Mineralogy of gold and the characteristics of host rock in the Podpolom (Klokoč) high sulfidation type epithermal deposit

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Mineralógia zlata a chrakteristiky východzích hornín vo vysoko-sulfidickom type epitermálneho ložiska Podpolom (Klokoč)

V strednej časti stratovulkanického komplexu Javorie sa vyskytuje v podpolomskej oblasti epitermálne zrudnenie báden-pannónskeho veku. Zrudnenie je radené k tým hydrotermálnym centrám, ktoré vznikli vo vrchnej časti stockovitých dioritových telies, ktoré prenikli cez stratovulkanický komplex. Výsledkom viacnásobných dioritových intrúzií je vznik vyššie termálnych minerálnych sukcesií. Nabohatenie zlata sa viaže k oxidickým, limonitizovaným, kremeňom a brekciám s priemerným obsahom zlata 1-2 g.t^{-1}. Primárne zlato sa objavuje vo forme 1-3 μm veľkých zlatiniek s vysokou čistotou, prirastené na povrch kremeňa, alebo pyritu. Možno to pozorovať v oxidovaných brekciách a kremencoch na okrajoch dutín goethitu. Nabohatenie zlata v pripovrchovej zóne je výsledkom supergénnych procesov.

Key words: *Podpolom, HS type epithermal deposit, native gold, oxidized hydrothermal breccia*

Introduction

The only economic mineral, which was mined in the Javorie area in the middle of the eighties, was native sulfur. Diorite porphyry intrusions with advanced argillic alteration as a potential of ore mineralization were identified in the sixties (Valach, 1966 in Štohl et al., 2000). As a result of a base metal exploration program in the Javorie Mountains in the seventies and early eighties a porphyry copper system was discovered. In the second half of the nineties exploration was focused on the Podpolom (Klokoč) area where significant gold anomalies were found (Štohl et al., 1999).

The purpose of this study was the examination of the morphological and mineralogical characteristics of gold in the Podpolom epithermal deposit. The composition of gold minerals as well the rock alteration was also examined. Clarifying the mineralogical features of gold is not simply a scientific question as it can be an important factor causing technological problems in mineral processing.

Geological setting of the Podpolom deposit

The Podpolom deposit is situated at the central part of the Javorie andesite stratovolcano. This stratovolcanic series belongs to the Central Slovakian Volcanic Field and was formed from the Badenien through Pannonian. The stratovolcanic complex is intruded by several stock-like form diorite-monzodiorite intrusions. These intrusions are 500-1500 m in width with a vertical extent of at least 2000 m downward (Stohl et al., 2000).

There are two well-defined types of mineralizations in the Javorie stratovolcanic complex: porphyry-type mineralization connecting to the intrusions and high-sulfidation mineralization located above them.

The mineralization centers appear on the surface as altered zones related to the intrusive stocks. These centres were formed at Zaježová, Banisko, Skalka, Podpolom and Stožok localities. The intrusions occur within a NE-SW striking volcanotectonic depression. This linear belt is discordant to the regional E-W striking Carpathian Volcanic Arc. It may define a transfer structure associated with transpressional tectonics during the regional development of the Volcanic Arc (Štohl et al., 2000).

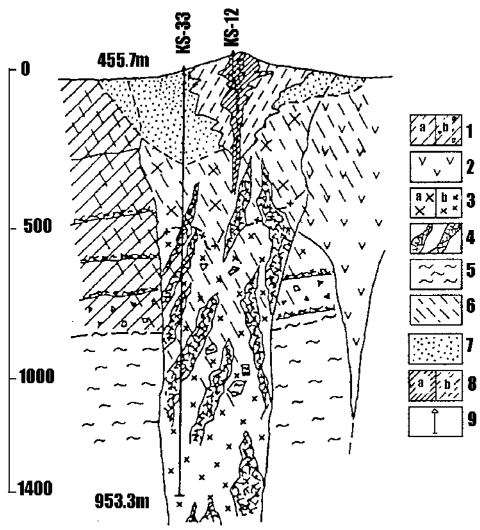

Fig.1. Schematic section of the hydrothermal center at Podpolom (After Konečný et al., 1998 in Štohl et al, 2000). 1: surrounding andesitic lava flows (a) and breccias (b), 2: hornblende-pyroxene andesite extrusive domes, 3: intrusive stocks of monzodiorite porphyry (a) and monzodiorite (b), 4: zones of magmato-hydrothermal breccias, 5: assumed crystalline schists of hercynian basement, 6: propylitic alteration, 7: argillic alteration, 8: advanced argillic alterations: a-quartzites, b-argillic quartzites, 9: drillhole.

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(Recenzované a revidovaná verzia dodaná 14.7.2003)

Connecting to the porphyry-type mineralizations the diorite-monzodiorite and the surrounding andesites underwent a propylitic alteration. In the central, upper part of the altered bodies, advanced argillic then argillic alteration developed *(Fig1)*. Rudimentary porphyry Cu mineralization appears with ore minerals like chalcopyrite, molibdenite pyrite, pyrrhotite, hematite and magnetite (Štohl et al., 2000).

At the Podpolom hydrothermal centre the zone of alteration is about one km in diameter. Drillholes in the Podpolom area intersected an epithermal gold mineralization with advanced silicic and argillic alteration zone above mesothermal zone with porphyry copper mineralization. Because of the too deep location of the copper ore, mining could not be an economic target.

The high sulfidation-type gold mineralization at Podpolom occurs in an oxidized, ferruginous breccia that has developed in the central core of a barren silicified zone. This breccia is also represented in the Podpolom quarry, where "sand" was exploited by the locals. This sand is fragmented residual silica, which developed in the internal part of the hydrothermal center or might be formed by the extreme hydrothermal leaching of the sulfide and argillic matrix from the very fine grained breccia (Štohl et al., 1999). A preliminary resource estimate at Podpolom is 250,000 t ore at the grade of 1.72 g/t and 0.7 g/t cutoff (Marlow et al., 1998). Due to the relatively low grade of ores and the small resource, mining activity has not been started at the locality.

Analytical methods

The basic aim of the study was to reveal the morphological and chemical characteristics of gold minerals and the genetic interpretation of the examination results. In accordance with this, the lithological features of the host rock as well as the rock alteration were also studied. The examined samples represent different rock types of the Podpolom hydrothermal centre. 30 samples were collected from the drillholes R4, R7 and R13 *(Fig.2)* in the Podpolom quarry. R4 dips 50° with the azimuth 0°, R7 is vertical and R13 is horizontal with the azimuth 347°. As gold assay results were known from former explorations, during the sampling the focus was on the parts of higher gold grades.

The microscopic characteristics were studied both in transmitted and reflected light by the AXIOLAB A-type polarization microscope with the MC 80 DX camera.

The scanning electron microscopic and microprobe analysis was carried out by the AMRAY 1830 I scanning electron microscope with the EDAX EDS detecting unit pv9700/36, (20 kV, SiLi detector, W cathode) at the Institute of Material Science, University of Miskolc, with the help of Árpád Kovács research engineer. The analysis were made place on the same thin sections that were subjected to the microscopic examinations.

The XRD analysis was carried out at the Hungarian Geological Institute by P. Kovács-Pálffy. The instrument was a computer-controlled and evaluated Philips PW 1730 diffractometer, with the Cu anticathode, 40 kV, 30 mA, graphite monochromator, goniometer-velocity 2°/min.

The gold assay results were produced by the ANALABS, Australia, Perth, using the fire assay – AAS finish method.

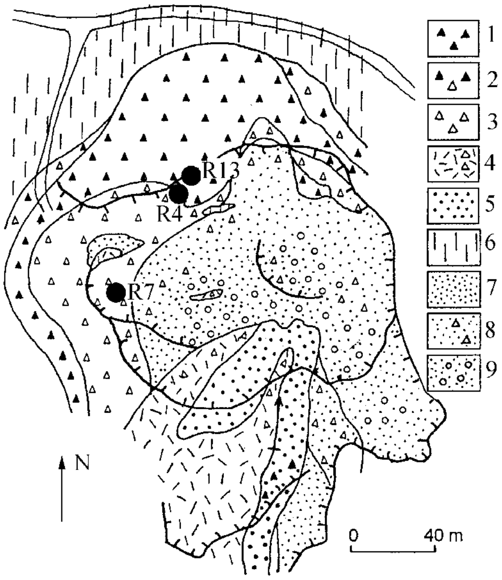

Fig.2. Distribution of rock types in the Popdpolom quarry showing the place of the examined drillholes (after Štohl et al., 1999).

Rock types and alterations at Podpolom

During the sample collection, the samples should represent the main rock types of the deposit. In the gold-bearing rock types the focus was on the parts with higher gold grades. Based on the application of the methods above, and comparing the data with the classification of Štohl et al. (1999), the examined samples were grouped into the following types: (1) vuggy silica; (2) limonitic breccia; (3) siliceous breccia; (4) grey, argillic silica. These kinds of rocks are common in the advanced argillic alteration zones of high sulfidation epithermal systems (Hedenquist & Arribas, 1999). At Podpolom deposit the biggest part of these zones are several generations of hydrothermal-explosive breccias (Štohl et al., 1999). Gold grades of the examined samples are shown in *Fig.3.*

Vuggy silica

This rock type is represented by the following samples: R4-1.5m, R4-19.6m, R4-24.8m, R13-0.7m, R13-2.3m, R13-3m, R13-4.9m, R13-6.1m, R13-13.9m, R13-27m and R13-34.2m. All samples from the horizontal drillhole R13 represent a near-surface position and the highest gold grades occur in these rocks. Gold content decreases with the depth in R4 *(Fig.3)*.

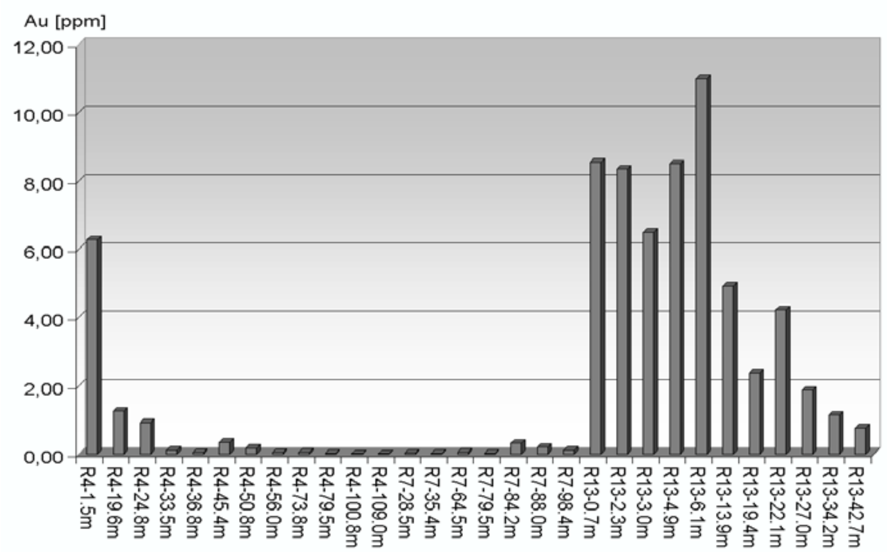

Fig.3. Gold assay results of the examined samples. Measured in ANALABS, Australia, Perth.

The rock is strongly siliceous, hard and porous with rare siliceous, subordinately white, argillic fragments. The colour varies from pale violet to ochre-brown depending on the iron oxide content. The size of vugs ranges mostly between 1-3 mm. The few grey, silicified or argillic, angular fragments are 1-4 mm.

Tab.1. XRD results of selected samples from the Podpolom deposit. Analyst: P. Kovács-Pálffy, Hungarian Geological Institute (wavelength 1,54186, Cu tube, 40kV, 30 mA, step 0,04, scanrange 2,2-66,2, max I 11174).

rock type	sample	ill/	illite		chlorite	alunite	pyr ll		ldsp.			pyrite	hem	goethite	augelite		cor	e
Vuggy silica	R13-6.1	-	4	2	-	-	-	80	-	-	1	-	5	8	-	-	-	-
Limon. breccia	R13-22.1	-	-	-	3	-	-	71	-	-	2	-	8	15	-	1	-	-
	R13-34.2	-	3	1	-	-	-	83	2	1	-	-	1	5	-	1	4	-
Siliceous breccia	R7-35.4	-	-	-	-	-	-	97	-	-	-	-	-	-	-	1	2	-
	R7-64.5	-	-	-	-	-	-	79	-	-	1	-	-	-	-	17	3	-
	R7-84.2	-	1	1	-	1	-	90	-	1	1	-	-	-	4	1	-	
	R7-88	-	-	10	-	-	21	36	-	-	2	4	-	-	-	-	-	27
	R7-98.4	-	3	14	-	-	13	60	-	-	1	3	5	-	-	-	-	1
Argillic silica	R4-79.5	-	-	4	-	26	10	55	-	-	1	5	-	-	-	-	-	-
	R4-100.8	17	-	-	-	-	-	58	4	-	-	18	-	-	-	-	-	3
	R4-109	20	-	-	5	4	-	47	-	-	1	21	-	-	-	-	-	2

Microscopically this rock type can also be considered as a strongly silicified breccia. It might have undergone multiply brecciation and silicification. Due to the multiply re-silicification processes the contours of the previously formed, silicified fragments are slurred, and the rock has become having a homogenous vuggy silica character. It is a chaotic mixture of fine-grained quartz and limonite, with irregular pores and vugs. The contours of the mosaic-textured quartz grains are slurred, the limonite appears in patches or veins. Second-generation, coarser-grained quartz fields occur in the finer-grained material but not with so sharp contour as fragments. The rare "real" fragments are angular, with definite contour, consisting of very fine-grained (first-generation) quartz, usually free of limonite. Some parts of the silica show cracled brecciation, in which the crackles are filled by limonite. A few, euhedral pyrite crystals also occur in the vuggy silica but these are rare and can be observed mainly by electron microscope because of their small size (1-3 μm). Kaolinite and illite are present in a few percent *(Table 1)*.

Limonitic breccia

This type of rock was studied in the following samples: R4-33.5m, R4-36.8m, R4-45.4m, R4-50.8m, R4-56m, R13-19.4m, R13-22.1m and R13-42.7m.

The colour of the rocks varies from light greyish-ochre to chocolate-brown depending on the limonite content. The breccia character can be matrix-dominated (R4-36.8m) to fragment dominated (R13-22.1m). The size of fragments ranges between 1-6mm, their colour is dark or light grey. Microscopically the matrix is strongly limonitic, rarely with vuggy character. The fragments can be angular or subrounded *(Fig.4)* and contain minor limonite or are free of limonite. All of the fragments are silicic, but the grain size and texture of the quartz in the fragments are different, indicating multiply brecciation. Many of the fragments consist of fine-grained or colloidal quartz with slurred grain-contour and limonitic patches. These are probably fragments of vuggy silica. In the other types of fragments the quartz is coarser-grained, has a definite contour and shows a mosaic-pattern.

In the sample R4-36.8m significant amount of alunite occurs, frequently intergrown with limonite. The alunites form tabular or laminar crystals of 0.2-0.5 mm and developed in clusters, in the vugs or pores of the matrix. Their presence indicates the strong acidic character of the mineralizing fluids. Zircon and rutile are frequent as a few μm to few tens of μm large euhedral-subhedral crystals. Goethite appears mainly in the surroundings of pores, often with a hematite rim.

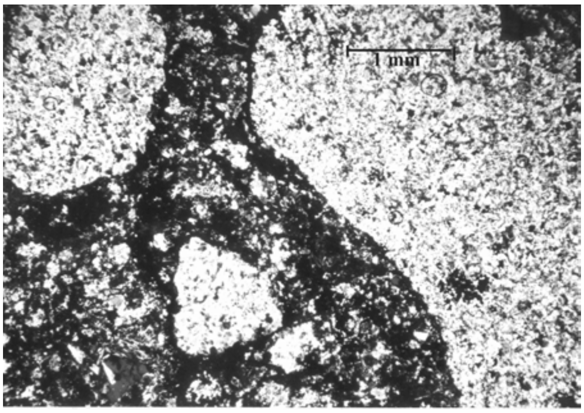

Fig.4. Subrounded, silicic fragments, strongly limonitic silica matrix with shattered rock flour. R4-36.8m, Limonitic breccia. Au: 0.05 g/t. Transmitted light, +N.

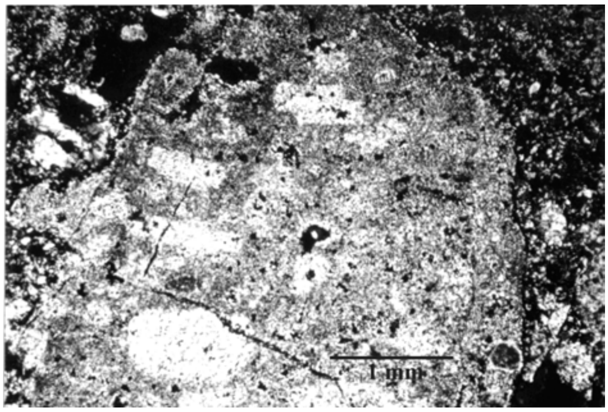

Fig.5. Multiply brecciated fragment in siliceous breccia. R7-84.2m. Au: 0.32 g/t. Transmitted light, +N.

Siliceous breccia

This rock type was observed in the following samples: R7-28.5m, R7-35.4m, R7-64.5m, R7-79.5m, R7-84.2m, R7-88m and R7-98.4m. In the examined samples gold content is very low in the highly silicified types down to 84 m but it is higher in the more argillic ones between 84-90m.

The rocks of this group are hard, strongly siliceous, and have light to darker grey colour, with 2-5 mm large, angular fragments. The fragments are mostly grey silica, subordinately white argillite or argillized andesite. The dominating matrix is grey, silicic rock flour. The rocks consist of almost pure quartz. Topaz and corundum are also present; topaz content can be is as high as 17%, according to XRD results *(Table 1)*. At deeper levels, (below 80m) the matrix is more argillic with abundant pyrophyllite, diaspore and kaolinite.

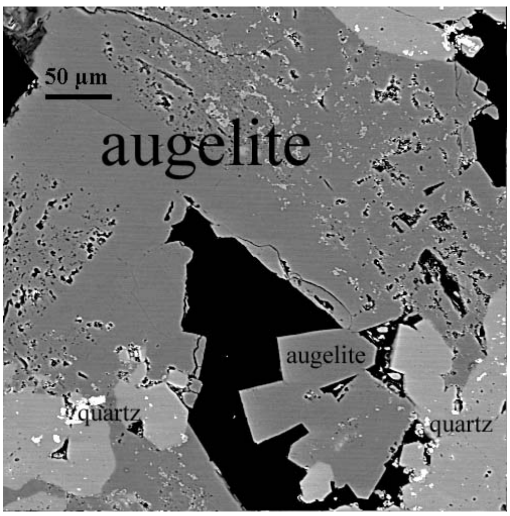

Fig.6. Augelite, quartz and topaz in a vug of matrix in siliceous breccia. Topaz: small, light grey patches in augelite. R7-84.2m. Au: 0.32 g/t. BSE image.

There are several types of fragments at different levels. Some fragments are sharply angular and built up by formerly shattered then cemented silica. At the other type of fragments the contour is not sharp because multiple silicification dissolved the edges of the fragments. Among this kind of fragments there are coarser- and finer-grained types. Several pieces of evidence indicate multiply brecciation. Fragments consisting of previously brecciated then cemented particles are frequent *(Fig. 5),* which also can be encrusted by limonite and goethite.

At R7-84.2 m augelite and topaz appear in he vugs and small cavities of the silica matrix. The augelite ($Al_2PO_4(OH)_3$ is a rare phosphate mineral, which may occur in relatively high-temperature Al-rich hydrothermal deposits (Visser et al., 1997). It forms tabular crystals of 0.1-0.5 mm *(Fig. 6).* Sometimes it completely fills the vugs. Minor amount of other phosphates were also observed in association with the augelite in form of irregular patches. These minerals are Ca-dominant Al-phosphate-sulphates (APS) with Sr, Ba and minor amount of REE. In certain vugs the augelite is intergrown with topaz. The topaz also forms clusters of tabular crystals about 10 μm across or encrustings of rutile cores.

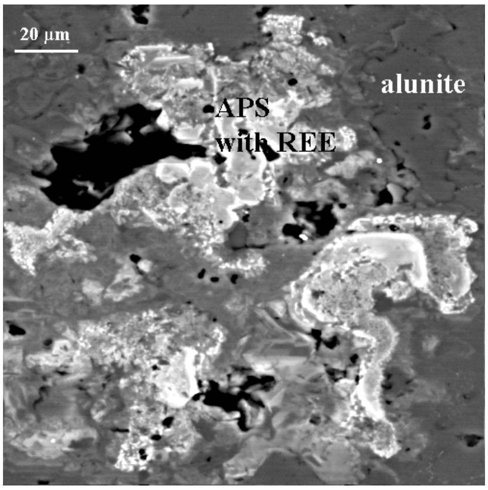

Fig.7. Irregular forms of Al-phosphate-sulphate minerals with REE content in alunite. Grey, argillic silica. R4-79.5m. Au: 0.02 g/t. BSE image.

Toward depth, after an interval of disintegrated, sand-like fragments, the rock becomes more argillic. At 88m and 98.4m significant pyrophyllite, diaspore and kaolinite are present. At 88 m the rock is coarse-grained, fractured silica with veins filled by pyrite, argillites and colloidal quartz. At 98.4 m a polimict, friable breccia occurs. The fragments are mostly grey silica of different shades, but pyrophyllite-rich, argillic fragments also occur. Microscopically, the boundary between the matrix and the fragments is not well defined. The matrix is predominantly argillic with pyrophyllite and kaolinite

Grey, argillic silica

This rock type was examined in the samples R4-73.8m, R4-79.5m, R4-100.8m and R4-109.0m. The rock samples from R4 are grey, silicified, compact and hard at 73.8m and 79.5m. Dense, very fine pyrite dissemination can be seen even by sight. At 100.8m and 109m it is more clayey, greenish, porous and disintegrated. Gold grades in the examined samples are very low, range between 0.01-0.06 ppm.

Microscopically the rock is a fractured silica with less argillites. The fractures are filled by coarse-grained, mosaic-textured quartz. The argillic assemblage is pyrophyllite-kaolinite with significant alunite *(Table 1)*.

Two generations of quartz can be observed. The contours of the first-generation quartz grains are slurred. This indicates the early silicification. The second-generation, mosaic-textured quartz grains appear with definite contour, in patches or fill the thin fractures in the silica.

Relatively much alunite and Al-phosphate-sulphate (APS) minerals were found at R4-79.5m. The alunite occurs as 20-200 μm large, elongated rhombs or lamellae. It forms clusters or fills veins and cavities with the second-generation quartz and pyrite. The abundant pyrite shows close, interstitial association with alunite, which proves the hydrothermal origin of the latter. The phosphate-bearing minerals can be detected only by scanning electron microscope. They appear mostly as bright, irregular-ragged portions in the alunite or form cores of alunite crystal groups *(Fig.7)*. They also occur in the silica matrix, separate from the alunites and are composed mainly of (K-Ca-Ba)-Al-(S-P) and (Ca-Ba-K-Sr)-Al-(P-S) elements, respectively. These minerals also contain REE elements at the rims or sometimes in the core.

Mineralization

The most abundant ore mineral in the Podpolom deposit was primarily pyrite, which turned into limonite, hematite and goethite above the oxidation level. However, a few, 1-2 μm large, euhedral pyrite crystals still can be observed as tiny inclusions in quartz or relict kernels in goethite. Pyrite is absent in the examined highly silicified grey breccias. Unaltered pyrite is abundant in the polimict, silicic-argillic breccias and the grey, argillic silica. In this rock types two generations of pyrite can be observed. A few, larger (30-100 μm across), corroded, euhedral-subhedral crystals occur in silicified fragments, mostly at small vugs and cavities. Being in a fragment, the formation of the first generation pyrite must have been followed by further brecciation. Much smaller (a few microns), second-generation irregular pyrites are disseminated in the siliceous-argillic matrix. Part of the pyrite in the matrix turned into limonite, displaying brownish patches in the silica material. Second-generation pyrite occurs not only scattered, but also in tiny veinlets crossing each other. In prints, chalcopyrite is also present in the argillic polimict breccias. These characteristics, together with the minor kassiterite, galena and sphalerite, which were detected by SEM may indicate the transition into the porphyry mesothermal system.

In the limonitic breccias and the vuggy silica a few μm large cinnabar and needle-like antimonite crystals were identified. These minerals indicate ore formation at low temperature, and cannot correlate with the pyrophyllite-topaz-corundum alteration assemblage, which was determined in the same rocks. There should have been at least two-phase alteration and the cinnabar and antimonite are linked to the lower-temperature alteration, associated with kaolinite-alunite-illite, which are present in the same rocks. Sphalerite was found in the same sample as antimonite and cinnabar, in form of a few 10-20 μm large irregular grains but it also occurred in the siliceous breccia at R7-84.2m. Kassiterite was found by SEM only in one sample at R4-100.8m in the grey, argillic silica as a 10 μm large, ahedral crystal.

Rutile is relatively frequent (1-2 wt %). It probably belongs to the high-temperature mineral assemblage. A few μm large, elongated or prysmatic rutile crystals appear both in the fragments and the matrix as fine dissemination. Sometimes rutile clusters are pseudomorphs after ilmenite. The siliceous breccias with high topaz content, which do not contain pyrite rutile is the only opaque mineral. Rutil sometimes occurs in association with zircon, but the latter was an accessorial mineral in the original rock, which came to the epithermal system without modification.

Characteristics of gold minerals

In the Podpolom mineralization gold occurs mainly in the matrix of the limonitic breccia and the vuggy silica, and is associated with goethite (Seres-Hartai & Földessy, 2000) *(Fig. 8, 9)*. However, Štohl et al. (1999) pointed out, that ferruginous breccias did not always contain gold. The amount of gold in the breccias shows definite correlation with the Fe-oxide content. Gold content decreases toward depth and with the dominance of argillic alteration. Gold in these breccias occurs at the edge of goethite in small cavities or fractures.

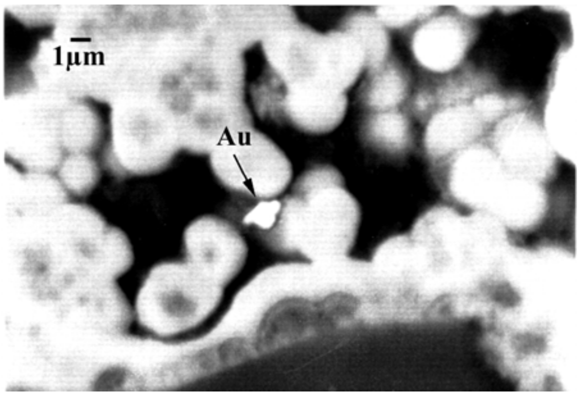

Fig.8. Native gold in ferruginous, vuggy silica, grown on the edge of goethite at a vug. R13-3m. Au: 6.5 g/t. BSE image.

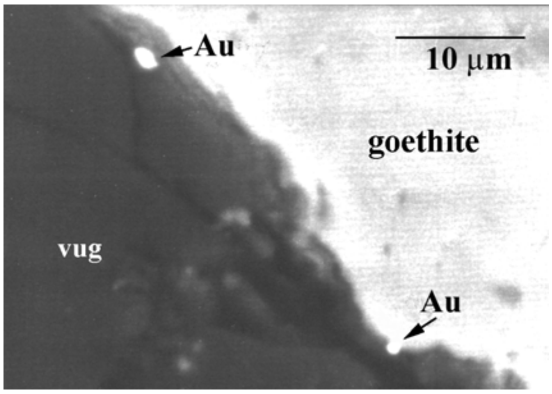

Fig.9. Native gold on goethite at a vug. Limonitic breccia, R4-45.4m. Au: 0.35 g/t. BSE image.

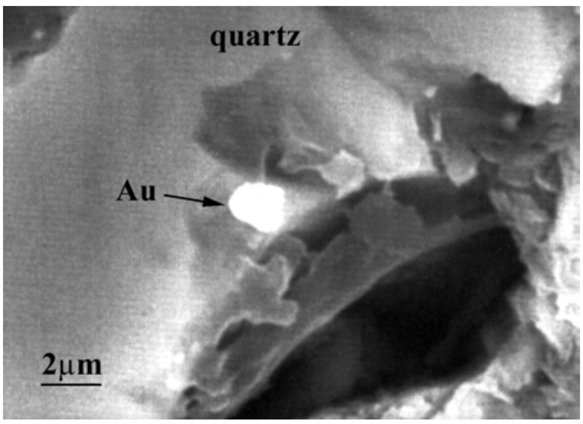

Fig.10. Native gold lamella on second-generation quartz. Grey, argillic silica. R4-79.4m. Au: 0.02 g/t. BSE image.

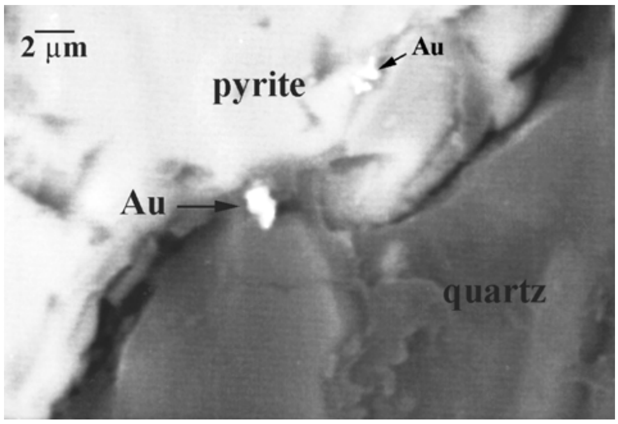

Fig.11. Irregular gold lamellae on veinlet-filling, second-generation pyrite. Grey, argillic silica. R4-79.4m. Au: 0.02 g/t. BSE image.

Below oxidation level, gold content is much lower. However, native gold was found by SEM with abundant pyrite in the grey, argillic silica, both in the compact and the disintegrable varieties. The gold occurs mainly related to quartz, grown usually on the surface of quartz crystals *(Fig.10)*. This quartz is the veinlet-filling, mosaic-textured, second-generation one. Gold also appears in close association with second-generation pyrite, forming tiny coating on it *(Fig.11)*, consequently its formation must have followed the crystallization of pyrite. Gold was not found on the first-generation pyrite.

The primary gold minerals in the argillic silica related to quartz or pyrite are usually irregular lamellae and their sizes are 1-3 μm. Gold grains in the ferruginous breccias and silica are rounded and their sizes in these rock types are a little smaller, range between a few tenth μm to 1 μm. This indicates that the gold grains could have been mobilized and rounded during the supergene processes and gold content must have increased in the near-surface levels. All native gold are of high fineness, no other chemical components beside Au could be measured in it.

Genetic aspects

Former studies pointed out the epithermal high sulfidation character of the Podpolom deposit: the mineralization is located above a diorite porphyry intrusion, and genetically linked to the porphyry copper mineralization in it; ore mineralization occurs in brecciated rocks, sometimes displaying multiply brecciation; the central mineralized core is characterized by pervasive silicification, vuggy silica is present; advanced argillic alteration also developed (Štohl et al, 1999, 2000). The recent examinations also support these statements.

However there are some new results, which can contribute to the better understanding of ore-forming processes. Beside the dominant quartz, as a result of rock alteration, topaz, corundum, pyrophyllite, diaspore and augelite occur in the same rocks, in which kaolinite, alunite and illite were also found. The formation temperature of the first groups of minerals is much higher than that is for the second group (Duggan et al., 1990, Bottrill, 1998, Hedenquist et al., 1999).

The relative abundance of phosphate minerals in the system is also worth emphasizing. Many studies show that secondary hydrothermal phosphates are formed by the decomposition of the primary apatites of the host rock (Bajnóczi et al, 2002). In the examined samples the abundance of augelite – and other phosphate minerals in the deposit - indicates that the hydrothermal system was rich in P and Al. It can be assumed that the hydrothermal fluids mobilized phosphor not only from the apatite of the host dioritic stock but that of the Herynian granitic basement as well. The aluminium can be originated by the extremely acid leaching of host rock.

Duggan et al. (1990) found augelite and other Al-phosphates with topaz and rutile formed by hydrothermal alteration of Triassic andesite and calculated the formation temperature between 450-500˚C. Visser et al. (1997) observed in metamorphic quartzites that augelite had developed after crandallite as a result of transformation due to an increase in temperature and pressure. During this stage Ca, Sr and Ce are partly removed from the rock. This kind of transformation could take place also in the Podpolom system. For the first, as a late phase of alteration, APS minerals formed in the vugs of the silica matrix of hydrothermal breccia. The formation of the dioritic stock is a result of multiply emplacement (Štohl et al, 2000), during which an increase in temperature could have resulted the transformation of former phosphates into augelite. The formation of topaz in close association with augelite can also be connected to this later, higher-temperature phase. In the drillhole R7, between 35-65m the topaz content increases as high as 16% with 2-3% corundum, which also suggests a higher-temperature effect.

The primary gold mineralization can be linked to the earlier, lower-temperature rock alteration, following the strong acid leaching. The formation of native gold and pyrite took place in distinct phases. The first-generation pyrite (corroded, in breccia fragments) was not associated with gold. The crystallization of second-generation pyrite (in veinlets and scattered) was followed by the gold mineralization as gold is grown on pyrite surface. The formation of antimonite and cinnabar can also be connected to the lower-temperature rock alteration.

The ferruginous, limonitic breccias and silica, which must have been abundant in pyrite, are usually characterized by high gold content (1-10 g/t). However, there are ferruginous breccias without gold. The grey argillic silica with abundant pyrite and low gold assay results (0,01-0,06 g/t) also suggest that there is no correlation between pyrite and gold content though Fe and Au shows correlation in the oxidized rocks. As Štohl et al. (1999) also pointed out, strong limonitization and gold enrichment in the near-surface levels are due to supergene processes.

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