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Original scientific paper

SOME IMPORTANT CHARACTERISTICS OF POLYMETALLIC LEAD-ZINC ORE IN HAJVALIA MINE, KOSOVO

Bedri Durmishaj[1], Sylejman Hyseni[1], Albert Hyseni[2]

[1]*University "Isa Boletini", Faculty of Geosciences, Mitrovicë, Republic of Kosovo*
[2]*Faculty of Natural and Technical Sciences, "Goce Delčev" University in Štip, Blvd. "Goce Delčev" 89, P. O. Box 201, 2000 Štip, North Macedonia*
sylejman.hyseni@umib.net

A b s t r a c t: In this paper are presented general geological data for the polymetallic Hajvalia deposit. In particular, we have presented the scientific results regarding the chemical and mineralogical composition of sulfur mineralizations from the Hajvalia deposit. The distribution of the main metals used during the mining activity in this deposit has been studied with special emphasis. From the microscopic description of the mineral samples in the Hajvalia deposit, the following mineral compositions result: sphalerite, galena, pyrite, chalcopyrite, marcasite, arsenopyrite, pyrrhotite, tenanntite, as well as non-metallic minerals. The geochemical associations indicate the strong connection of Pb with Zn and Ag, and less with Cd and Cu. There is another association of Sb with Cd in antagonism with Cu, and an association of Bi with Cu in antagonism with As. Electron probe analysis shows that galena is quite pure with sporadic high content of Ag (up to 100 ppm). The Fe content in the sphalerite is quite homogeneous and varies from 9 to 11.5%, or 0.18 Fe atoms in the crystallochemical formula. As is known, the Fe content in sphalerite depends on the temperature of the mineral formation and, in some cases, can be used as a geothermometer. The main characteristic of galenites is their "purity", with a very high content of Pb. Ag stands out among the other elements, as does the presence of Hg, more Sb than Bi. The characteristic is the presence of Te and Se in isomorphism with S, always with a predominance of Te. In contrast to galena, the contents of Ag as well as Se and Te are lower while it is found more often in Bi than Sb.

Key words: polymetallic sulphide; lead-zinc ore; geochemical association; Hajvalia mine

INTRODUCTION

Hajvalia mine is located around 12 km southeast of Prishtina (capital city of the Republic of Kosova), and it is part of the Hajvali-Badovc-Kizhnic ore field (Figure 1). From a metallogenic point of view, the mineralization of the Hajvalia deposit is genetically and timely related to Tertiary age andesites [8].

The Hajvalia ore deposit is one of the richest polymetallic deposits in Europe and is particularly distinguished from other deposits in this region for its high content of zinc (Zn) metal. Are evaluated on 5 Mt ore reserves with average content of main metals of 12.20% Zn, 6.50% Pb and 72 g/t Ag [5, 16, 19]. The main associated elements with zinc (Zn) in this mineral deposit are lead (Pb) and silver (Ag), while other elements such as Cd, Bi, Au, etc., can be found in smaller quantities. The geochemical data presented in this manuscript will help to a real interpretation of these geological and mining data. The study is based on the data of the Hajvalia ore deposit, taken during the years of exploitation.

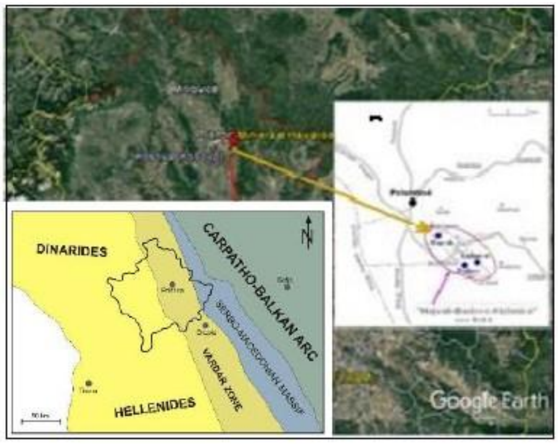


Fig. 1. Geographical position of the Hajvalia mine (a) within the Vardar zone (SZV) and (b) within the Hajvalia-Badovc-Kizhnica ore field [11]

REGIONAL GEOLOGY

The Hajvali-Badovc-Kizhnica Pb-Zn-Ag deposits are located at the southern end of the so-called zone II of the NNW-SSE-trending Vardar zone [14], a zone of Tertiary activity [8], and continental plate [7]. From the structural-tectonic point of view, this area belongs to the metallogenic region of Kopaonik [13] mineral belt Trepča [10] (Figures 2a, 2b).

Fig. 2a. Location of the Trepča mineral belt within Vardar tectonic zone [10]

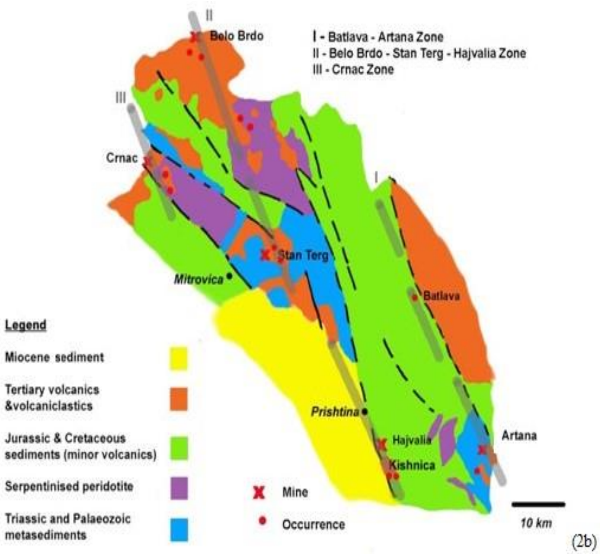


Fig. 2b. Hajvali-Badovc-Kizhnica and Artana ore fields, and other [11]

The deep tectonic fracture II (Belo Brdo-Stan Tërg-Hajvali-Kizhnica zone, Figure 2b), in the region of the Hajvali-Kizhnica, is divided into three structural-tectonic units of the lowest order: the Hajvali-Badovc (1); Kizhnica (2); and "Okosnica" (3). All these fracture zones are factors that control the location of the lead-zinc mineralization in the deposits (Figure 3).

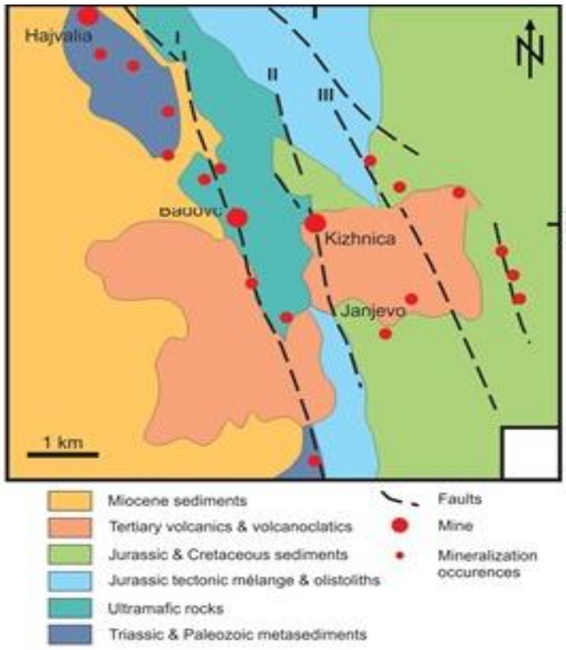


Fig 3. The Hajvalia-Badovc-Kizhnica ore field [9]

The main geologic subunits in the region of Hajvali-Kizhnica are the metamorphic series (Paleozoic age). The metamorphic series includes crystalline schists, carbonates (limestone, marbles, and crystalline dolomites), and quartz rocks, which are present in the Hajvali and Badovc deposits but not in the Kizhnica deposit. Jurassic sedimentary and magmatic formations are composed of sedimentary-terrigenous rocks, whereas the magmatic rocks, mostly serpentinites, are rare. The carbonates are the main ore-bearing rocks in the Hajvali mineral deposit, but to a certain extent also in the Badovc deposit. Cretaceous sedimentary depositions (conglomerate, sandstones, clays, marl and limestone sub-layers) are found in all three mineral deposits but are more spread in the mine of Kizhnica, where scarce mineralization of lead (Pb) and zinc (Zn) are located in them.

Tertiary sedimentary breccia, conglomerates, sandstones, marl, and mostly clays (Paleogen age) are present around the mines of Badovc and

Kizhnica, but they are missing in the mineral deposit of Hajvali, and Neogen volcanic rocks (lava flows, and pyroclastic rocks). The volcanic of the Neogen rocks are believed to be the source of hydrothermal fluids that produced polymetallic mineralization in this region [18], Quaternary depositions.

GEOLOGY OF THE HAJVALIA MINE

The zinc-lead-silver sulphide deposit of Hajvalia is the carbonate replacement type, and occurs as lenses within a series of Paleozoic schists with intercalated carbonate layers, which host the main mineralization.

Structurally, the ore bodies occur at the hinges of folded carbonates and schist within an anticline structure. The mineralized lenses can be followed along a strike length of 300 m and down-dip extensions of up to 900 m (open at depth). The host carbonates occur as beds and lenses within schist and have been intensely deformed. The mineralization is particularly well developed in the hings of the folded carbonates within an anticline plunging approximately 200 SE which is bounded by the western fault (marked by breccias and possibly a feed for hydrothermally altered (silification and chloritization) adjacent to the mineralization (Figures 4, 5). The sulphide paragenesis includes sphalerite, galena, and pyrrhotite with minor chalcopyrite.

Fe-Mn carbonates of the skarn-type association (oligonite) with only minor sulphides occurring peripheral to the mine ore zones. Examinations in the Hajvalia lead-zinc mineral deposit revealed the presence of cemented gold [22].

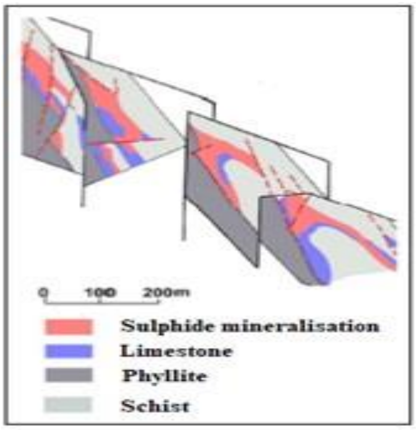


Fig 4. Simplified section, Hajvalia mine [9]

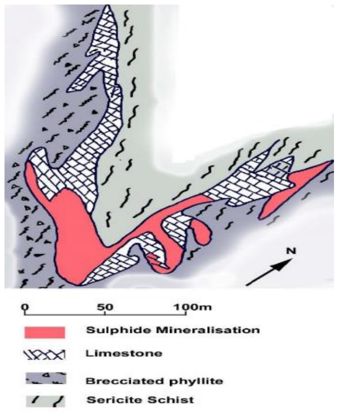


Fig 5. Detail from level plan showing metasomatic replacement of carbonate on fold hinge, Hajvalia mine [9]

CHEMICAL COMPOSITION AND MAIN GEOCHEMICAL ASSOCIATIONS IN THE HAJVALIA DEPOSIT

Regarding the chemical composition of the mineralizations in the deposit, we have used the analysis of the ore minerals deposited for processing in the Kizhnica factory/enrichment plant (flotation), as well as the analysis of the samples taken from the ore bodies in the Hajvalia deposit.

The distribution of the contents of main chemical elements and accompanying chemical elements in the deposit is shown through statistical parameters in Table 1. From this table, the content of Pb and Zn main metals is high, with a predominance of Zn versus Pb (14.41% versus 9.35%) and that of Ag at about 108 g/t. Whereas in Table 2 we present the geochemical associations using correlative analysis.

According to the correlative analysis, the results show these geochemical associations: Pb-Zn-Ag; Zn-Ag-Cd; Sb-Bi. Whereas data on geochemical associations according to another method known as factor analysis from [5]. result in: Pb-Zn-Ag-(Cd); Sb-(Cd); Bi-(Cu).

In the following, we present the results on the chemical composition of sulfur mineralizations at the Hajvalia deposit (Table 3), as well as the diagram of the result of lead and silver content for 30 analyzed samples from [12].

From Table 3 we constructed the diagram in Figure 6, from which the statistical data suggest a very strong correlation of silver with lead, which can be related to both the association of pyrargirite with galena as well as the isomorphic enrichment of galena with silver.

Table 1

Statistical parameters of the distribution of metal contents in the Hajvalia deposit [5]

Parameters\Elements	Pb %	Zn %	Ag g/t	Bi %	Cd %	Cu %	As %	Sb %
Average	9.35	14.41	107.90	0.02	0.05	0.06	0.09	0.15
Median	10.25	14.02	78.00	0.02	0.05	0.06	0.06	0.13
Standard deviation	5.48	4.84	64.01	0.01	0.02	0.01	0.06	0.08
Minimum	4.68	5.38	24.00	0.01	0.02	0.05	0.03	0.05
Maximum	24.34	21.06	204.00	0.06	0.10	0.10	0.21	0.29
No. of samples	14	14	14	14	14	14	14	11

Table 2

Correlation matrix, Hajvalia deposit. Correlation coefficients are significant for ρ <0.0500 when they have a value of > 0.52. Number of samples = 14 [5]

Elements	Pb %	Zn %	Ag g/t	Bi %	Cd %	Cu %	As %	Sb %
Pb %	1							
Zn %	**0.653215**	1						
Ag g/t	**0.671419**	**0.659889**	1					
Bi %	0.060286	0.24774	0.08904	1				
Cd %	0.448503	**0.598125**	0.407301	0.03885	1			
Cu %	0.192734	0.33962	0.405992	0.347134	0.18641	1		
As %	0.27762	0.34705	0.19854	0.15269	0.30433	0.12497	1	
Sb %	0.37315	0.1202	0.28466	**0.526416**	0.196047	0.278879	0.042862	1

Table 3

Average chemical composition of minerals in the studied ore bodies, according to the representative mining, and in the enrichment plant Kizhnica [5]

Elements	Pb %	Zn %	Ag g/t	Au g/t	Cu %	Bi %	Sn %	As %	Cd %	Sb %
*EP	5.5	8.70	60	0.5	0.05	0.016	0.014	0.026	0.090	trace
*EOBD	9.35	14.41	107.90	NA	0.06	0.02	NA	0.06	0.098	0.15
Nonmetals	SiO_2 %	MgO %	MnO %	CaO %	Zn/Pb	Ag/Au	MgO/CaO			
*EP	24.60	0.87	5.18	1.23	1.57	120	0.71			

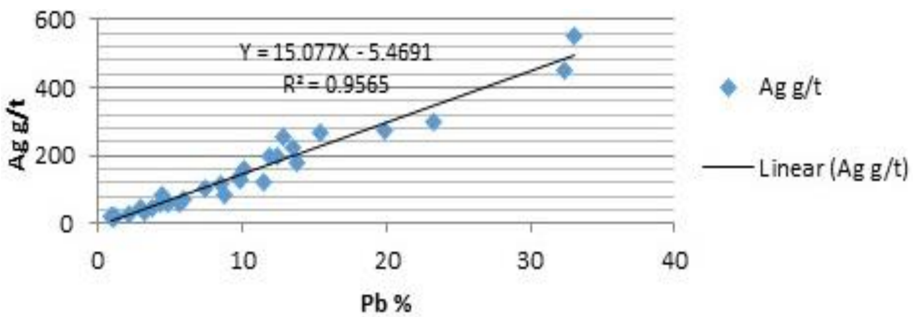
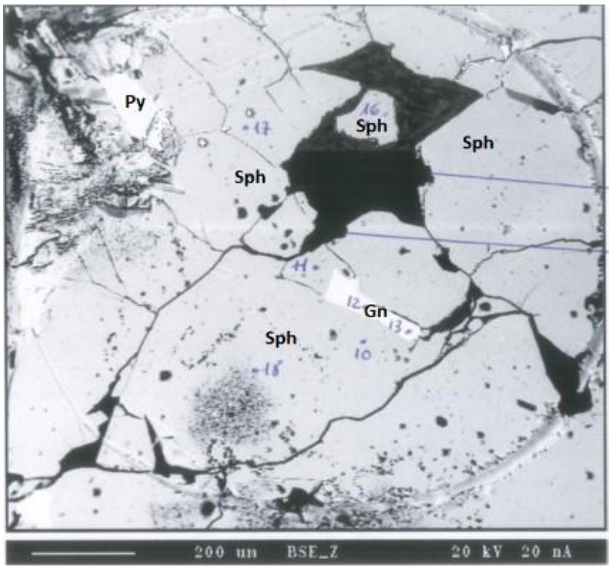


Fig. 6. Diagram of the correlation of silver contents with lead contents [5]

MICROSCOPIC STUDY AND THE RESULTS OF CHEMICAL ANALYSIS OF THE MAIN MINERALS IN THE HAJVALIA DEPOSIT

From this deposit, four samples were analyzed, performing a total of 86 chemical analyses in different minerals. SX50 electron microprobe analyses were performed at the BRGM laboratory in Orleans, France. The working conditions for the quantitative determination of chemical elements were: kosecande of the capture angle is 1.556, the acceleration voltage 20 kV, the current 30 nA, and the counting time 10″.

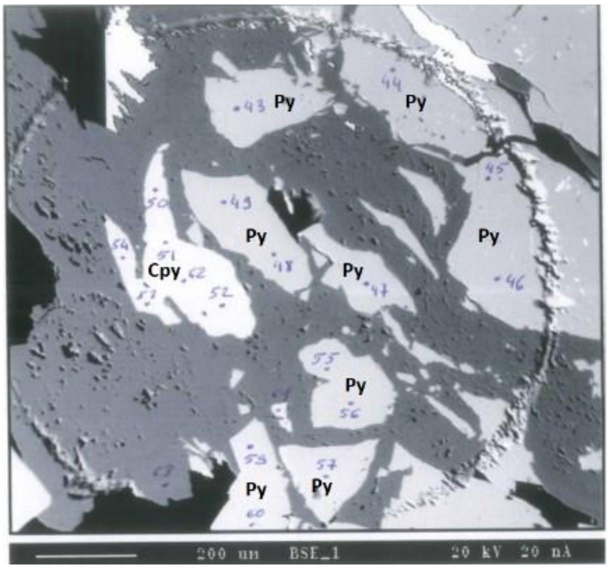
From the microscopic description of the preparates/polished section (H2, H3, H5, and H6) that result in this mineralogical composition: H2-polished section, results to have 20% sphalerite composition, 5% galena, then pyrite, marcasite, tenanntite, and traces of chalcopyrite and arsenopyrite, as well as pyrrhotite transformed into marcasite and pyrite. Iron sulfides occupy the majority part of the polished section about 60% (Microphoto 1). The marcasite does not have a definite orientation, as we saw on the polished section H6. The sphalerite is with drops of chalcopyrite, maybe even with very fine tenanntite.



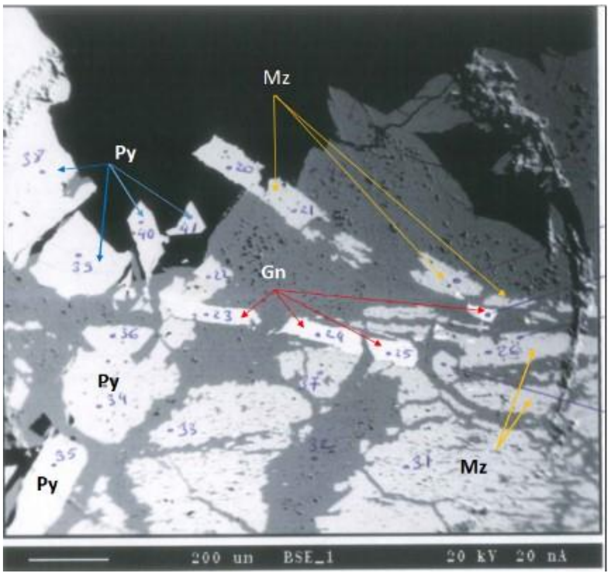
Microphoto 1. Mineral composition of H2-polished section, Hajvalia deposit, Kosovo [5]

H3-polished section consists of the following minerals: pyrite 40%, marcasite 20%, galena and chalcopyrite 1%, as well as non-metallic minerals (Microphotos 2 and 3).

Pyrite forms: relatively large crystals with dimensions ranging from millimeters up to 1 cm. Its crystals are well grown with a cocardine texture. They are often described as having cracks and fractures filled with non-metallic minerals. We also find microcrystalline pyrite mixed with marcasite and that for melnikovite. These formations are described and replaced in their lateral parts with microcrystalline marcasite



Microphoto 2. Mineral composition of H3-polished section, Hajvalia deposit, Kosovo [5]



Microphoto 3. Mineral composition of H3-polished section, Hajvalia deposit, Kosovo [5]

Marcasite meets in two morphological forms: fine microcrystalline marcasite and crystals with a size of hundreds of millimeters, while in another form, plate marcasite measuring four to tenths of a millimeter. Fine crystal marcasite generally forms pseudomorphs according to an elongated crystal that appears to have been pyrrhotite. While large crystals of plate marcasite appear to be genuine formations of marcasite. Starting from microcrystalline marcasite accumulations, we can say that the pyrrhotite crystals were of centimeter scale, and no marcasite inclusions were observed between the pyrite macrocrystals, which indicates a relatively high pH (above 5) of the initial crystallization environment and maybe at a high temperature. Apparently, the granular pyrite will have crystallized simultaneously with the initial pyrrhotite.

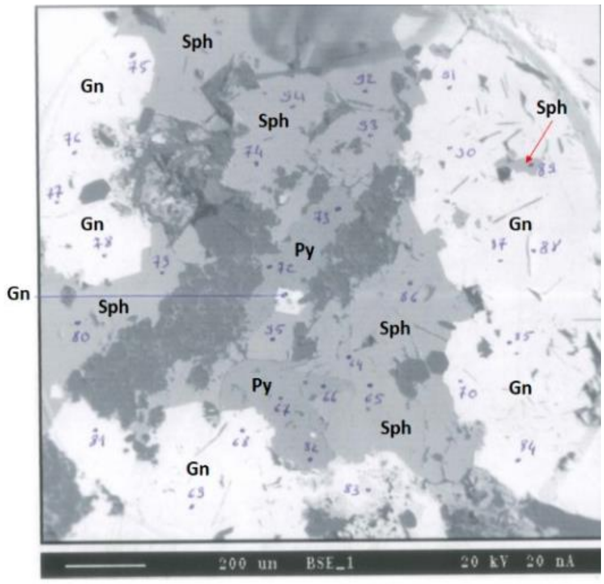
Chalcopyrite. This mineral is generally found outside pyrite macrocrystals and is generally surrounded by non-metallic minerals. Chalcopyrite forms elongated clusters up to 0.5 mm in an irregular shape. It appears that this mineral was formed together with the non-metallic minerals behind the pyrite macrocrystals. Even in some cases, when chalcopyrite is in contact with pyrite, it seems as if you are wearing it. The fact that chalcopyrite is found mainly between the non-metallic minerals that interrupt and surround the pyrite suggests that we are dealing with a recycling of copper and its re-deposition together with quartz and other non-metals.

Galena. This mineral occurs both in the form of elongated formations enclosed in macrocrystalline pyrite or in isometric junctions between them and in pseudomorphoses according to elongated crystals that may have been pyrrhotite. So, on the whole, we seem to have two moments of crystallization: The first is mainly iron sulfides, pyrite-pyrrhotite with quartz and with traces of galena, and under two: pyrite-marcasite with pseudomorphoses of galena according to pyrite and marcasite. Galena may have formed pseudomorphoses even according to pyrrhotite, taking into consideration its elongated forms.

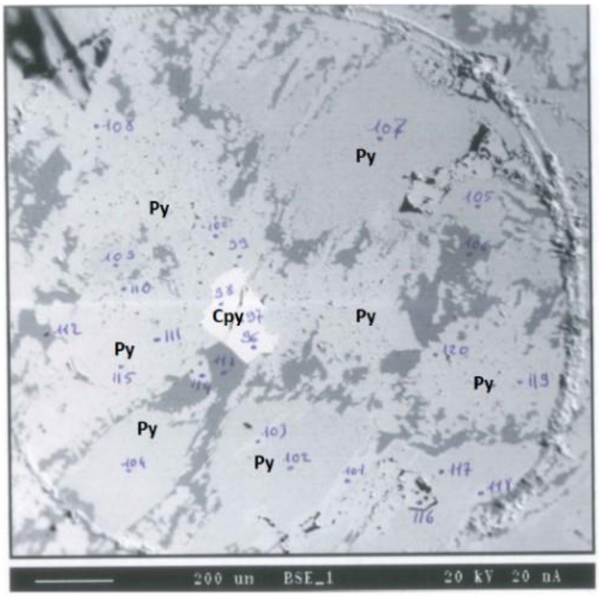
H5-polished section consists of 30% sphalerite, 25% galena, chalcopyrite, and a trace of pyrite, as well as non-metallic minerals (Microphoto 4). Sphalerite has more, and with micro-inclusions of chalcopyrite, probably also of tenanntite that is surrounded by galena.

H6-polished section consists of the following minerals: 50% pyrrhotite, 40% marcasite, traces of pyrite, chalcopyrite, and sphalerite, as well as non-metallic minerals (Microphoto 5). In this anschlife, we have pyrrhotite broken down in exogenous conditions with typical "bird eye" textures.

Typical is the orientation of the marcasite crystals, which is dictated by the orientation of the crystallographic planes of any other mineral (pyrrhotite with more in our concrete case). The characteristic color of marcasite is a fairly strong anisotropy, with a characteristic color range from blue to dark brown. The orientation of the marcasite is seen according to three directions that formed an angle of approximately 60º, as a result of the existence of structural planes of an earlier mineral, which most likely was pyrrhotite



Microphoto 4. Mineral composition of H5-polished section, Hajvalia deposit, Kosovo [5]



Microphoto 5. Mineral composition of H6-polished section, Hajvalia deposit, Kosovo [5]

T a b l e 4

Crystallochemical formulas of major minerals according to electron microprobe analyses, orebodies of the Hajvalia deposit, Kosovo [5]

Polished section No.	Crystallochemical formulas	Mineral
1	$Pb_{1.003}(S_{0.996}, Te_{0.001})_{0.0997}$	Galena
5	$(Pb_{0.994}, Sb_{0.002}, Ag_{0.0005}, Hg_{0.0005})_{0.997}S_{1.003}$	
	$(Pb_{0.993}, Fe_{0.003}, Sb_{0.002})_{0.998}S_{1.002}$	
2	$(Zn_{0.802}, Fe_{0.180}, Co_{0.001})_{0.983}S_{1.017}$	Sphalerite
5	$(Zn_{0.806}, Fe_{0.180})_{0.986}S_{1.014}$	
3	$(Fe_{0.992}, Pb_{0.001})_{0.993}(S_{2.005}, As_{0.002})_{2.007}$	Pyrite
4	$(Fe_{0.997}, Pb_{0.001})_{0.998}(S_{2.00}, As_{0.002})_{2.002}$	
5	$(Fe_{0.984}, Zn_{0.005}, Pb_{0.001})_{0.990}S_{2.010}$	
6	$(Fe_{0.995}, Co_{0.001}, Pb_{0.001})_{0.997}(S_{2.001}, As_{0.002})_{2.003}$	
3	$Cu_{0.992}(Fe_{0.984}, Co_{0.001})_{0.985}S_{2.023}$	Chalcopyrite
	$(Cu_{0.993}, Bi_{0.001}, Pb_{0.001})_{0.995}Fe_{0.995}(S_{2.009}, As_{0.001})_{2.010}$	
5	$(Cu_{0.987}, Pb_{0.001}, Hg_{0.001})_{0.989}(Fe_{0.971}, Co_{0.001}, Zn_{0.013})_{0.985}(S_{2.025}, As_{0.001})_{2.026}$	
6	$(Cu_{0.993}, Pb_{0.001})_{0.994}(Fe_{0.974}, Co_{0.001}, Zn_{0.002})_{0.997}(S_{2.028}, As_{0.001})_{2.029}$	

In galenites there are contents of Sb, Ag and sporadically Hg, which isomorphically replace Pb. The Sb^{3+} replaces Pb^{2+} according to the scheme: $3Pb^{2+} \rightarrow 2Sb^{3+} + \dagger$ proposed by [17]. This heterovalent isomorphism is associated with the creation of point defects (voids). Sb^{3+} enters the galena structure as a paired isomorphism together with Ag^{+} in the equivalent positions of $2Pb^{2+}$ according to the scheme: $Sb^{3+} + Ag^{+} \rightarrow 2Pb^{2+}$. Such isomorphic substitutions are analogous to those observed in the polymetallic facies of the Munella deposit [20].

In Figure 7, it can be seen a clear correlation between antimony (Sb) and mercury (Hg), so, with the content reduction of the antimony, it also happens the reduction of the mercury and vice versa. Antimony contents are higher than those of mercury. Also, in this figure we can clearly see a distinct correlation between the difference in sulfur and that of antimony.

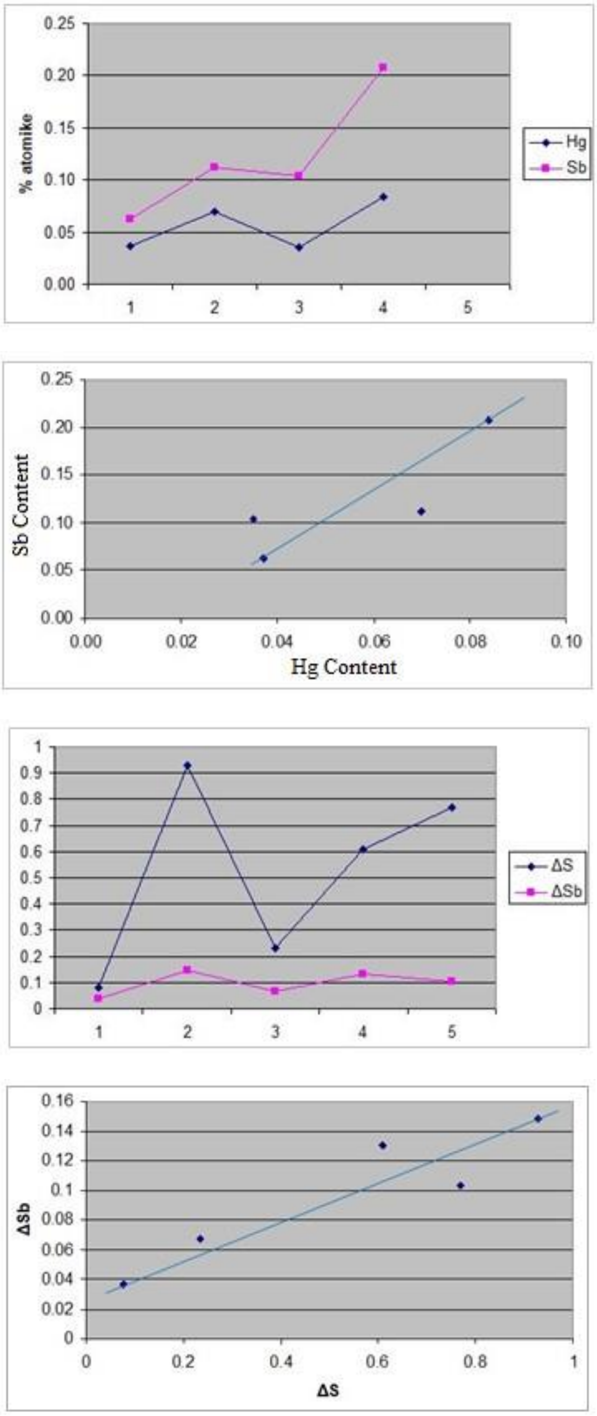


Fig. 7. Diagram of mercury (Hg), antimony (Sb), as well as the difference between sulfur (S) and antimony (Sb) in galena ore, Hajvalia deposit [5]

Sphalerite is characterized by high Fe content and a Fe/Zn = ¼ ratio. So we are dealing with the marmatite variety. The Co content is apparently associated with Fe in the isomorphism with Zn. The presence of metalloids (As, Bi), although within the limits of sensitivity, should be related to the heterovalent isomorphism $2(As, Bi)^{3+} \rightarrow 3Zn^{2+} + n$.

In the analyzed pyrite, the content of As and the predominance of Co, as it is noticed Ni, as well as Bi against Sb, stand out. The high contents of As are connected to the presence of As^{3-} in isomorphism with S^{2-}, as it is noticed in the pyrite from Qafë-Bari South, where the zonation formed by the concentration of As in pyrite is emphasized by the formation of arsenopyrite veins [1].

According to [15], the Ni and Co contents are quite low, implying that mineralizations formed below 400°C. Electron microprobe analysis relates to the Pb content, which is apparently due to the presence of galena micro-inclusions in pyrite. Although at the limit of sensitivity. Even in chalcopyrite we have a predominance of Co as it is noticed Ni and Bi as it is noticed Sb, but the most recent bindings of As. Should it be noted that the contents of Co and Ni are quite low and they are caught even at the limit of sensitivity.

In the studied preparations/polished section, e.g., by **[**4] from the mineralizations of the Hajvalia deposit, are shown the contents of Ag and Fe in the mineral of galena, sphalerite, and pyrite (Figures 8, 9).

In Figure 8 we notice that the high content of Ag (0.05–0.11%) is found in galena, sphalerite, and pyrite. At the same time, in many points analyzed in these minerals, the content of Ag is quite small, which speaks more about the presence of micro-inclusions than about an isomorphic state.

Figure 9 shows the distribution of Fe content in the sphalerite-galena mineralogical association. As is known, the Fe content in sphalerite depends on the temperature of the mineral formation and, in some cases, can be used as a geothermometer. According to **[4**] the sphalerite of the Hajvalia deposit has formation temperatures below 500°C.

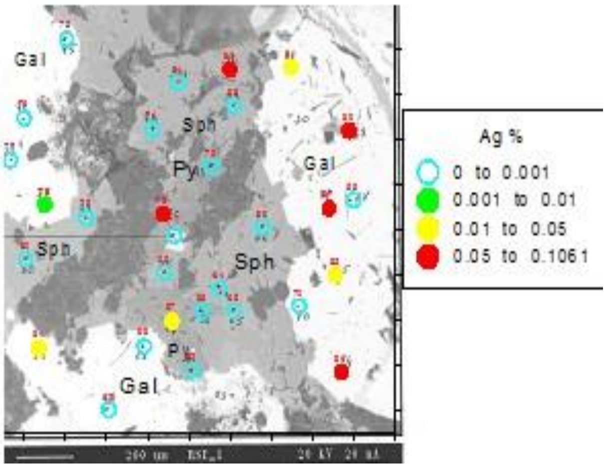


Fig. 8. Contents of Ag, H5-polished section [5].

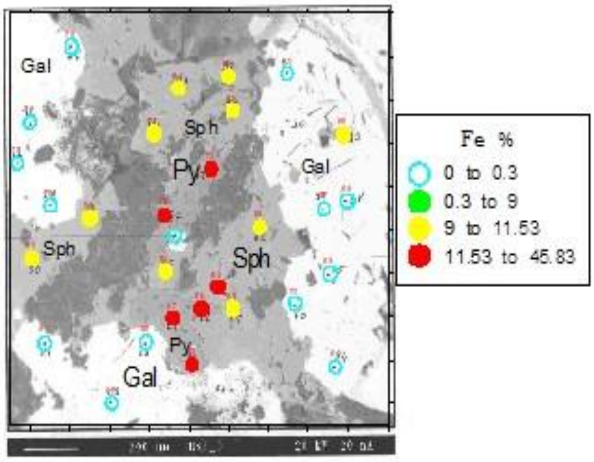


Fig. 9. Contents of Fe, H5-polished section [5].

CONCLUSIONS

The Hajvalia ore deposit is one of the richest polymetallic deposits in Europe and is particularly distinguished from other deposits in this region for its high content of zinc (Zn) metal. The main associated elements with zinc (Zn) in this mineral deposit are lead (Pb) and silver (Ag), while other elements such as Cd, Bi, Au, etc., can be found in smaller quantities. Contents of main metals in Hajvalia deposit are high with a predominance of Zn versus Pb (14.41% vs. 9.35%), and those of Ag about 108 g/t. The results of the correlative analysis are these geochemical associations: Pb-Zn-Ag; Zn-Ag-Cd; Sb-Bi. Our research found that factorial analysis produces the most accurate data for the following geochemical associations: Pb-Zn-Ag-(Cd); Sb-(Cd); and Bi-(Cu). In this study, a clear correlation between antimony (Sb) and mercury (Hg), was found (Figure 7). Antimony contents are higher than that of mercury. Also, we can clearly see a distinct correlation between the difference in sulphur and that of antimony.

From the microscopics description of the mineralogical preparations/polished section in the Hajvalia deposit, result this mineral composition: sphalerite, galena, pyrite, chalcopyrite, marcasite, arsenopyrite, pyrrhotite, tenanntite, as well as non-metallic minerals. Regarding silver, the statistical data in the diagram suggest a very strong correlation of silver with lead (Figure 6), which can be related to both the association of pyrargirite with galena as well as the isomorphic enrichment of galena with silver.

The Fe content in the sphalerite is quite homogeneous and varies from 9 to 11.5%, or 0.18 Fe atoms in the crystallochemical formula.

The main characteristic of galenites is their "purity", with a very high content of Pb. From the other elements Ag stands out and from once the presence of Hg. Contains more Sb than Bi. Characteristic is the presence of Te and Se in isomorphism with S, always with a predominance of Te.

Sphalerites are notable for their high Fe content. In contrast to galenite, the contents of Ag as well as Se and Te are lower, while it is found more often in Bi than Sb. In the analyzed pyrite, the content of As and the predominance of Co over Ni, as well as Bi against Sb, stand out. In chalcopyrite, we also have a predominance of Co over Ni and Bi over Sb, but lower As content.

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Р е з и м е

НЕКОИ ВАЖНИ КАРАКТЕРИСТИКИ НА ПОЛИМЕТАЛНАТА ОЛОВНО-ЦИНКОВА РУДА ОД РУДНИКОТ ХАЈВАЛИЈА, КОСОВО

Бедри Дурмишај[1], Силејман Хисени[1], Алберта Хисени[2]

[1]Универзитет „Иса Болетини“, Факултет за геонауки, Митровица, Република Косово
[2]Универзитет „Гоце Делчев“, Факултет за природни и технички науки, Штип,
Гоце Делчев 89, 2000 Штип, Република С. Македонија
sylejman.hyseni@umib.net

Клучни зборови: полиметален сулфид; олово-цинкова руда; геохемиска асоцијација; рудникот Хајвалија

Во овој труд се претставени општи геолошки податоци за полиметалното наоѓалиште Хајвалија. Посебно ги презентиравме научните резултати во врска со хемискиот и минералошкиот состав на сулфурните минерализации од наоѓалиштето Хајвалија. Со посебен акцент е проучена распределбата на главните метали користени при рударската дејност во ова наоѓалиште. Од микроскопскиот опис на примероците минерали од наоѓалиштето Хајвалија се добиваат следните минерални состави: сфалерит, галена, пирит, халкопирит, марказит, арсенопирит, пиротит, тенантит, како и неметални минерали. Геохемиските асоцијации укажуваат на силната поврзаност на Pb со Zn и Ag, а помалку со Cd и Cu. Постои уште една асоцијација на Sb со Cd во антагонизам со Cu, и асоцијација на Bi со Cu во антагонизам со As. Анализата на електронска сонда покажува дека галената е прилично чиста со спорадична висока содржина на Ag (до 100 ppm). Содржината на Fe во сфалеритот е прилично хомогена и варира од 9 до 11,5%, односно 0,18 атоми на Fe во кристалохемиската формула. Како што е познато, содржината на Fe во сфалеритот зависи од температурата на формирањето на минералот и, во некои случаи, може да се користи како геотермометар. Главна карактеристика на галенитите е нивната „чистота“, со многу висока содржина на Pb. Ag се издвојува меѓу другите елементи, како и присуството на Hg. Повеќе има Sb отколку Bi. Карактеристика е присуството на Te и Se во изоморфизам сп S, секогаш со доминација на Te. За разлика од галената, содржината на Ag, како и на Se и Te е пониска, додека почесто се среќава во Bi отколку во Sb.