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TELLURIDES OCCURRENCES IN THE BAIA MARE REGION, ROMANIA

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Abstract: Tellurides are significant minerals in gold deposits both as gold carriers and as indicators of PTx-conditions. In the Inner Carpathians belt tellurides are abundant in most deposits of the golden Quadrilateral (Apuseni Mountains) and sporadic in the Banska Stiavnica zone and the Zlata Bania zone. In the presenr paper we describe for first time altaite and tetradymite occurences in the Baia Mare district proved by e-probe data. Altaite (PbTe) was found in the Cavnic deposit in assemblage with sphalerite, galena, and rhodonite. It's chemical composition is close to stoichiometric: Te 37.61 wt. % and Pb 60.51 wt. %, total 98.12 wt. %. In the Cu-Bi assemblage of the Nistru deposit occurs tetradymite (Bi_2Te_2S) in intergrowth with chalcopyrite, gustavite, and native Au. Tetradymite appears as very small grains with the composition Te 36.99 wt.%, Bi 61.93 wt.% S 4.55 wt.% and calculated formula $Bi_{2.03}Te_{1.99}S_{0.98}$. Paratellurite ($TeO_2$), found in assemblage with native gold and gustavite, forms intergrowth with a 2-microns phase containing Au and Te. Thermodynamic constrains supported by fluid inclusions data indicate that altaite formation at Cavnic was likely to be a result of temperature decrease from 300 to 250-200°C and fS_2 drop. At Nistru tetradymite formation does not require temperature decrease and might be a result of fS_2 drop and/or fTe_2 increase.

Keywords: epithermal, Baia Mare, tellurides, altaite, tetradymite, fahlore, mineral assemblages, fluid inclusions

1. INTRODUCTION

Tellurides, especially those of Au, Ag, Pb, and Bi are typical minerals of epithermal precious and base-metal deposits. Tellurides and sulfotellurides have been reported as rare minerals from most of the deposits in the Inner Carpathians belt: e.g. hessite and tetradymite for the Banska Stiavnica zone and hessite, petzite, and sylvanite for the Zlata Bania zone (Mato et al., 1987). Moreover, tellurides are abundant in most deposits of the Golden Quadrilateral (Ciobanu et al., 2006). Despite this, no tellurides have been found in the Baia Mare district. Although Butucescu et al. (1963) pointed out Au-Ag tellurides (hessite, sylvanite and petzite) as scarce minerals at Nistru and, later, Istvan et al. (1995) mentioned petzite and calaverite at the Suior deposit and non-specified Au-Ag tellurides at the Cavnic-Roata deposit, these observations were made only by optical microscopy and were not confirmed by any analytical equipment. In the present study, we describe two new telluride occurrences within the Baia Mare region – at the Nistru deposit and at the Cavnic-Boldut deposit.

Chemical composition of tellurides and associated minerals were carried out with the Camscan Vega II scanning electron microscope equipped by Oxford Instruments EDX detector (IEM RAS, A. Mukhanova the analyst) and with the Jeol 5600 scanning electron microscope equipped by Link EDX detector (IGEM RAS, A. Mokhov the analyst). Fluid inclusions study were carried out with THMSG-600 "Linkam" thermokriocamera and the Amplival microscope by "Carl Zeiss" (V. Prokofiev the analyst).

2. GENERAL GEOLOGY AND MINERALOGY

Baia Mare ore district is a part of the Inner Carpathians, which represents a subduction-related continental margin arc of Neogene age (Fig. 1). This structure hosts numerous world known Pb-Zn-(Ag) low-sulfidation epithermal ore districts, e.g. Majarovo (Transcarpathians, Ukrania) and in the Banska Stiavnica zone (West Carpathans, Slovakia).

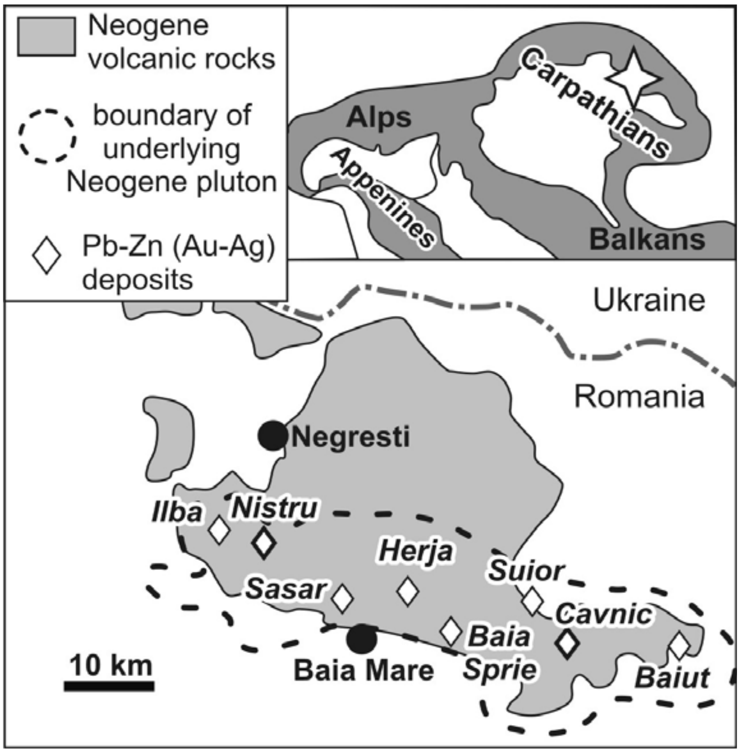

Figure 1. Schematic map of the Baia Mare region showing locations of major ore deposits.

The Baia Mare ore district includes numerous low-sulfidation epithermal gold-polymetalic ore fields, e.g. Nistru, Ilba, Sasar, Herja, Baia Sprie, Suior, Cavnic, Baiut, etc (Borcoş et al., 1972; Lang, 1979; Grancea et al., 2002).

2.1 The Cavnic deposit

The Cavnic deposit is located in the western part of the Baia Mare district and consists of two zones: Cavnic-Bolduț in the North-West and Cavnic Roata in the South-East. Geology and mineralogy of the Cavnic deposits were described in details by Mariaş (1996 and 2005). The deposit is hosted by Neogene volcanic rocks and Paleogene-Miocene sedimentary units intruded by Pannonian (N_1) dioritic bodies. Mineralization occurs in numerous NNE-SSW trending parallel veins that are disposed as 120-350 m distance (Fig. 2). Veins are 200 to 2,500 m long, and with up to 900-m vertical extent. The thickness ranges from 0.5 to 7 m and usually makes up 1m to 2 m (Mariaş, 1996; 2005). Breccias, collomorph, banding, and rugs are most common textures. Hydrothermal alterations consist of extensive argillization with local silicification and pyritisation, (Mariaş 1996; 2005).

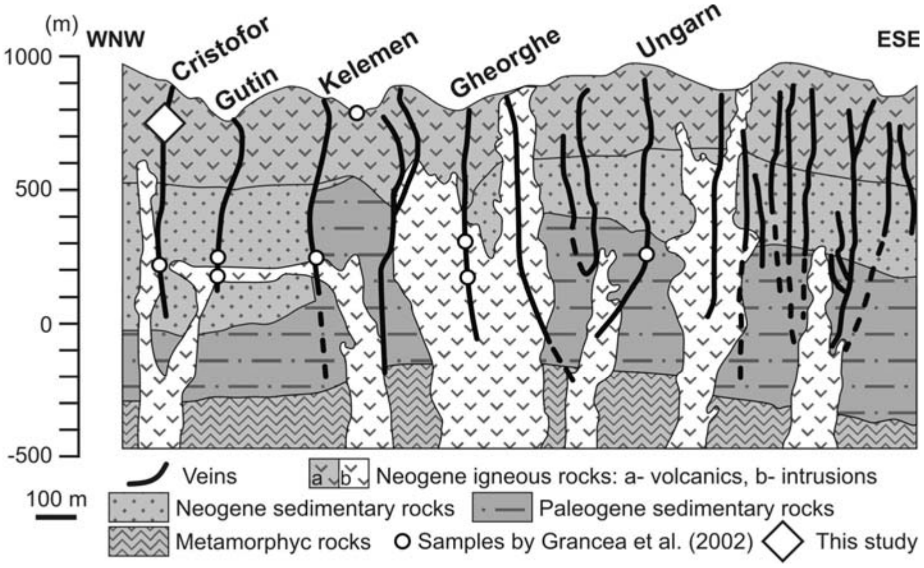

Figure 2. Schematic geological section across the Cavnic-Bolduț zone, modified after (Borcoş et al., 1975; Mariaş, 1996; Grancea et al., 2002).

Mariaş (1996) proposed five mineralization stages for the Cavnic deposit: M0 (pre-ore) − regional propylitic alteration, M1 (Fe-W) − quartz, pyrite, magnetite, scheelite etc., M2 (Cu) − chalcopyrite, pyrite, and covellite, M3 (Pb-Zn-Au) − sphalerite, galena, native gold, quartz, adularia, and M4 (Pb-Zn-Au-Mn-Sb) − rhodonite, rhodochrosite, adularia, galena, sphalerite, tetrahedrite, stibnite, native gold, etc. The mineralization has a base metal character (Pb+Zn±Cu, Au) with a vertical zonality: Au-Ag mineralization with native Au and Ag sulphosalts occur in the upper part of the deposit, base-metal mineralization is more common in the deeper part.

3.2. The Nistru deposit

The Nistru ore field is located in the western end of the Baia Mare ore district (**Fig. 3**). The host rocks at Nistru are represented by Neogene volcanic rocks: pyroclastic rocks with ignimbritic character of Badenian age, pyroxenic andesites of Sarmatian age and quartz andesites of Pannonian age. The Nistru ore field is subdivided into NW and NE-SE ore fields (Fig. 3). The NE-SE ore-field establishes a remarkable horizontal mineral zoning around the stock of quartz-monzodiorites: (1) Cu-Bi-Au mineralization is confined to the central part, within the stock, (2) Pb-Zn mineralization occurs in the periphery, and (3) Au-Ag veins are confined to the margins of the ore field (Damian, 2003). This study was focused on the Bi-Cu-Au mineralization from the central part.

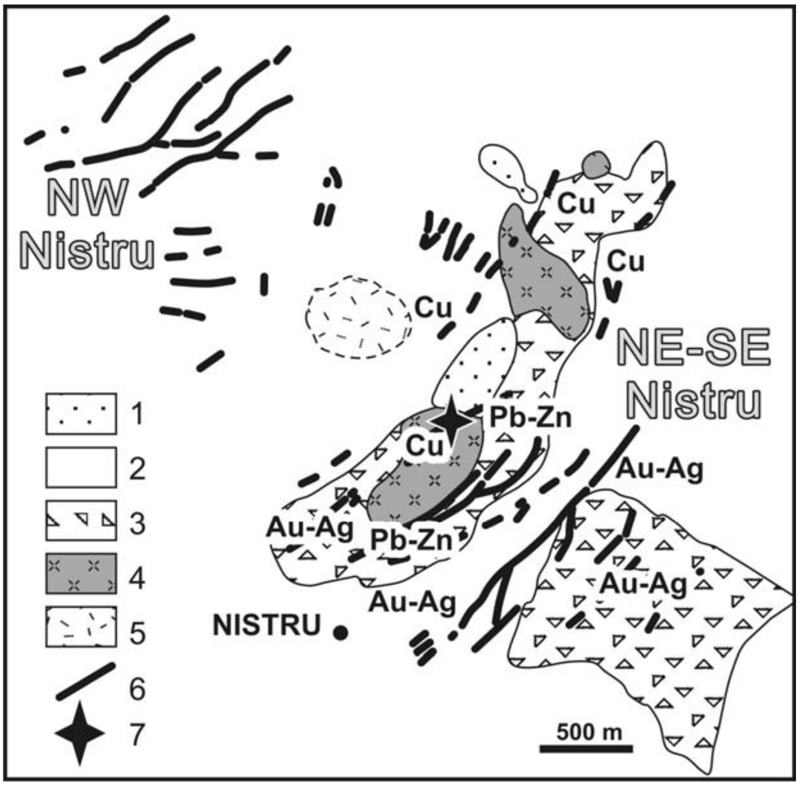

Figure 3. Schematic map the Nistru area, modified after (Borcoş et al., 1972). 1– Quaternary, 2–Pyroxene andesites (Sarmatian age), 3– Pyroclastic rocks (Badenian age), 4– Quartz-monzodiorites, 5– Volcanic neck, 6– Veins, 7– sample point.

Three ore stages were distinguished at the Nistru: (1) Pyrite-copper ± Au and Bi, (2) Base-metal, and (3) Gold ones. The mineralization of Pyrite-copper stage is represented by abundant chalcopyrite, pyrite, minor pyrrhotite, and arsenopyrite. Those are followed by minerals of Au-Bi assemblage whish occurs as lamellas or fill cracks in earlier sulfides or in quartz. Bi mineralization is represented by numerous

Bi±Pb±Cu±Ag sulfosalts. Previous studies (Damian, 1999) revealed minerals of bismuthinite–aikinite (pekoite $CuPbBi_{11}S_{18}$, krupkaite $CuPbBi_3S_6$, gladite $CuPbBi_5S_9$) and lillianite-gustavite ($Pb_3Bi_2S_6$ – $PbAgBi_3S_6$) series, matildite $AgBiS_2$, and cosalite $Pb_2Bi_2S_5$.

4. TELLURIDES ASSEMBLAGES

4.1. The Cavnic deposit

The only telluride found in the investigated samples is altaite (PbTe). The sample comes from the Cavnic-Boldut zone, from the Cristofor vein (+750 level), and represents the M4 stage. Sulfide mineralization in the altaite bearing sample is represented by sphalerite, fahlore, galena, chalcopyrite and pyrite, while gangue minerals are: quartz, rhodonite, ankerite and rhodochrosite. Altaite occurs as small (10-15 µm) grains overgrowing galena, and followed by rhodonite (Fig. 4). Galena, in turn, with tetrahedrite replaces low-iron sphalerite. Rhodonite is followed by ankerite and kutnagorite or rhodochrosite. Thus, Pb telluride occurs directly on board between base-metal and manganese assemblages. Its chemical composition is close to stochiometric: Te 37.61 wt. % and Pb 60.51 wt. %, total 98.12 wt. % (*L* series were measured, synthetic PbTe was used as a standard).

Sphalerite assosiated with altaite has remarcably low iron contents (less then 0.4 wt.% and usually below the detection limit) and high Mn admixtures (up to 2.6 wt.%). Sphalerite from samples where altaite was not found has much higher Fe quantities (0.7 to 1.3 wt. %) and lower contents of Mn − from 0.5 to 1.4 wt. % (Fig. 5), it also contains Cd admixtures from 0.3 to 0.7 wt. %.

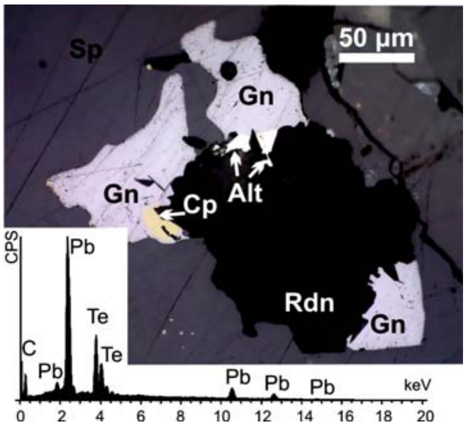

Figure 4. Altaite (Alt), chalcopyrite (Cp), galena (Gn), sphalerite (Sp) and rhodonite (Rdn); reflected light. The inset shows the altaite EDX spectra.

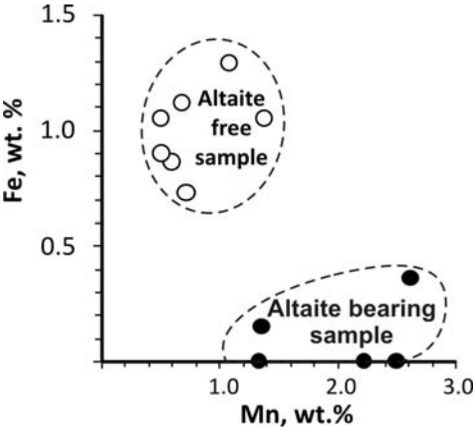

Figure 5. Admixtures in sphalerite associated with altaite and galena (black circles) and one associated with galena only (open circles).

Fahlore group minerals associated with altaite are represented by three generations varying in mineral assemblages as well as in chemical compositions (Figs.

6 and 7). The first one forms ultra thin veinlets that crosscut sphalerite. It is tennantite with variable contents of Zn and Fe and average composition $(Cu_{9.16}Ag_{0.06})_{9.22}(Fe_{0.96}Zn_{1.04}Cd_{0.03})_{2.03}(Sb_{0.59}As_{3.46})_{4.05}S_{12.94}$. The second generation is tetrahedrite with an average calculated formula $(Cu_{9.80}Ag_{0.07})_{9.87}(Fe_{0.11}Zn_{1.72})_{1.82}(Sb_{3.06}As_{1.12})_{4.18}S_{13.13}$ which forms eutectic-like intergrowths with galena forming a band on the margin of sphalerite grains. Fahlore of the third generation is close to the second one in chemical composition $[(Cu_{9.58}Ag_{0.12})_{9.69}(Fe_{0.17}Zn_{1.86})_{2.03}(Sb_{3.53}As_{0.58})_{4.11}S_{12.96}]$ but has slightly higher Sb contents and overgrowths sphalerite (Fig. 6).

Although, it's difficult to determine which generation of fahlores is in equilibrium with altaite it could be supposed that a dramatic change from tennantite to tetrahedrite is consistent to the change from galena to altaite. Thus, altaite bearing assemblages are marked by almost Fe free sphalerite as well as by high variability of fahlore group minerals.

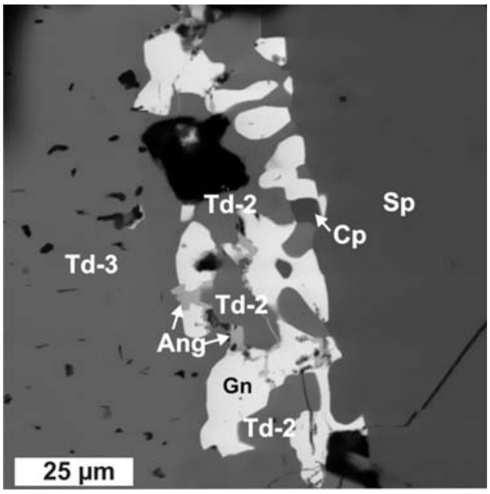

Figure 6. Fahlore 2 (Td-2) in intergrowth with galena (Gn), anglesite (Ang), chalcopyrite (Cp), and Fahlore 3 (Td-3), Sp – sphalerite; SEI image.

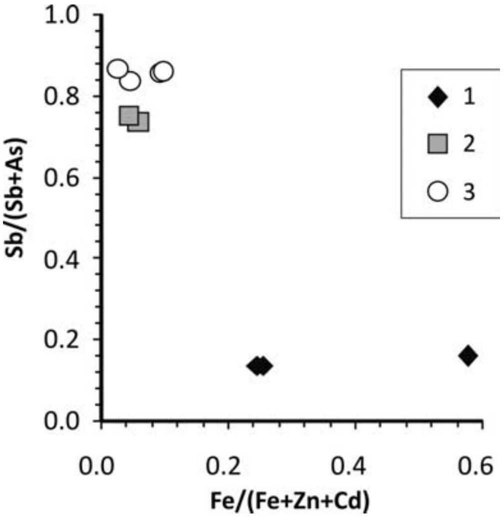

Figure 7. Chemical composition of fahlore group minerals, numbers correspond to generations, see text for explanations.

4.1. The Nistru deposit

Tetradymite Bi_2Te_2S occurs as small (usually 2 to 5 µm) inclusions in chalcopyrite and forms intimate intergrowths with native gold and matildite (Fig. 8). It's composition is difficult to determine dew to very small grains and permanent matrix captures but in the largest grain where the matrix influence was removed the composition is Te (L_α) 36.99 wt.%, Bi (M_α) 61.93 wt.% S (K_α) 4.55 wt.% and calculated formula $Bi_{2.03}Te_{1.99}S_{0.98}$.

In assemblage with native gold and gustavite was found **paratellurite** (TeO_2) which forms intergrowth with a 2-microns phase containing Au and Te (Fig. 9). The phase has the following composition: Au (M_α) 40.23 to 10.15 wt. %, Ag (L_α) 3.98 to

1.76 wt. %, Te (L_α) 38.23 to 65.73 wt.% and O (K_α) 19.75 to 22.08 wt. %. It is supposed to be **paratellurite** with small inclusions of native gold and such an assemblage can be interpreted as oxidized **calaverite** $AuTe_2$ or low-Ag **krennerite** $(Au,Ag)Te_2$.

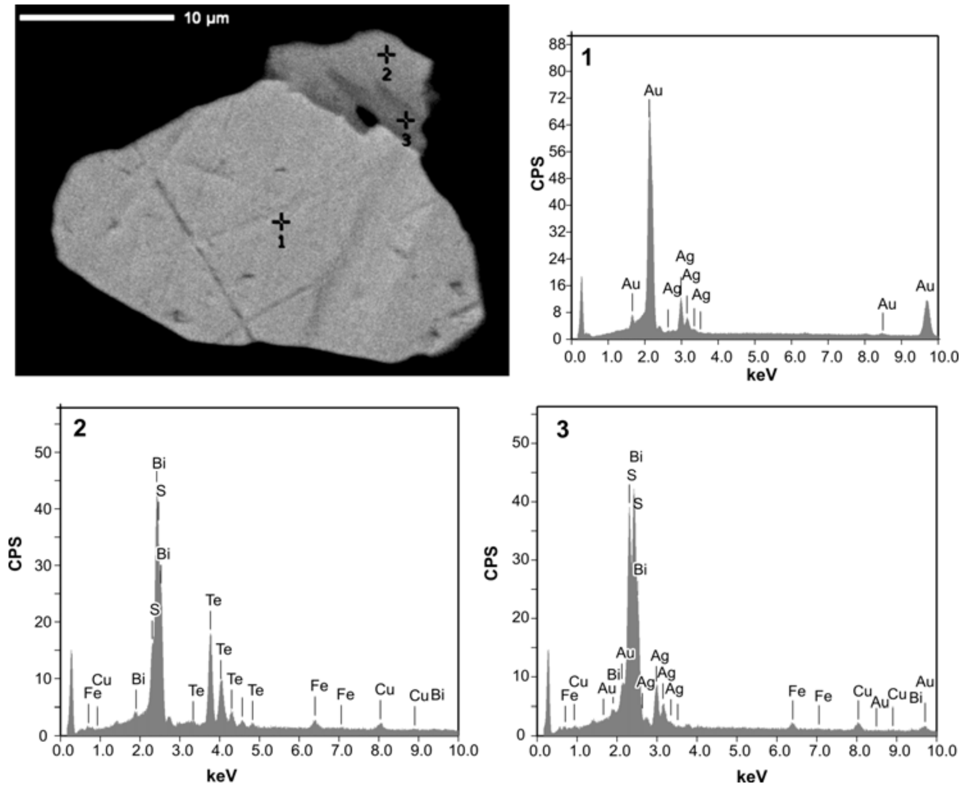

Figure 8. Native gold Au_2Ag (1), tetradymite (2), and matildite (3) in chalcopyrite (black), SEI image and EDX spectra.

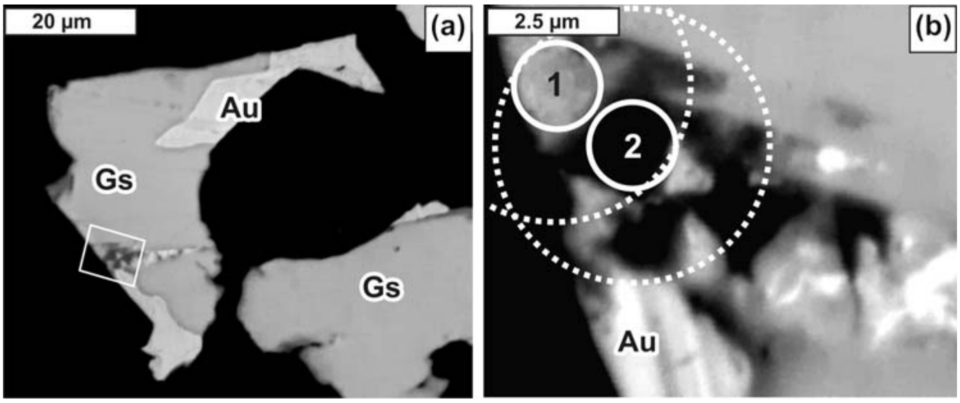

Figure 9. (a) − gustavite (Gs) and native Au; (b)- enlarged part of (a): supposed oxidized krennerite (1) and paratellurite (2), SEI. Solid circles show the beam diameter, dotted circles mark the measured zones.

Native gold occurs as small lamellas mostly in chalcopyrite and pyrite as well as in quartz and forms intimate intergrowth with Bi minerals. In analyzed samples it shows remarkable narrow range of Ag admixture (from 20.46 to 23.35, average 22.37 for n=7) and its composition is closed to stoichiometric Au_2Ag. The Ag contents are lower than those in native gold from telluride-rich gold deposits from the Apuseni Mountains, (Neacşu et al., 2009).

Sphalerite is the main mineral of the base metal stage. It has relatively high Fe contents (6 to 16 wt.%) and contains chalcopyrite emulsion along growth zones. Subordinate minerals are **galena** and **freibergite** with 16.5 wt. % of Ag and calculated formula $(Cu_{6.7}Ag_{2.7})_{9.4}(Fe_{0.9}Zn_{1.3})_{2.2}Sb_{4.0}S_{13.2}$.

5. TELLURIDES FORMATION CONDITIONS

5.1. The Cavnic deposit

The most representative fluid inclusion study for the Cavnic deposit was performed by Grancea et al., (2002). They estimated temperatures of M0 and M1 stages to be as much as 290-316°C, M2 stage − from 260 to 294°C, M3 stage 225 to 240°C, and M4 stage from 214 to 290°C (only primary fluid inclusions data are listed here). The lowest temperature of 214° was reported for carbonate. Since Grancea et al. (2002) did not investigate M4 stage mineralization from the Cristofor vein, extra fluid inclusion study has been carried out. Results of fluid inclusion study in quartz hosting sulfide mineralisation of M4 stage (Table 1; Fig. 10), in general, show a good agreement with the studies of Grancea et al. (2002). Small increase in salinity with temperature decrease (Fig. 10) is a typical trend for fluid boiling. Dilution trend (salinity decrease with temperature drop) also takes place. The eutectic temperature range from -37 to -29°C evidence the presence of both Na-K and Mg and/or Fe chlorides (Borisenko et al., 1979). Boiling indicates homogenisation temperatures were the real temperatures of mineral deposition. Pressure 60−70 bar points to the depth of ore formation as less than 600 m if the environment was hydrostatic.

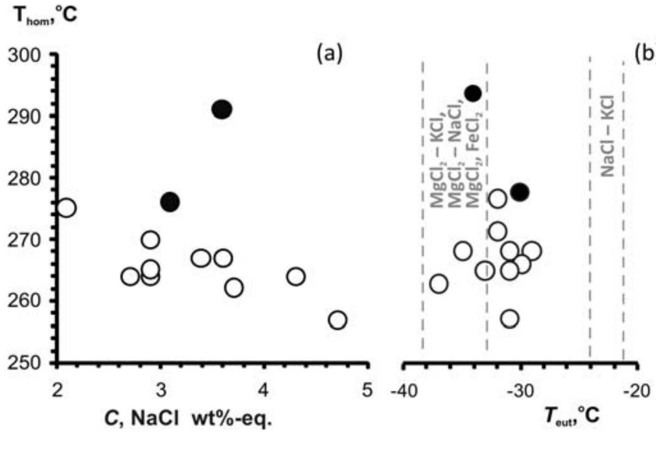

Figure 10. Homogenization temperature via salinity (a) and eutectic temperature (b) diagrams for M-4 stage quartz, of the Cristofor vein, the Cavnic-Boldut deposit; black circles mark boiling.

Table 1. Fluid inclusion data for M-4 stage quartz; n/d = not detected; * marks boiling

№	sample	n	T_{hom}, °C	T_{eut}, °C	$T_{ice\ melt.}$, °C	C, wt. % eq. NaCl	d, g/cm^3	P, bar
1	Cav 220g	15*	276	-30	-1.8	3.1	0.78	60
2	<<	6*	291	-34	-2.1	3.6	0.76	70
3	Cav 233	3	267	-29	-2.1	3.6		-
4	<<	4	267	-31	-2.0	3.4		-
5	<<	2	270	-32	-1.7	2.9		-
6	<<	3	264	-33	-1.7	2.9		-
7	<<	4	265	-30	-1.7	2.9		-
8	<<	3	264	-33	-1.6	2.7		-
9	<<	3	275	-32	-1.2	2.1		-
10	<<	11	264	-31	-2.6	4.3		-
11	<<	3	262	-37	-2.2	3.7		-
12	<<	7	267	-35	-2.1	3.6		-
13	<<	8	257	-31	-2.8	4.7		-
Grancea et al., 2002			214...290	n/d	-3.0...-0.9	1.6...5.0	n/d	n/d

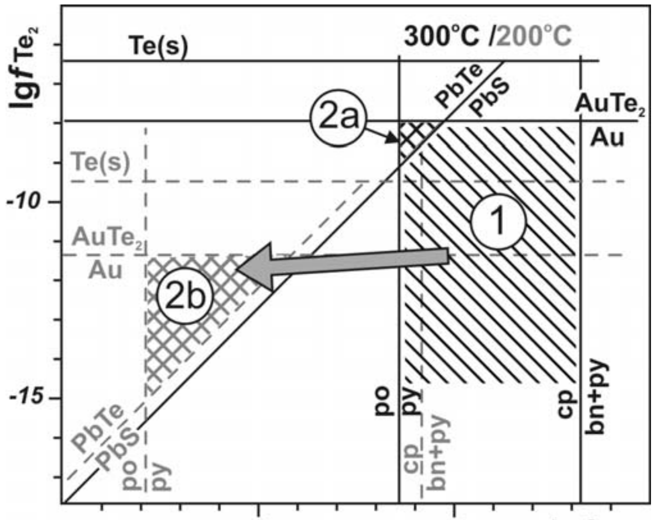

Figure 11. $fS_2 - fTe_2$ diagram for 300°C (solid lines) and 200°C (dashed lines) for galena and altaite stabilities. Assemblages: 1 – galena, pyrite, chalcopyrite, native gold at 300°C, 2a – altaite, pyrite, chalcopyrite, native gold at 300°C, 2b – the same as 2a at 200°C.

Altaite and galena stability limits were calculated in fS_2-fTe_2 space using data from (Barton, Skinner, 1979) and (Afifi et al., 1988). It was assumed that all the mineral assemblages of M-4 stage were formed: (1) within pyrite-chalcopyrite stability space and (2) within native gold but not gold tellurides stability, and (3) that minerals deposition started at maximum 300°C with sulfides formation and continued till about 200°C with carbonates precipitation. As shown on figure 11, the position of the altaite/galena monovariant line is not much reflected by temperature. However, if temperature 300°C remains constant and galena replacement by altaite is a function of $f\mathrm{S}_2$ decrease/$f\mathrm{Te}_2$ increase, only altaite stability occupies a very small field (2a on Fig. 11) and thus altaite deposition seems unlikely. In contrast, if temperature drop till 250−200°C takes place (e.g. as a result of boiling) pyrite-chalcopyrite stability will require much lover $f\mathrm{S}_2$ and that enlarges altaite formation limits remarkably (2b on Fig. 11). S_2 fugacity decrease in turn can be a result of sulfur loss during boiling and subsequent mass sulfides deposition. Thus, Pb telluride formation at Cavnic occurred due to fluid boiling which resulted in temperature decrease and dramatic $f\mathrm{S}_2$ drop.

5.2. The Nistru deposit

Little has been published on formation conditions at the Nistru deposit. Fluid inclusions data presented in Borcoş et al., (1974) reported deposition temperatures of more than 325°C for the pyrite-copper stage, 325 to 210°C for the base-metal stage and 160 to 250°C for the gold stage. Fluid inclusions data obtained by Damian (1999) indicated deposition temperature of 230-240º C for copper vein with Bi-minerals.

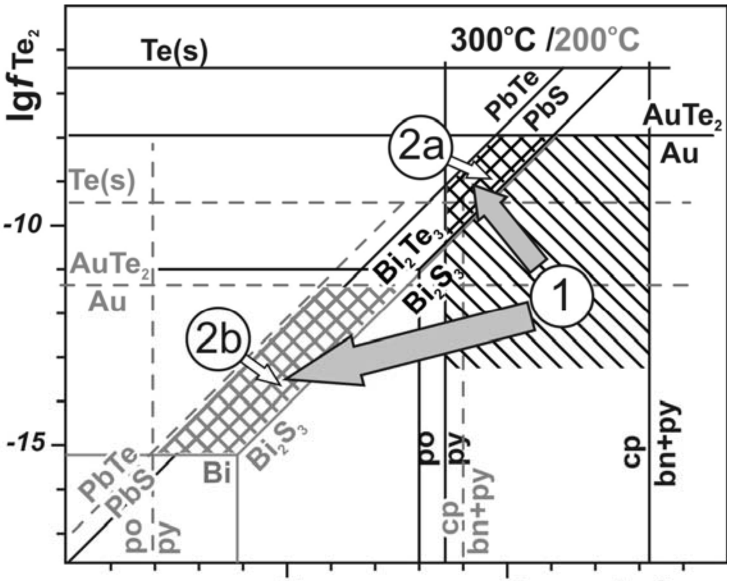

Figure 12. $f\mathrm{S}_2 - f\mathrm{Te}_2$ diagram for 300°C (solid lines) and 200°C (dashed lines) for galena and bismuth minerals stabilities. Stability fields: 1 – bismuthinite and other Bi sulfosalts at 300°C, 2a – tellurobismuthite (and, presumably, tetradymite) at 300°C, 2b – the same as 2b at 200°C.

The absence of thermodynamic data for tetradymite as well as for most of Bi sulfosalts reported at Nistru embarrasses to determine their formation limits but we can assume that tetradymite could have formed from bismithinite according to the reaction:

$Bi_2S_3 + Te_2 = Bi_2Te_2S + S_2$ (1)

or vice versa from tellurobismuthite:

$Bi_2Te_3 + ½ S_2 = Bi_2Te_2S + ½ Te_2$ (2)

i.e. tetradymite can be formed at small fluctuations in Te_2 or S_2 fugacities in the system and that it's stability is close to those of tellurobismuthite.

Assuming an average temperature of the pyrite-copper stage as 300°C and that mineral deposition took place within pyrite-chalcopyrite stability limits and that galena but not altaite is stable we can see that tellurobismuthite (and, presumably, tetradymite) field forms a narrow band (2a on Fig. 12). At temperature as much as 200°C tellurobismuthite stability field is only slightly larger (2b on Fig. 12) and that explains rare occurrence of tetradymite at the Nistru deposit.

6. CONCLUSIONS

The data obtained in our study shows that tellurides (especially those of Bi and Pb and, perhaps, of Au or Ag) are typical for epithermal Au-Ag base-metal deposits of the Carpathians and therefore, tellurides can be found at other deposits of the Baia Mare district. The relative abundance of tellurides compared to sulfides is a problem of future research.

7. ACKNOWLEDGEMENTS

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