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GENESIS OF LOW SULFIDATION TYPE EPITHERMAL ORE INDICATIONS AT ARANYOSBÉRC (MÁTRAKERESZTES), MÁTRA MTS., NORTH HUNGARY

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ABSTRACT

Indications are situated at the western border of the low sulfidation type western Mátra ore field, representing a characteristic part of the polymetallic-epithermal zone of the Inner Carpathian Volcanic Belt. The host rock shows intense silicification and K-metasomatic alteration. The poor, original sulfide mineralization (marcasite and arsenic pyrite) is accompanied by silicification, in form of stockworks and little veins with banded structure. Later these minerals were transformed into goethite and jarosite (gossan formation), indicating the high potassium content (13% K₂O) of the host rock. In the area there is an As-Sb-(Hg-Ba) anomaly (8500ppm As, 1000ppm Sb) belonging to the secondary minerals. The mineralization proceeded at 190-200°C. Two types of fluids could be distinguished of different salinity and composition (vein-forming and "apophysis" stage). The dilution of solutions by meteoric water was characteristic. Consequently, the upper region of the vein system of a low sulfidation type mineralization can be identified, which is located in shallower depth than the vein system of the western Mátra mineralization.

INTRODUCTION

Aranyosbérc is the largest continuous and most intensely K-metasomatized area of the Mátra Mts., and is situated at the western border of the K-metasomatized belt surrounding the western Mátra ore field (Fig.1.). Many scientist studied the genesis of this potassium-rich rock: according to VARGA (1966a, 1966b, 1992) and VARGA et al. (1975) this rock crystallized from potassium rich lava. On the other hand, KUBOVICS (1965, 1966, 1970) and MEZŐSI (1968, 1970) are of the opinion that this rock obtained its high K_2O (12-13%) content during an intense K-metasomatism. This later view is corroborated by the investigations accomplished in the Mátra Mts. by BAKSA et al. (1982), and in the Tokaj Mts. by SZÉKY-FUX (1970) and by MOLNÁR (1993).

The hydrothermal indications in these rocks were described by KUBOVICS (1966). He found 10 ppm Ag, and in traces Pb, Zn and Cu in the intensely pyritized quartzite at Aranyosbérc, generated by low temperature silicification. VARGA (1992) recognized three

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types of silicification here. According to MEZŐSI (1970) the silicification is closely related to the K-metasomatism.

Our purpose was the genetic study of the epithermal indications of this hydrothermally strongly altered area.



Fig.1. Important potassium metasomatite occurrences in the Mátra Mts. after Varga (1992).



Fig.2. Generalized geological map of Aranyosbérc and its surroundings after Varga (1966b) showing the sites of collected samples.

1 pyroxene-andesite ("middle-andesite"), 2 K-metasomatized lava rock, 3 K-metasomatized tuff, 4 andesite agglomerate and tuff ("upper-andesite"), 5 pyroxene-andesite ("upper-andesite"), 6 fresh andesite, 7 Kmetasomatized lava rock, 8 K-metasomatized tuff, 9 breccia, 10 gossan, 11 spoil, 12 quartzite, 13 vein quartz, 14 "apophysis" quartz, 15 borehole

GEOLOGICAL SETTING

The low sulfidation type (GATTER, 1997) western Mátra ore mineralization represents a characteristic part of the epithermal-polymetallic zone of the Inner Carpathian Volcanic Belt. It developed in close relation with the caldera structure in the middle part of the Mátra andesitic stratovolcano, which is characterized by three eruption cycles. According to the K/Ar data, the age of the whole volcanic sequence is about 12-16 Ma (HÁMOR et al., 1978). The last section of stratovolcanic series was strongly altered by hydrothermal fluids after the caldera formation (BAKSA et al., 1982). This potassium rich rock belt (average 8.50% K₂O) surrounds the uniform western Mátra vein system from western direction (VIDACS, 1965). The K-metasomatite is underlain by pyroxene andesite (so-called "middle andesite"), and overlain by more basic andesite tuffs and lavas (so-called "upper andesite"). The geological map of the studied area is shown in Fig.2.

The K-metasomatized rocks of Aranyosbérc and its surroundings are mainly of vesicular texture, but subordinately they appear as massive and brecciated types, as well. Separation of these rock types is not possible in the field. According to KUBOVICS (1965) the high frequency of the vesicular texture is due to the K-metasomatism of the originally vesicular andesites. In the late stage of K-metasomatism an intense silicification proceeded with the formation of quartz of stockwork structure (MEZŐSI, 1970). The second and third generations of silica consist of concentric or radial chalcedony, red jasper and cristobalite of low temperature origin (KUBOVICS, 1966). From the Hidegkút-hill south-southwestwards the rate of silicification and argillization (illite, celadonite, and montmorillonite) decreases, and the rate of K-metasomatization increases, because the low temperature silicification displaces the potassium feldspar from the rock (VARGA, 1992).

In the studied area two boreholes were drilled (Fig.3.), both of them penetrated the Kmetasomatized lavas and tuffs. The thickness of the potassium rich rock sequence is between 27 and 64 m. It is built up by two eruption cycles, and interrupted by three younger andesite apophyses (VARGA et al., 1975 and VARGA, 1992).





Fig.3. Boreholes drilled in the studied area (Gyöngyöspata-1. And Gyöngyöspata-3.) after Varga et al. (1975) and Varga (1992). 1 soil, 2 pyroxene-andesite ("upper-andesite"), 3 K-metasomatized lava rock, 4 Kmetasomatized tuff, 5 pyroxene-andesite ("middle-andesite") asl = above the sea level

ANALYTICAL METHODS

To distinguish the different rock and ore types and to recognize their alteration characteristics the polarization microscope was used.

The identification of constituents of ore mineralization, the distinction between SiO_2 phases and identification of clay minerals were accomplished by XRD analysis (Siemens D-5000 type X-ray diffractometer, Department of Mineralogy, Eötvös Loránd University).

Chemical analyses were performed by neutron activation (Canberra model, Technical University of Budapest) and by electron microprobe analysis (Amray 1830 I/T6, Department of Petrology and Geochemistry, Eötvös Loránd University).

Microthermometric studies were carried out by means of Chaixmeca freezing-heating stage on 0,1-0,5 mm thick sections of quartz (Department of Mineralogy, Eötvös Loránd University).

RESULTS

1. Rock types

VARGA (1992) distinguished five different rock types on the basis of the mineralogical and petrological study of K-metasomatites in the Mátra Mts. We recognized the massive, the vesicular, the brecciated, and the scoriaceous types at Aranyosbérc and in its surroundings. Each of these rock types was silicified in different rate.

The metasomatized rock consists almost solely of potassium feldspar and quartz. Mafic minerals are absent in the lava rocks, their previous presence is indicated by the limonitized and argillized nodes. These rocks are of flow texture: pilotaxitic or hyalopilitic, and subordinately perthitic. According to KUBOVICS (1966) this latter texture type is due to the ringwise precipitated opaque and clay minerals. Among the K-metasomatized tuffs the dust tuffs are prevailing. They contain a few little lapillis only very rarely, and we found pumice only in one case. However, in the tuffs the mafic minerals appear in form of fresh biotite. Both the lavas and the tuffs are often brecciated, their matrix consists mainly of quartz, and of limonite (goethite and jarosite).

The K-feldspar phenocrysts are often sericitized, and rarely limonitized (Fig.4a.). Moreover, fresh generation of hydrothermal K-feldspars (adularia) appears in the open vesicles and in fractures of the host rock (Fig.4b.). MOLNÁR (1994) recognized two generations of K-feldspars in the K-metasomatites of the Telkibánya ore deposit, as well. This indicates the increase of pH of the hydrothermal fluid, which is the result of loss of the acid dissociation gases, or of silicic acid release. The first process is due to the decrease of pressure by the opening of fracture system in the host rock, the second one to the sericitization of potassium feldspars. At Aranyosbérc the K-feldspars of metasomatites do not contain any fluid inclusions.

2. Alteration of the host rock

70-80% of the rocks consists of potassium feldspar, which is the result of strong metasomatism. The K-feldspar phenocrysts which displaced the primary plagioclases are often spotted and inhomogeneous. The fresh K-feldspars of the metasomatized rock underwent strong sericitization caused by acidic fluid migration. During this process silicic acid could be released in large quantities. According to the investigations of Mezősi







Fig.4a. Strongly sericitized K-feldspar phenocryst. Fig.4b. Fresh hydrothermal K-feldspar in the vesicle of the host rock. Fig.4c. Totally silicified metasomatite with mosaic structure quartz and chalcedony encrustation.

(1968) at Mátraszentistván, in the initial stage of metasomatism the mafic minerals and the groundmass were dissolved and replaced by K-feldspar, this process was followed by the displacement of plagioclase. In the late stage of metasomatism intense silicification proceeded, which resulted in quartz crystals in the leached vesicles and in fractures of the rock (stockwork structure) as well as dispersed in the groundmass.

Several generations of the strong silicification consisting solely of quartz can be observed in the fractures or in the vesicles of the host rock. Usually, the oldest quartz generation of mosaic structure is followed by idiomorphic quartz grown loose in the vesicles. The youngest chalcedony generation forms sheets and incrustations. The hydrothermal K-feldspar (adularia) appears in these quartz veins, indicating the boiling, and the "quasi" neutral pH in the epithermal systems (HEDENQUIST, 1995). Limonite sheets are often between quartz generations. This silicification is characteristic of the whole area, and its grade extends from the weakly silicified rock to quartzite (Fig.4c.). In addition, at the Korlát-hill we found typical vein formations: saccharoidal quartz and amethyst.

Argillization and hematitization are of secondary importance. According to the X-ray diffractometric analyses, this argillization is indicated by sericitization of K-feldspar phenocrysts, and by montmorillonitization of the groundmass.

3. Ore mineralization

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In the metasomatites of Aranyosbérc poor secondary limonitic ore mineralization appears in form of vesicle and fracture fillings as well as of massive ore (gossan). The primary sulfide mineral, the pyrite is in microscopic form, disseminated in the silica veins and displaced by goethite in the oxidized ore. The original ore minerals are indicated by the pentagonal and the hexagonal pseudomorphs after pyrite (Fig.5a.) and the pseudomorphs with rhomb-shaped cross section after marcasite (Fig.5b.). The pseudomorphs after pyrite are often built up by alternating limonite and jarosite zones. We recognized two generations of goethite: the older generation is of boxwork or radialspherolitic texture (Fig.5c.), the younger one is of incrusting, colloform texture (Fig.5d.). In these ores the two texture types of limonite suggest their formation from weathering of pyrite to boxwork habit or aqueous precipitation to botryodal-colloform habit (SANGAMESHWAR and BARNES, 1983). The structure pattern of the boxwork habit goethite is not homogeneous: the porous and the massive rhomb-patterned zones alternate often symmetrically. This phenomenon suggests rhythmic precipitation, or the zoned quality of the primary fracture filling phase. The spherolitic structure is characteristic of the thin encrustations. Jarosite occurs in form of fine-grained radial mass, mainly associated with goethite.

At Aranyosbérc and its surroundings the rock altered by hydrothermal fluids consisted of K-feldspar, quartz, and pyrite (marcasite). Close to the surface these minerals reacted with the O_2 -rich groundwater, which resulted in acid solutions as well as goethite and jarosite. During the weathering of pyrite, the first reaction product is always goethite. The sequence of reaction products following goethite varies with the irreversible flux of Fe and Al to the solution from dissolving minerals: reactions with molar ratios of Fe/Al greater than 1.0 precipitate jarosite. The formation of jarosite depends on the quality of both the sulfide phase and the K-bearing phase (BLADH, 1982).



Fig.5a. Hexagonal-shaped goethite and jarosite pseudomorphs after pyrite.
Fig.5b. Rhomb-shaped goethite pseudomorphs after marcasite.
Fig.5c. The older goethite generation of boxwork texture.
Fig.5d. The younger goethite generation of colloform texture.

4. Geochemistry

In the studied area there is an extended As-Sb-(Hg-Ba) anomaly, which is associated with Zn, as well (Tab.1.). The large quantity of Fe belongs to goethite and jarosite. In certain samples there is large quantity of potassium, which derives from the host rock. According to HEDENQUIST (1995) the As, Sb, Ba, Hg, Zn and K are the typical element association of the low sulfidation type epithermal systems. SZÉKY-FUX (1970) recognized that the K-metasomatism promotes the mobilization and accumulation of Ba, As, Hg, Pb, Zn, and Ni. The high arsenic content of these rocks is due to their high pyrite content. Accordingly, the geochemical anomaly, the ore mineralization and the silicification may be the result of processes linking with the potassium metasomatism at Aranyosbérc.

Table 1

Important anomalies at Aranyosberc and its surroundings (ppm).

 jarosite (Aranyosbérc), 2 breccia (Aranyosbérc), 3 limonitic breccia (Hidegkút-hill), 4 limonite (Korlát-hill), 5 limonite (Korlát-hill), 6 limonitic metasomatite (Aranyosbérc), 7 silicified metasomatite (Aranyos-brook), 8 vein quartz (Korlát-hill), 9 soil (Boc-brook)

	1	2	3	4	5	6	7	8	9
As	1386	6078	9.8	596	6526	8589	510	34	15
Ba	1423	604	578	369	352		450		190
Hg		10		5	7	7			
Sb	117	1181	84	71	739	1052	36	96	54
Zn	79	103	78	118	157	229		20	
Fe (%)	16.7	31	4.1	14	45	38	4.8	1.4	0.4
K (%)		11.2			7.8	13.6			
Na	4514	3822	6800	1020	7140	3920	750	540	630

The quantities of As, Sb, Hg and Zn correlate well with one another, and they belong to the Fe-bearing minerals (Tab.2.). According to microprobe analyses, the distribution of arsenic is not homogeneous in the ore samples, thus it does not belong solely either to goethite or to jarosite. The same investigations verified 2wt% arsenic content of pyrite (Fig.6.). Some sulfide minerals contain the largest quantities of arsenic as trace element. According to KISS (1982) the pyrite may contain 110-20000 ppm As and 25-1160 ppm Sb.

TABLE 2

Correlation matrix of As, Sb, Hg, Zn, and Fe at Aranyosbérc and its surroundings.

	As	Sb	Hg	Zn	Fe
As	1				
Sb	0.95	1			
Hg	0.84	0.90	1		
Zn	0.89	0.89	0.90	1	
Fe	0.94	0.86	0.84	0.88	1



Fig.6. Result of the microprobe analysis of arsenic-bearing pyrite from the gossan at Aranyosbérc. This sample contains 2wt% arsenic.

The limonite ore mineralization was formed by the weathering of the primary ore minerals under oxidizing conditions. At low temperature the iron and manganese were separated, and the anions were adsorbed by iron-hydroxide, and the cations rather by manganese-hydroxide (WEDEPOHL, 1972), so the limonite phase could strengthen significantly the original anomaly.

TABLE 3

Chemical composition of potassium metasomatites at Aranyosbérc and its surroundings after Varga (1992) (wt%).

1 borehole, Gyöngyöspata-1., 5 m, 2 Southern slope of Hidegkút-hill, 3 South-western slope of Hidegkút-hill, 4 Southern slope of Aranyosbérc, 5 South-western slope of Aranyosbérc, 6 Korláthill, 7 Southern slope of Korlát-hill, 8 Northern slope of Korlát-hill

	1	2	3	4	5	6	7	8
SiO ₂	54.97	68.01	65.25	67.64	66.79	66.46	66.00	65.79
TiO ₂	0.97	0.58	0.70	0.46	0.54	0.45	0.57	0.54
Al ₂ O ₃	18.81	16.02	14.23	12.09	15.83	15.58	15.46	15.57
Fe ₂ O ₃	6.27	1.97	5.04	4.75	3.34	0.01	1.48	1.00
FeO	0.40	0.24	0.33	0.47	0.56	0.42	0.45	0.40
MnO	0.10	Trace	0	0.09	0.02	0.04	0.06	0.04
MgO	2.90	Trace	0.27	0.91	0.91	0.52	0.74	0.37
CaO	1.45	Trace	0.71	0.90	0.72	1.03	0.63	1.26
Na ₂ O	0.49	0.34	3.56	0.39	0.92	0.21	036	0.25
K ₂ O	7.73	8.18	6.07	10.23	7.35	12.05	13.08	11.75
H_2O^+	4.41	3.31	2.08	2.19	2.41	1.56	0.93	1.65
H ₂ O [•]	1.00	1.22	0.83	0.10	0.19	0.83	0.57	1.37
CO ₂	0	Trace	0.19	0	0	0.50	0.34	0.32
P ₂ O ₅	0.16	0.05	0.71	0.16	0.07	0.02	0.06	0.12

In the analyzed samples the large quantities of potassium are due to their high jarosite content, which indicates also the high K content of the host rock as weathering product. According to VARGA (1992) 76-77% of the metasomatites consists of K-feldspar at Aranyosbérc, so the K_2O content of these rocks may be as high as 12-13% (Tab.3.). The jarosite may contain larger quantities of lead (plumbojarosite) and silver (argentojarosite), too (BROWN, 1970). The presence of lead and silver was not verified in the jarosite at Aranyosbérc, which indicates that either these elements were not present in the original ore mineralization, or they were completely leached during the weathering.

5. Genesis

According to fluid inclusion studies the homogenization temperatures determined from two-phase inclusions of hydrothermal quartz fall between 160 and 250°C (Fig.7.). The average density of fluid is $0,873g/\text{cm}^3$. The low salinity fluids are site-dependent: the salinity of fluid is 2-2,5wt% (NaCl eqv.) at the Hidegkút-hill and Korlát-hill, while at Aranyosbérc the salinity is 0,1-0,5wt% (NaCl eqv.). The development of fluids varies also with different sites: according to the T_h/c diagrams the low salinity fluid was fully diluted and depleted with decrease of the temperature at Aranyosbérc. Cooling and dilution took place at the Hidegkút-hill. The development of fluid was more complicated at the Korlát-hill, but small number of data do not allow sure conclusions (Fig.8.).



Fig.7. Homogenization temperatures measured from the fluid inclusions of quartz at Aranyosbérc, Hidegkút-hill, and Korlát-hill.

The composition of fluids differs, too: the composition of vein forming fluid is Ca-Nachloride type, and that of "apophysis" quartz forming fluid is alcaline bicarbonate/sulfate type (GATTER, 1983). These two types were recognized in the measured data as well, which verify the presence of chloride type fluid at Aranyosbérc (it proves the dilution of fluid), and the presence both chloride and bicarbonate/sulfate type fluid at the Hidegkúthill and Korlát-hill. The chloride type fluid predominates at the whole area suggesting the greater role of the vein forming stage.

DISCUSSION AND CONCLUSION

The epithermal mineralization at Aranyosbérc consists of strong silicification, which took place at the late stage of potassium metasomatism, and of poor secondary limonite mineralization (gossan). The original sulfide minerals were the arsenic-bearing pyrite and marcasite, which were transformed into goethite and jarosite under oxidizing conditions (gossan formation). The extended As-Sb-(Hg-Ba) anomaly belongs to the secondary Febearing minerals, and it could be increased during the weathering. The primary ore mineralization is accompanied by strong silicification in form of stockworks. In these rocks the hydrothermal potassium feldspar (adularia) is frequent. The argillization (sericitization of the K-feldspar phenocrysts, and montmorillonitization of the groundmass of host rock) and the hematitization are of secondary importance. The mineralization proceeded at 190-200°C. The salinity of the fluids was low, and the composition was mainly Ca-Na-chloride type. The dilution of solutions was characteristic, since the originally mixed fluids could be diluted-cooled by the direct influence of the meteoric water.

According to the alteration characteristics, the stockwork-type structure, the geochemical element association and the fluid characteristics obtained by the fluid inclusion studies the presence of a low sulfidation type epithermal system can be identified at Aranyosbérc. This system is located in shallower depth than the western Mátra vein system (Gyöngyösoroszi-Mátraszentimre) and at greater distance from the main upwelling channels, and represents the stockwork zone of the low sulfidation type systems, which forms above the vein zone (Fig.9.). Later this system was exposed on the surface, and was transformed under oxidizing conditions, therefore the original element association was enriched. According to Fig.9., the Pb, Zn and the Cu enrich in the vein zone below the brecciated zone. Above it is the zone of boiling, where Au and Ag accumulate. The western Mátra polymetallic vein type mineralization corresponds to the vein zone.

According to the boreholes drilled in the studied area, the thickness of the hydrothermally altered zone is small here. From the Aranyosbérc westwards important hydrothermal alteration zones are absent. The studied area is located vertically and horizontally at the greatest distance from the center of the western Mátra ore mineralization.

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