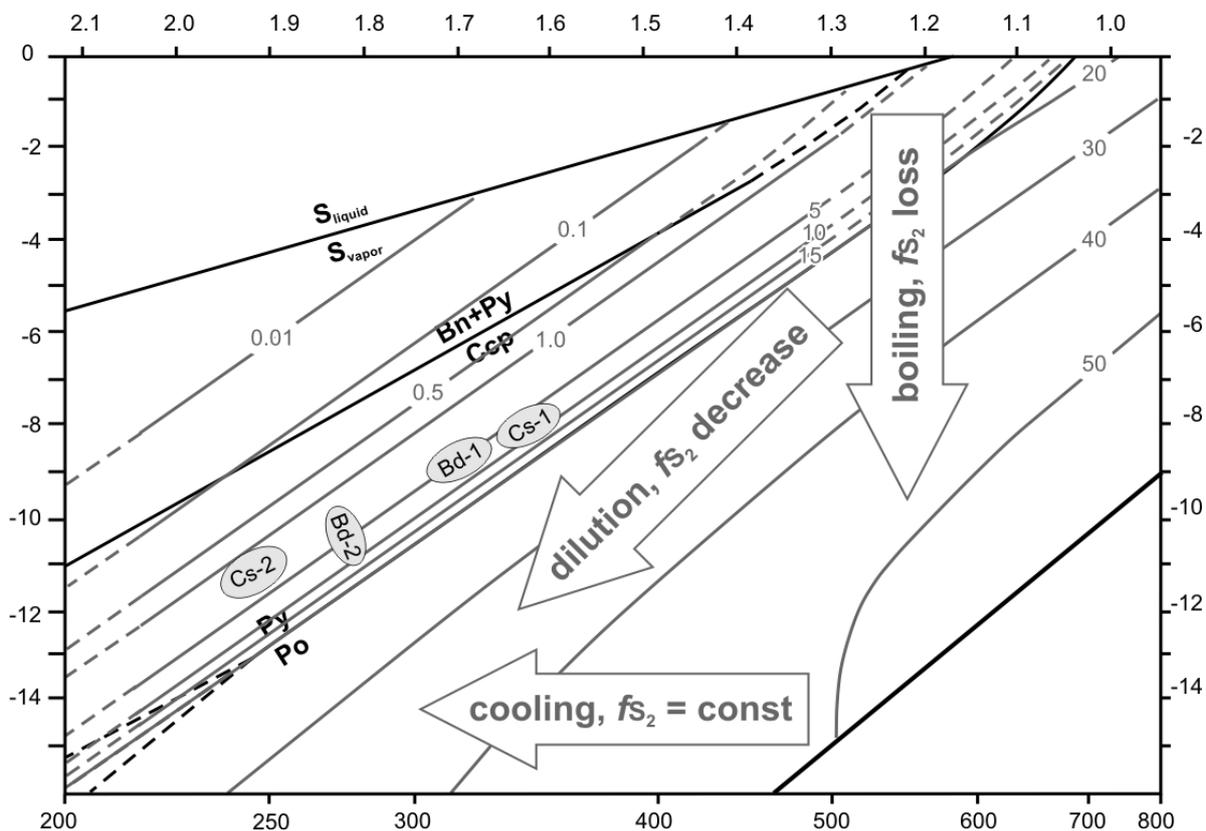


Figure 8. fS_2 -T diagram of the Fe-Cu-S system. X_{FeS} isopleths for pyrite-chalcopyrite stability field are derived from Czamanske (1974) and for pyrrhotite stability field from Scott and Barnes (1971). Cs-1, Cs-2, Bd-1, Bd-2 ovals mark sphalerites of the first and second generations from the Cisma and Bandurita veins, respectively.

seems to be unimportant in epithermal, i.e. shallow environments and thus sphalerite composition can be discussed in $fS_2 - T$ space (Fig. 8). If temperature remains relatively constant fluid boiling will result in a loss of gaseous sulfur, i.e. in fS_2 decrease (Fig. 8). This should lead to an increase of X_{FeS} in sphalerite. On the contrary, in case of temperature decrease from 350 to 150-200°C, as it was estimated for some samples from the Cisma vein, and if fS_2 remains constant, X_{FeS} in sphalerite will slightly decrease. This explains a small X_{FeS} decrease noted in the Cisma vein (from 8 to 3-4 mol.%). However fS_2 can also decrease during cooling because of sulfide deposition or due to dilution by meteoric waters (Fig. 8). A combination of boiling and cooling may lead to relatively small X_{FeS} variations as it was noted for the Bandurita vein.

6. CONCLUSIONS

Sphalerite from the Cisma deposit is characterized by two generations formed in different environments. Sphalerite-1 has a negative correlation between Fe and Mn contents and precipitated in equilibrium conditions. Sphalerite-2 was deposited in non-equilibrium environment; it contains chalcopyrite inclusions and shows positive correlation of Fe and Mn.

Fluid inclusion data point to depositional temperatures of 270 to 316°C in the Bandurita vein and 143 to 344°C in the Cisma vein. The fluids were of low salinity (up to 10 but usually less than 5 wt.% eq. NaCl). Pressure ranges from 50 to 80 bar, i.e. 500 to 800 m paleodepth (hydrostatic).

The various zoning patterns in sphalerite were derived from a combination of boiling, cooling, and dilution of the ore-forming fluid.

7. ACKNOWLEDGEMENTS

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