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Copper Ore Deposits in the Gradski Potok Ore Field of the Trgovska Gora District of Croatia

Ivan JURKOVIĆ

Rudarsko-geološko-naftni fakultet Sveučilišta u Zagrebu, Pierottijeva ul. 6, YU — 41000 Zagreb

In this paper the author reports the results of investigations of the copper ore deposits Gradski potok, Švinica and Katarina of the Gradski potok ore field in the middle part of the Northern Trgovska gora ore district built up from Upper Palaeozoic sediments. The author describes the history of mining and geological investigations, the morphology of ore occurrences, the microphysiography of hypogene and hypergene minerals and the succession of mineralisation.

U ovom radu autor iznosi rezultate istraživanja bakarnih rudnih ležišta Gradski potok, Svinica i Katarina u rudnom području Gradski potok u središnjem dijelu sjeverne Trgovske gore izgrađene od gornjopaleozojskih sedimenata. Autor je izložio historijat dosadanjih rudarskih i geoloških istraživanja, prikazao način pojavljivanja bakarnog orudnjenja, izložio rezultate detaljnih mikroskopskih ispitivanja hipogenih i hipergenih minerala sa sukcesijom mineralizacije.

INTRODUCTION

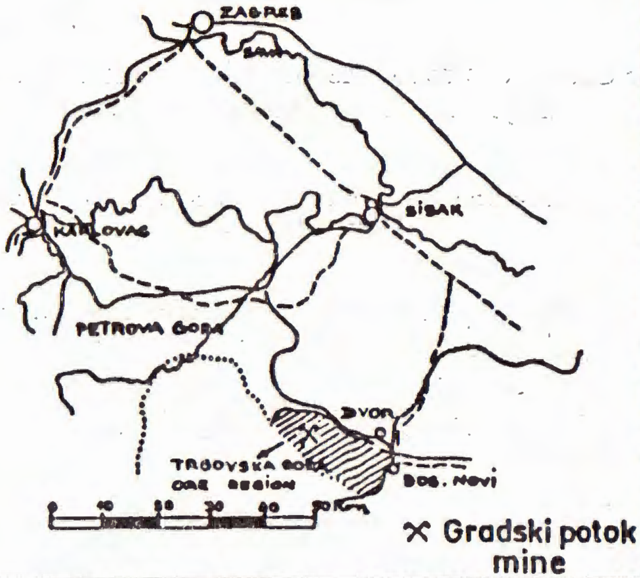
The copper-bearing siderite deposits in the Gradski potok area are situated further east than the silver-bearing galena deposits in the Zrin area (Jurković & Durn, 1988) and the Čatrnja area (Jurković, 1989) hitherto described. The area extends in the South as far as the Velebit brook from point + 230 to point + 162 below the village of Stambolije, on the West along the Hasanov Grob spur (+ 385 m), in the East along the Stambolije brook, a left bank tributary of the Velebit brook as far as point + 381, and on the North to the upper reaches of the Svinica water-course, a tributary of the Sredorak, and the Dragiševac brook, a tributary of the Lesser Majdanski brook. The central part of the ore district is 3.5 km from the village of Bešlinac (Fig. 1 and 2). Three copper deposits were located here and partially exploited: the largest is the Gradski potok mine (August), with workings from no. 27 to no. 37 on Fig. 2, then the Svinica mine (workings nos 38 and 39), and the Katarina mine (workings nos 40 and 41).

HISTORY

In Illyrian and Roman times mining was carried out in the Gradski potok area for hard limonite ore from the oxidation zone of the siderite deposits. With the

Fig.1

POSITION OF THE TRGOVSKA GORA IN CROATIA
(položaj Trgovske gore u Hrvatskoj)



collapse of the Roman Empire mining activity came to a stop. Mining for limonite was resumed by Saxon miners during the tenth and eleventh centuries. At a time of intensive mining under the Counts Zrinsky between 1453 and 1577 only silver-bearing lead ore was extracted (mainly in the Zrin and Čatrnja districts), while the copper ore, which was the principal ore in Gradski potok, remained untouched. At the beginning of the eighteenth century the Austrian court financed small scale prospecting works. Following the loss of the Saxon mines, the Empress Maria Theresia encouraged mining in Croatia from 1768 onward, and according to Wojtanek (1772) copper deposits in Katarina and Gradski potok were prospected, while Hacquet (1789) reports prospecting for copper ore in Kornelija and some other localities. The emphasis in mining was nevertheless on iron ore, and the »Trgovi Iron Works« founded in 1794 went on producing nothing but iron ore and smelting crude iron until 1832. It was the Klagenfurt merchants, Planker and Jäger, who converted production to copper ore, having purchased the mining rights from the Austrian state. These included concessions for Gradski potok, Katarina, Upper and Lower Breunner, Upper Gradski potok and Kornelija (Szedelmy, 1845; Lipold, 1855). The ore was smelted in a copper smelting plant in Bešlinac which had been built by the Austrian government in 1842. From 1858 to 1870 the mining and smelting rights were in the hands of the Viennese business man Désiré Gilain, who founded the »Trgovski mines and smelting works Bešlinac«. v. Hauer (1870) gives an account of this period. Following a fall in the price of copper on the world market, the new proprietor Frohm and his successor Mulay stopped producing copper ore and converted the smelting works into a blast furnace for the production of crude iron (Zloch, 1897). From 1894 until 1901 this mining activity was at a standstill, and the blast furnaces and the mining rights were taken over by the »Société anonyme des hauts fourneaux, mines et fôrets en Croatie, Trgovo-Bruxelles«. This company resumed mining for copper ore, deepened the Gradina and Gradski potok pits and went on producing ore until 1914. Work ceased in that year, because the mine workings revealed that the ore-bearing level deteriorated downwards on account of major faults in the NW and SE parts of the deposits which ran counter to the direction of the decline, and also because of an impoverishment in the copper content of the ore-bearing zone. This period is described by Reuter (1910) and v. Papp (1919). Between 1914 and 1945 the mining rights changed hands several times, but mining operations were not resumed, except during the period 1936—1941, when the remaining mining machinery was removed from the main extraction gallery. It was not until 1952 that the »Sisak Iron Works« cleared the main adit at Lower Breunner (no. 30) in Gradski potok and the Lower Katarina adit (no. 30), but these operations were soon abandoned.

THE GEOLOGICAL STRUCTURE OF TRGOVSKA GORA

The central parts of Trgovska gora, where iron, copper and lead ore deposits occur, are formed from Upper Palaeozoic sediments. Palaeontological analyses carried out to date have established *Lower Devonian* (D_1) in the valley of the Ljubina brook, then *Lower Carboniferous* (C_1) near the confluence of the Ljubina with the Žirovac brook and near the junction of the Majdanski brook and the Žirovac, *Middle Carboniferous* (C_2) in the SE part of Trgovska gora, and *Upper Carboniferous* (C_3) near the village of Kosna and at point Klupica and the Meterize area. The Devonian and Carboniferous strata consist mainly of shales, with layers of grauwacke, subgrauwacke and other types of sandstones of varying thickness. No fossils were detected in the roof series, which is generally of a sandstone character (more detailed account in Jurković' study, 1988). In a tectonic sense, Trgovska gora represents a unit constructed from the Palaeozoic, the Triassic and reduced Jurassique and Cretaceous. It is allochthonic in situation, overlying a base of Cretaceous sediments in the SW and W direction (Šparica, 1981).

THE MORPHOLOGY OF THE COPPER DEPOSITS

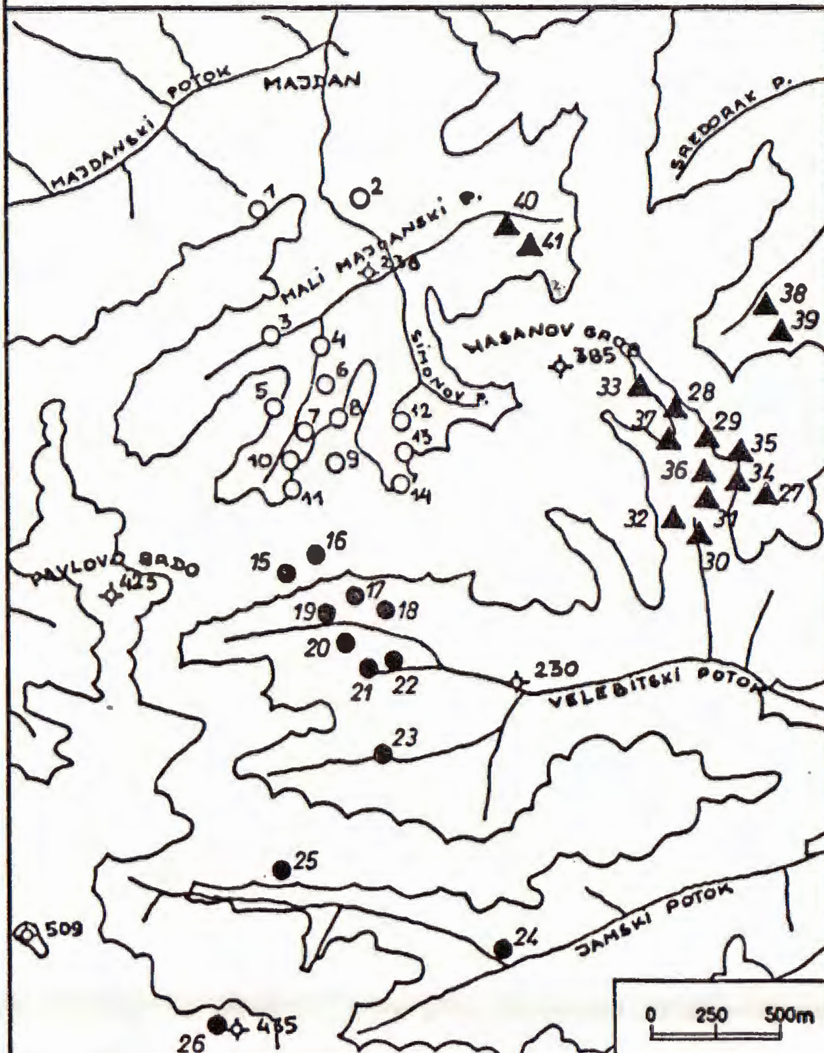
The Katarina ore bed (40 and 41) is situated by the upper reaches of the Dragiševac brook in Upper Palaeozoic (Carboniferous) shales with intercalations of sandstones. The mineralisation is represented by two ore-bearing zones running parallel to each other in a NW—SE direction, with a gradient downwards of 35° to 55° towards the SW, the first thicker and richer in ore 240 m long, opened up by the Lower Katarina adit (40), the other a thinner, poorer ore bed, 120 m in length, opened up by the Upper Katarina adit (41). Within the ore-bearing zone the mineralisation takes the form of stratified veins (beds), or else lenses concordantly embedded in shales (Wojtanek, 1772; Szedelmy, 1845; v Andrian, 1868; Tućan, 1907; Reuter, 1910; Reithofer, 1910; Jurković, 1962). The main paragenetic mineral is siderite with varying quantities of quartz and sulfides, amongst which pyrite is most frequent, followed by chalcopyrite, a great deal less tetrahedrite, and only here and there some sphalerite and very rarely galena.

The Svinica ore-bed (workings 38 and 39) is located on the upper reaches of the Svinica brook, a right bank tributary of the Sredorak brook, which flows into the Žirovac water-course near the village of Trgovi. Investigations have established the existence of two parallel mineralised

Fig. 2

Pb-Cu ORE DEPOSITS IN TRGOVSKA GORA

ORE DEPOSITS: O Pb (ZRIN) ● Pb (ČATRNJA)
▲ Cu (GRADSKI POTOK)



27. NOVA NADA 28. UPPER BAPTISTA 29. LOWER BAPTISTA 30. LOWER BREUNNER 31. KORNELIJA 32. KEMPEN 33. GLÜCKAUF 34. UPPER BREUNNER 35. UPPER GRADSKI POTOK 36. LOWER GRADSKI POTOK 37. GRADINA SHAFT 38. LOWER SVINICA 39. UPPER SVINICA 40. LOWER KATARINA 41. UPPER KATARINA

zones concordantly embedded in carboniferous shales with intercalations of sandstones. The extension of the series is in a NW-SE direction, declining towards SW. The foot-wall of the ore-bearing zone was explored by the Lower Svinica adit extending for 140 m (38), while the hanging wall zone was opened to a distance of 80 m by the Upper Svinica adit (39) (Szedelmy, 1845; Reuter, 1910). The paragenesis and the forms of the ore bodies are very similar to those in Katarina.

The Gradski potok ore field (workings 27 to 37) is located beneath the Gradina spur round the source and the upper reaches of the brook of the same name and its tributaries, amongst them the Baptista and Nova Nada brooks. On the Gradina spur numerous spoil heaps with limonite ore may be observed, signs of mining operations in Illyrian, Roman and Saxon periods. There is no reliable evidence of attempts to mine silver-bearing tetrahedrites and galena, which are accessory minerals in the Gradski potok ore field, because at that time the miners did not extract copper ore. Gradski potok was the largest copper mine of the Austro-Hungarian monarchy in the nineteenth century (Lipold, 1855). The basic morphological characteristics of the Gradski potok ore fields may be reconstructed from the relatively large number of accounts given by mining engineers and geologists in the eighteenth, nineteenth and twentieth centuries: Wojtanek, 1772; Hacquet, 1789; Szedelmy, 1845; Lipold, 1855; v Zepharovich, 1859; v Andrian, 1868; v Hauer, 1870; Zloch, 1897; Kišpatić & Tućan, 1914; v Papp, 1919; Aleksijević, 1922; Turina, 1933; Tućan, 1941; Nedêla-Devidé, 1953; Jurković, 1962; Šinkovec, (1962) and Braun (1977).

The ore body is concordantly intruded in a series of Carboniferous shales with intercalations of sandstones, grauwackes and subgrauwackes (Nedêla-Devidé, 1953 and Majer 1964) extending generally NW--SE, or alternatively NNW-SSE, with a dip of 45—62° towards SW, or alternatively WSW. Outcrops of the ore body are found at various absolute altitudes on the Gradina spur, because erosion has penetrated deep into the ore-bearing zone in the bed of the Baptista brook, as far as the first horizon level at point + 265 m, while an even broader area or erosion is located in the bed of the Gradski brook, where erosion also extends to the level of the first horizon (sector C). Because of these eroded areas on the Gradina spur, the first phases in the exploitation of the ore took place in separate sectors A, B and D. It was only from the level of horizon I that a coordinated mine was developed.

In the *first phase* of prospecting and opening the Gradski potok mine, which lasted from 1768 to 1832, the mineral reserves were investigated to an absolute altitude of + 265 m, i. e. between 33 and 80 m above the excavation levels, depending on the configuration of the terrain. The extreme NW part of the ore-bearing zone, which was the highest on the site, was opened up by the Glückauf adit (33) at point + 312 m. By means of adits Nova Nada (27) at point + 283 and Upper Baptista at point + 278 and + 285, the zero horizon of Gradski potok was opened to a length of 575 m. The major part of this adit embraced the primary zone with siderite, sulfides and sulfosalts. According to recorded data it was highly productive along 50% of its length. The thickness of the orebearing zone is between 5 and 6 meters, richer in chalcopyrite in its NW part than in the SE part, where the incidence of chalcopyrite declines in favour of pyrite. By means of extended prospecting in depth, the first horizon of the mine was opened up, with adits Upper Gradski (35), Lower Baptista (29) and Lower Breunner (34), all at points on level + 265 m. At this horizon the ore-bearing zone was 540 m in length, extending to the NW fault zone, with a dip to SE, and the SE fault zone, with a dip to the NW, which curtailed the ore-bearing slab wedgewisely.

To that level of the first horizon the Gradski potok deposits were richest in chalcopyrite, and the ore-bearing zone thickest, with a massive »ore shoot«. Before the construction of a second horizon at point + 232 m, the ore was extracted via intermediate horizons, with the adits Lower Gradski (36) at point + 248, and Kempen (32) at + 238, and Kornelija (31) at point + 236. The configuration of the ground in the valley of the Gradski potok brook permitted the economic working of the lowest extraction adit Lower Breunner at point + 236. This adit ran N 40° E, and having traversed the ore-bearing zone at 300 m, opened it by extensions on both flanks to a distance of 490 m, as far as the two major fault zones. At a later period it served as the main adit for the extraction of ore, being linked by 9.4 km of narrow gauge railway with the smelting works in Bešlinac. In the first phase of the development of the Gradski potok mine as described here, the main mining effort was concentrated on prospecting the abundance and the quality of the copper ore, while exploitation was concentrated on the production of limonite and siderite that were poor in chalcopyrite, with a view to obtaining crude iron. There are no data concerning the production of copper ore, and it was on a negligible scale.

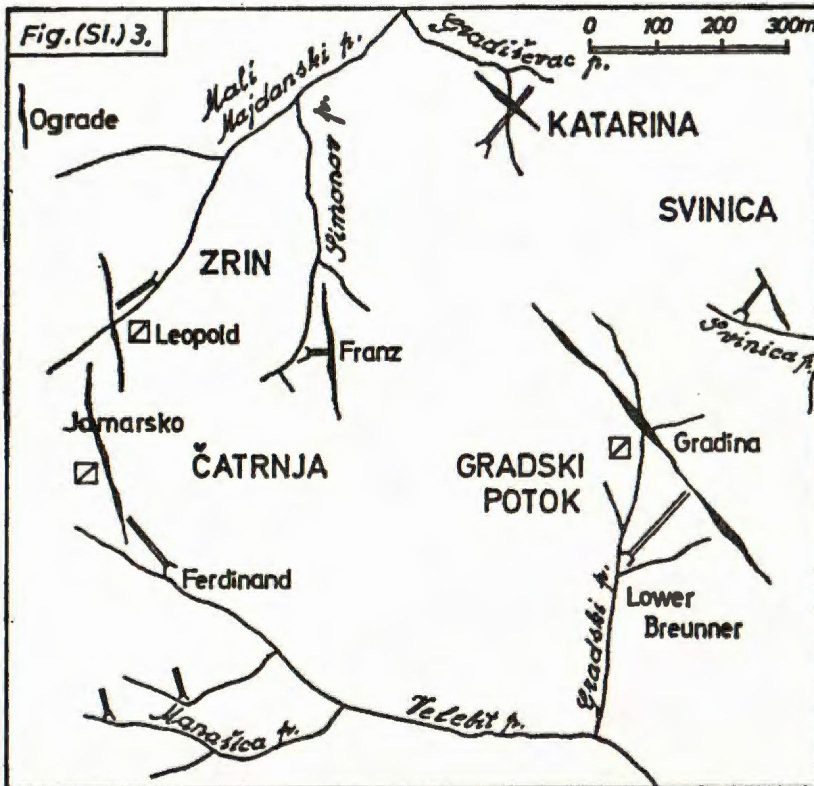
During the s e c o n d p h a s e in the development of the Gradski potok mine, which lasted from 1832 to 1870, copper ore began to be produced from the first and second horizon, particularly during the period from 1832, a copper smelting plant was constructed in Bešlinac (1842), while in 1862 the construction of the Gradina (Emilija) shaft was begun at point + 287 m, in the hanging wall part of the ore-bearing zone. Of the planned 180 m depth of the shaft, 120 m were completed by 1870, as far down as + 167 m, when work was suspended because of the fall in the price of copper in the world market. The Gradina shaft was linked by a lateral galery 110 m long with either the second horizon or the Breunner adit (30) at + 232, as well as by an 80 m long lateral gallery with the fourth level at + 167 m, which ran for 350 m as far as both fault zones. The third horizon at + 187, running 380 m to both faults, was not linked to the shaft, but to the fourth horizon by means of drifts. During the period from 1858 to 1870 the development of the Gradina shaft permitted intensive exploitation of the copper ore. From 1839 to 1858, it was mainly iron ore that was extracted, along with siderites poor in copper minerals, and limonites.

In the t h i r d p h a s e, from 1870 until 1901, the production of copper ore ceased, while on the upper horizons, in the Nova Nada and Upper Baptista adits, where the deposits were most plentiful, the remaining limonite and siderite with low copper content were to some extent extracted. (Z l o c h, 1897).

In a f o u r t h p h a s e, from 1901 until 1914, a Belgian company resumed work in Gradski potok, following a renewed rise in the price of copper. The Gradina shaft was deepened to + 140 m, and reached a total depth of 147 m. The shaft as provided with electricity, and linked by means of a transverse gallery 55 m long with the fifth horizon at 140 m., which ran 260 m. to both fault zones. From this fifth horizon only six short drifts were run downwards to about 15 m. in each case, the negative yield of which led to the cessation of attempts to sink further workings. During this period, siderites rich in chalcopyrite were extracted from horizons IV and V, while on horizons I, II and III, only the SE parts of the NW sector (B) and the hitherto unexploited parts of the A sector in the SE sector (A) were worked. Extraction was carried out by the pillar method.

After 1914 the Gradski potok mine was no longer in production.

Through these mine workings it was established that there were three ore-bearing zones running parallel to each other, with siderite and variable quantities of quartz, sulfides and sulfosalts, amongst which chalcopyrite was the main ore mineral. The lowest stratigraphical zone was the richest and the thickest (up to 3 m.). Its foot-wall salband included an interpolation of almost pure chalcopyrite from 30 to 50 cm. thick on average (up to 1.2 m. in places) of a stratified texture, blending into the central part of this footwall zone consisting of siderite with sprink-



ORE FIELDS ZRIN, ČATRNJA AND GRADSKI POTOK
(Rudna područja Zrin, Čatrnja i Gradski potok)
adit (potkop) shaft (okno)
ore outcrops (izdanci rude)

lings, impregnations and small nests of chalcopyrite. Larger nests of chalcopyrite may also be observed locally in the siderite, while there are also similar nests in the hanging wall and foot-wall shales. In the hanging wall and the foot-wall of the ore-bearing zone, a relatively large number of slender chalcopyrite strata may be seen, several millimeters thick, with local networks of chalcopyrite lamellae up to 4 cm, in thickness (R e u t e r, 1910). This lowest cre-bearing zone is separated from the central ore-bearing zone by an interposed layer of shale of 1.4 m. thickness on average; it is 1—2 m. thick on average and is significantly poorer in ore content. The interspersed deposits of chalcopyrite were 1 to 10 cm. thick. This zone was again separated from the uper ore-bearing zone by an interpolated layer of shale 1.4 m. thick. The hanging wall zone was the thinnest, about 1 m., with low sulfide content, and the least known in terms of its extension. The extension of the entire tripartite zone of mineralisation was NE-SE,with a dip of 45-62° towards SW. The ore-bearing zones on the upper levels were fairly regular and consistent in their extension. As they went deeper they grew progressively thinner and became less regular in the direction in which they ran. Minor faults were encountered, a relatively rapid reduction in thickness with a relatively great increase in the occurrence of quartz and noticeably less chalcopyrite while the productive length was reduced downward at both ends of the mineralisation zone on account of the two fault zones previously mentioned. As a result the ore-bearing surface acquired a trapezoidal shape. Its upper side was 575 m. long at the surface of the Gradina spur, but was of an irregular shape because of the erosion clefts represented by the Gradski brook and the Baptista brook. Its lower surface at the fifth horizon level was significantly shorter, no more than 322 m. From these measurements we obtain a trapezoidal surface of about 90,000 m^2. While on the upper horizons the richer parts of the ore-bearing zone (6.5 % Cu) constituted up to 50 % of the total extent of the ore-bearing zone, on the lower horizons this was reduced to 30 % and even less. According to the classification of Smirnov (1967), the ore field of Gradski potok most nearly approximates in its morphology to »multiple strata with intercalations of country rock, divided into benches separated by intercalation of rock. The benches are, in turn, broken locally into layers.«

QUALITY OF THE ORE FIELD

According to v. Hauer (1870), in the case of under ground workings in the nineteenth century, the Gradski potok ore containing 9.38 % of Cu on average was considered to be of first-class quality. Such ore constituted about 10 % of the excavated ore. Second-class ore with 6.18 % Cu made up 90 % of the ore mined. Accordingly, the average Cu content in the production of that period was 6.55 %. The same author claims that by 1859, 1945 tons of copper ore with 6.77 %, i. e. 131.8 tons of copper had been extracted, and then, down to 1870, 8,600 tons of ore with 6.97 % Cu, i. e. 600 tons of copper. v. Andrian (1868) writes that the richest ore was on the higher horizons, the zero and the first horizon, where a very rich ore shoot (Erzmittel) was discovered; of a total length of 380 m. on the third horizon, only 50 m. contained rich ore, the remaining segments were either sterile, or else poor in chalcopyrite, in place of which an increase in pyrite was found. The third horizon yielded only 1.3 tons of ore per 1 $m.^2$, with the fourth horizon giving no more than 1 ton of ore per 1 $m.^2$ of the ore-bearing zone on average. Reserves were estimated at 4,000 tons, with 7.12 % Cu. It is considered that the zone a few meters above the fifth horizon, as well as the drifts below the horizon are sterile. Lipold (1855) estimated the Gradski potok reserves at 2,400 tons of copper metal, considering it to be at that time the largest copper mine in the Austro-Hungarian domains. Reuter (1910) gives an analysis of the rich and the poorer ores from Gradski potok. The rich ore contains: 7.10 % SiO_2, 8.70 % Cu, 9.63 % S, 35.60 % Fe, 0.84 % CaO, 0.21 % MgO, 2.30 % Al_2O_3, 1.55 % Mn, and 34.07 % CO_2 and ignition lost. The low-grade ore contained 16.85 % SiO_2, 3.88 % Cu, 3.42 % S, 33.80 % Fe, 1.70 % CaO, 0.30 % MgO, 2.66 % Al_2O_3, 1.21 %

Mn, and 36.10 % CO_2 and ignition lost. Jurković (1962) gives an analysis of the composition of the copper siderite ore found on the mine's main spoil heap: 15.40 % SiO_2, 3.77 % Cu, 3.45 % S, 31.50 % Fe, 0.55 % CaO, 1.50 % MgO, 0.44 % Al_2O_3, 1.12 % MnO, 0.33 % Pb, 0.03 % P, strong traces of Ni, traces of Co. This composition would correspond to the so-called low-grade ore of the earlier periods of exploitation. Analyses of the copper content from the Nova Nada (27) spoil heap were also carried out, with the following results: 6.51 % Cu and 9.41 % Cu, while samples from the Breunner adit yielded 2.14 % Cu and 3.67 % Cu. Particularly interesting were the results of an analysis of siderite ore containing very little sulfides such as was formerly used as an iron ore in the Gradski potok output: 6.6—10.0 % SiO_2, 0.4—0.56 % Cu, 0.1—0.2 % S, 37—38 % Fe, 0.6—0.8 % CaO, 0.3—0.5 % MgO, 1.5—3.0 % Al_2O_3, 1.17—1.24 % MnO, 0.02—0.03 % P, 27—29 % ignition lost. After roasting the same ore yielded 17.7 % SiO_2 and 43.6 % Fe. According to v. Papp (1919), output in 1909 amounted to 2,086 tons of copper, and in 1912 to 229 tons of copper.

PARAGENESIS OF THE COPPER DEPOSITS

The majority of earlier investigations quote only the basic characteristics of the mineral composition of the copper deposits. Lipold (1855), for instance, considers the deposits in the wider Gradski potok area to be siderite deposits, with chalcopyrite as the principal ore mineral; v. Andrian (1868) writes that the main ore minerals are siderite and chalcopyrite, with quartz as the principal gangue mineral. In the poorer sectors of the deposits, pyrite prevailed over chalcopyrite. There is little galena in the paragenesis. v. Hauer (1870) enumerates in the paragenesis siderite, chalcopyrite, quartz and a little galena and pyrite. Reuter (1910) writes that, apart from siderite, chalcopyrite, pyrite and quartz, the paragenesis involves bornite.

Only a few researchers write in more detail of a paragenesis established by the naked eye: on the spoil heap of the Kornelija adit and other prospecting sites for copper ore Hacquet (1789) detected: chalcopyrite (minera cupri cinerea) in quartz with a small amount of schists, a brown copper ore (minera cupri cinerea) in quartz, occasionally in calcite and siderite, less frequently »Leberkupferkies« (pyrite cupri) in schists. Even rarer was »minera cupri versicolorata« or a mixture of various copper minerals. v. Zepharovich (1859) provided the most detailed account of the Gradski potok minerals: bornite as dense masses in association with chalcopyrite, quartz, calcite and malachite, then chalcopyrite in drusy spaces of siderite or else as close-grained masses, or sprinklings in siderite, locally in association with malachite and »Ziegelerz«. He alo refers to the presence of cuprite with ochre and malachite in its cavities, as well as elemental copper in the form of fine films on the cuprous materials. As for siderite, he says that it is fine- to coarsegrained and contains crystals or sprinklings of galena, pyrite, chalcopyrite, talc or dense quartz. In the drusy spaces of the siderite he observed small, flat rhombohedrals of siderite, isolated or polysynthetically associated. He writes that the talc is foliate, silvery white in colour in the drusy spaces of siderite, together with other minerals, amongst which there is also foliate or fine crystalline pyrolusite. Among the primary minerals Kišpatić (1901) enumerates siderite, chalcopyrite, bornite, tetrahedrite, sphalerite, pyrite, gersdorffite, calcite. He gives the following composition for the beautiful crystalline gersdorffite on the basis of a chemical analysis: $(Ni,Fe,Co)_{12}As_{10}S_{11}$. Amongst hypergenic minerals he notes covellite (as a coating on chalcopyrite), cuprite of a dark red colour, with minute needles or coatings of malachite, as well as »limonite« of kidney-shaped, clustered, stalactitic or fibroid structure.

MICROSCOPIC EXAMINATION OF THE PARAGENESIS

The first microscopic analysis of ore samples from the Gradski potok deposits was provided by Jurković (1962). In this paper we give a detailed account of examinations of thin ground and polished sections with the microphysiographic characteristics of all the hypogenic and hypergenic minerals identified, along with an account of their succession in the course of mineralisation.

Paragenesis

Hypogenic minerals: siderite I, pyrite, quartz, gersdorffite, millerite, sphalerite I, chalcopyrite I, chalcopyrite II, sphalerite II, bornite, tetrahedrite, bravoite, galena, calcite.

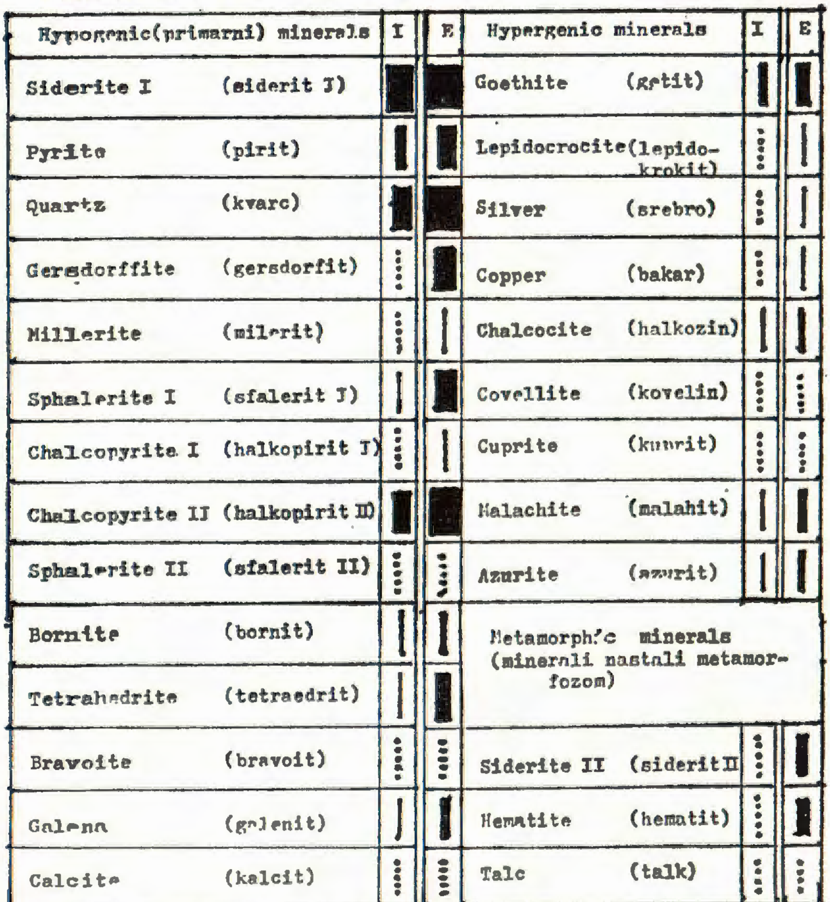
Hypergenic minerals: goethite, lepidocrocite, elemental copper, elemental silver, chalcocite, covellite, cuprite, malachite, azurite.

Metamorphic minerals: siderite II, hematite, talc.

THE MICROPHYSIOGRAPHY OF HYPOGENIC MINERALS

Siderite I is the main mineral of hypogenic paragenesis in the ore deposits of the Gradski potok region. Its structure ranges from finegrained to coarsegrained. The cleavage is distinctly rhombohedral. It is silicified to varying degrees of intensity, silification taking place along the plane of cleavage, at micro cracks, or in intergranular spaces. Many samples of siderite are of brown or reddish brown colour, so-called »Braunspat«, which display red and reddish brown internal reflections with crossed nicols, arising from sub-microscopically fine particles of hematite dust. At the lower levels of the Gradski potok deposits, oxidation is barely perceptible, and is visible then along the rhombohedral plane of cleavage or at cracks, while in the upper parts of the deposits it is total. According to Tućan (1907), a quantitative analysis of siderite from the Katarina mine give the following results: 59.60 % FeO, 0.87 % CaO, 0.10 % MgO, 0.87 % MnO, 37.73 % CO_2, 0.78 % insoluble residue and 0.20 % Al_2O_3, which on recalculation indicates the following constitution for siderite: 96.75 % $FeCO_3$, 1.55 % $CaCO_3$, 1.48 % $MnCO_3$, 0.21 % $MgCO_3$. An analysis of coarsegrained siderite from Gradski potok gave the following results: 57.61 % FeO, 0.79 %

Intensity(I) and Extensiveness(E) of hypogenic and hypergenic minerals in the Gradski potok brook ore district of Trgovska gora



Fig(sl.)4. Intenzitet(I) i ekstenzitet(E) pojavljivanja minerala u paragenezama rudnih pojava područja Gradski potok

CaO, a trace of MgO, 1.15 % MnO, 36.39 % CO_2, 3.57 % insoluble in HCl, 0.31 % Al_2O_3, which indicates the following constitution on recalculation: 96.03 % $FeCO_3$, 1.98 % $CaCO_3$, 1.98 % $MnCO_3$, and a trace of $MgCO_3$.

Q u a r t z is an important constituent of these ore deposits. There is 5 to 15 % on average, but on the lower horizons, with attenuated ore-bearing zones, there is considerably more, to the point where it matches the quantity of siderite. It replaces siderite in the form of systems of relatively thinner or thicker veins, nests and small masses of varying dimensions. Quartz in the form of veins is typimilky white in colour, fine to moderately grained in structure. It is associated with sulfides and sulfosalts. Corroded grains of quartz may be observed in chalcopyrite. Palinkaš (1988), investigating inclusions in quartz from Gradski potok, established that twin phase inclusions of a very irregular shape (L + V) were predominant, while single phase inclusions (L) played a subordinate part. Low temperature research gave T_{FM} = —20 °C, which indicates an almost pure NaCl—H_2O system. The salinity varies from 0 % to 11 % NaCl ekv. with a maximum around 8 %. By homogenising samples it was possible to obtain a wide distribution, with two maximum values of + 150° and 250 °C. There was no indication of boiling.

Pyrite is a subordinate ore mineral in the richer sectors of the deposits, but there is more of it in the SE parts of the ore-bearing zones, as well as on the lower horizons. It is more often present in the attenuated parts of the ore-bearing zones, where larger amounts of quartz make their appearance at the expense of siderite, and the amount of chalcopyrite diminishes. As regards the paragenesis of Gradski potok overall, pyrite is the most abundant sulfide after chalcopyrite. We observe it in the form of corroded grains in siderite, chalcopyrite and quartz, but also as individual idioblasts of hexahedral habit, or alternatively as clusters of such crystals. Many idioblasts (porphyroblasts) of pyrite are enveloped in aligned incrustations of gersdorffite. In some pyrite crystals bravoite even selectively replaces certain zones or segments of the crystals, particularly at the edges, which indicates the position of Ni minerals in the sequence of mineralisation. Chalcopyrite replaces pyrite enveloped in gersdorffite incrustations, forming insular structures. The larger aggregations of the more coarsely crystallised pyrite are cataclased, the cataclases being cemented by quartz and here and there by sphalerite.

Gersdorffite was first observed with the naked eye by Kišpatić (1901) as small crystals of 1 to 2 mm. On the basis of chemical analysis he worked out the formula $(Ni,Fe,Co)_{12}As_{10}S_{11}$. Through microscopic examination we established that gersdorffite is a very prevalent mineral, we found it in almost all the specimens examined, but always in microscopic quantities. It occurs either in the form of idiomorphically developed small crystals with a diameter ranging from several micrometers up to 50 micrometers, or else in the shape of relatively coarse grains, the largest of them measuring 1 mm × 0.5 mm. All the larger crystals are very finely structured in zones, these zones being numerous and varying with minute differences in outline and in colour tones. At points individual zones or segments of the crystals have been selectively replaced by chalcopyrite of more recent origin. In this way, skeletal forms of gersdorffite arise within chalcopyrite. We also observed pseudomorphosis of chalcopyrite and galena on gersdorffite. In some crystals alfa and beta components may be detected which differ mainly in configuration and in slight discrepancies of colour and brilliance. According to Ramdohr (1983, p. 836) the brighter and harder component is the alpha variety, while the darker and softer component is the beta modification. Gersdorffite evolves symmetrically in the form of incrustations on larger pyrite idioblasts, these accrued crystals have very regular outer surfaces of octohedral habit. At points gersdorffite embodies clusters of pyrite grains, especially when it is situated in siderite. A characteristic phenomenon is the accretion of sphalerite on gersdorffite, which leads to the conclusion that gersdorffite is older than sphalerite I in the sequence of mineralisation. In various specimens we observed worm-like residues, ribbonshaped clusters and isolated structures of gersdorffite in chalcopyrite, in association with corroded grains of pyrite and quartz. The coarser grains and crystals of gersdorffite are fissured, and the cataclases are filled with quartz and chalcopyrite. A cleavage of (100) was observed. The relief of gersdorffite is considerably lower than the relief of pyrite, but higher than that of siderite and chalcopyrite. In reflected light it is white in colour, with a creamy hue. Its lustre is greater than of chalcopyrite, but rather less than that of pyrite. In a number of specimens it was associated with millerite.

Sphalerite I is of marmatite type, its internal reflections are brown or reddish brown. Quantitatively, it is a accessory component in the deposits, but it is found in more than 80 % of the specimens, which indicates the significant extensiveness. A basic characteristic of sphalerite I is the very common occurrence of sub-microscopic and microscopic droplets (blebs) of chalcopyrite in the central parts of masses of sphalerite I, especially in the larger masses. Sphalerite I is very unevenly distributed in the individual specimens examined, in some very thinly scattered, very small masses of sphalerite I may be seen, in other specimens there are very numerous, but predominantly microscopic small masses. Sphalerite I can rarely be observed with the naked eye. It is associated with tetrahedrite and galena. At points it adheres to gersdorffite, enveloping it. It cements cataclases in pyrite and gersdorffite.

Chalcopyrite I is found exclusively in the form of exsolutions in sphalerite I, in its central parts, which indicates that these are in fact exolutions and not generated by subsequent diffusion from the surrounding chalcopyrite. These exsolutions are on a microscopic scale, at the limit of visibility with large magnification.

Sphalerite II is observed in a smaller number of specimens as small starlets in chalcopyrite II. These exsolutions suggest a relatively high temperature for their formation.

Chalcopyrite II is the principal sulfide mineral of the Gradski potok ore field. The largest amounts of this mineral are located at the foot-wall interface of the lowest ore zone between siderite and shale, where it forms almost pure compact masses of 30—50 cm. in size, there is considerably less of it within siderite, where it is in the shape of impregnations, small nests or irregular masses of varying size, replacing siderite, or even as larger nests in the hanging wall and foot-wall salband of shale. At the salbands in the central and hanging wall ore-bearing zone there are significantly smaller amounts of chalcopyrite, and siderite itself is also rarer on chalcopyrite II. As a younger mineral it replaces siderite, pyrite, quartz, gersdorffite, sphalerite I, it is deposited almost at the same time as tetrahedrite, while it is older than galena, which it replaces. In chalcopyrite we observe skeletal, worm-like forms, or corroded grains of gersdorffite, corroded grains of pyrite, minutely small masses, which minerals are subject to replacement in varying degrees. At certain points it selectively replaces gersdorffite, and there are also pseudomorphoses of chalcopyrite II on gersdorffite. At the edges of small masses of chalcopyrite II, and sometimes within it, millerite of a granular structure may be seen. Larger masses of chalcopyrite are regularly in association with tetrahedrite, or embody it, while they also enclose small masses of sphalerite I with exsolutions of chalcopyrite I. In places the chalcopyrite II is crumbled into sharp-edged fragments. It is changed in various degrees of intensity into hypergenic minerals by processes of oxidation and cementation. Amongst oxidation minerals we detected goethite, malachite, cuprite, and among cementation minerals were chalcocite, covellite and elemental copper. In cataclased parts of chalcopyrite II, microscopically minute masses of elemental silver were observed. The weathering of chalcopyrite II proceeds by planes (111) or along systems of very fine cracks, forming networks of microveins of broken down products, of which the commonest are goethite and descendant, low-temperature chalcocite. As they proceed, these veins coalesce, forming irregular small masses of secondary minerals. At places minute leaves of covellite may be seen along (111) planes of chalcopyrite II.

M i l l e r i t e is intimately associated with chalcopyrite II, we find it on the edges of small masses of chalcopyrite but also corroded inside it. It is microgranular in structure, the grains being allotriomorphic. Its contur is rather higher than that of chalcopyrite II, its lustre is also somewhat greater, it is yellow in the reflected light of the polarised microscope. It has distinct reflective pleochroism and powerful anisotropic effects. It is rare in the ore bed, observed in a few specimens only.

B o r n i t e was first noticed by v. Z e p h a r o v i c h (1859), who observed it with the naked eye in the upper sectors of the Gradski potok ore-field (August), as dense masses associated with chalcopyrite II, quartz, calcite and malachite. Later on, K i š p a t i ć (1901) refers to the presence of bornite in association with siderite, chalcopyrite, tetrahedrite, sphalerite, pyrite, gersdorffite and calcite. In specimens of ore from Gradski potok and in polished sections prepared from them we were unable to identify bornite.

T e t r a h e d r i t e is found in half of the specimens examined. It is small in quantity, and is mainly linked with chalcopyrite II. It may be observed with the naked eye, and by microscope as minute masses of microscopic dimensions. It is partially weathered into the secondary minerals chalcocite and goethite, less frequently into lepidocrocite and covellite, while there is also malachite and very rarely azurite. Weathering is reticular, evolves along microcracks or octohedral plates (111), this applying especially to microplates of covellite.

G a l e n a is a relatively rare sulfide in the ore filed and very subordinate in amount. It is silver-bearing. It is the youngest mineral in the sequence, replaces all the other minerals, especially chalcopyrite II, which is the most abundant sulfide in Gradski potok. We observe it in the form of relatively large, irregular masses and nests, while pseudomorphoses were observed on gersdorffite. It was found in half of the specimens examined.

Bravoite was observed in one prepared sample from Gradski potok. It replaces porphyroblasts of pyrite of a hexahedral habit, along its edges or along certain zones of zonal crystals of pyrite, or even in certain parts of pyrite crystals. It has the customary optical features.

MICROPHYSIOGRAPHY OF HYPERGENIC MINERALS

Goethite is the most abundant hypergenic mineral. In the superficial sectors of the Gradski potok deposits, all the siderite and all the iron-bearing sulfides and sulfosalts (pyrite, chalcopyrite, sphalerite, tetrahedrite, bravoite) break down totally or partially into goethite, and a limonite ore is formed which was the object of mining operations with the aim of producing crude iron. At the lower horizons, oxidation has only partly affected the hypogenic minerals. The oxidation of siderite, which is the main mineral in the ore field, develops along the rhombohedral planes of cleavage, in networks of cracks and in relatively rare intergranular spaces. In the chalcopyrite II masses, goethite is associated with descendant chalcocite, and where there are visible accumulations of pyrite and lepidocrocite. Goethite is crypto- or microcrystalline, of frequently colloidal structure. In the oxidation belt of the limonite mass there are kidney-shaped, clustered and stalactitic structures formed from small fibrous crystals.

Lepidocrocite is predominantly a product of the oxidation of pyrite and is very much subordinate to goethite.

Malachite and azurite are mainly visible in the superficial parts of the deposits, with malachite being considerably commoner and more abundant than azurite, which arises mainly from the weathering of tetrahedrite, while malachite is formed from chalcopyrite II. Malachite is clustered, kidney-shaped, in the form of films and coatings. Its internal structure is fibrous. Azurite is observed only as a very fine coating.

Cuprite was first noticed by v. Zepharovich (l. c.) and his data were quoted by Kišpatić (l. c.). Seen with the naked eye, it consists of dark brownish-red small masses in association with »ochre« and malachite. Seen under the microscope, cuprite is fine-grained and is distinguished by bright bloodred internal reflections.

Elemental copper is found only in association with cuprite, characteristic in its colour in the reflected light of the microscope.

Chalcocite is most frequently found in the cementation zone, where it is deposited directly on chalcopyrite or tetrahedrite as a margin of varying thickness. This exceptionally fine-grained chalcocite is transformed by subsequent oxidation into cuprite, malachite and elemental copper, and mixes with goethite. Chalcocite also arises along cracks in chalcopyrite and tetrahedrite, along with goethite, weathering relatively rapidly into tetrahedrite. Here and there along the octohedral plane of these two minerals chalcocite is associated with the occurrence of covellite in the form of microscopically minute plates.

Covellite is very rare, observed in chalcopyrite II and tetrahedrite affected by cementation processes, in which chalcocite and covellite are produced by the reduction of descendant Cu-sulfate. It is developed as small plates with characteristic optical qualities.

Elemental silver is observed in several specimens, i. e. in the cataclased segments of siderite I, or else of chalcopyrite II, as microscopically minute small masses of very high metallic lustre, creamy yellow in colour. It arose by extrusion from silver-bearing galena, but possibly also from silver-bearing tetrahedrite.

Pyrolusite, foliate or micro-crystalline, is very uncommon.

THE MICROPHYSIOGRAPHY OF MINERALS PRODUCED BY ADDITIONAL METAMORPHIC CHANGES

Siderite II is micro-granular, arising through the precrystallisation of siderite I in tectonically disturbed parts of the ore bed.

Hematite is found in siderite I in the form of submicroscopically fine dust (»dust-like inclusions of hematite«), which gives the siderite the appearance of so-called »red spar or Braunspat«. Such siderite displays internal reflections of a brownish-red or dark red colour.

Talc was described by v. Zepharovich (1859) as a mineral of foliate texture, silvery white in colour on siderite or in the drusy spaces of siderite, in association with other sulfides and quartzes. In the course of our microscopic investigations and examination of the specimens we did not detect any talc.

SEQUENCE OF MINERALISATION

The first phase of mineralisation is also the major phase, in which two or three sub-parallel siderite zones were formed. Masses of siderite are clearly crystallised out, the structure ranging from fine-grained to coarse-grained. An occasional corroded grain of quartz and pyrite in the siderite suggests the possibility that a very small amount of these two minerals was secreted in this phase. Low manganese content in the siderite (from 0.9 to 1.2 % MnO) indicates the probable epithermal character of this main phase of mineralisation.

The second phase of mineralisation has a rejuvenative character. It begins with a mesothermal first sub-phase, the basic characteristic of which is represented by a process of silification with mesothermal quartz, or so-called veined quartz, for which Palinkaš (l. c.) gives a formation temperature of +250 °C. Silification is accompanied by coarse crystalline pyrite I and gersdorffite typical of these mesothermal conditions, along with very small quantities of sphalerite I and with a clear exsolution of chalcopyrite I in the central parts of the sphalerite crystals. The succeeding second sub-phase is characterised by considerable crystallisation of chalcopyrite II of a mesothermal character, as indicated by the star-like exsolutions of sphalerite II with a little tetrahedrite and bornite, and very rarely allotriomorphically grained millerite. In this sub-phase crystallisation of quartz continues right down to a temperature of +150 °C. Towards the end of the mineralisation process a small quantity of silver-bearing galena crystallises, the replacement of pyrite with bravoite begins.

Because of tectonic stresses in the course of the more recent orogenic phase, certain optical anomalies in the hypogenic minerals were caused, cataclasing, recrystallisation of relatively small quantities of siderite and siderite II, hematitisation in siderite I and its conversion into »red spar«.

In the younger Tertiary period goethite is formed through oxidation processes, while goethite, lepidocrocite, malachite, azurite and pyrolusite are produced by the oxidation of sulfides and sulfosalts. In the cementation zone chalcocite and covellite are produced from descendant sulfate solutions, and through their oxidation elemental copper, cuprite, elemental silver, malachite, azurite, goethite and lepidocrocite are produced.

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Bakarna rudna ležišta u rudnom području Gradski potok Trgovske gore u Hrvatskoj

I. Jurković

U gornjim tokovima potoka Dragiševac, Svinica i Gradski potok nalaze se bakarna ležišta Katarina, Svinica i Gradski potok. Područje je izgrađeno od gornjopaleozojskih šejlova s tanjim ili debljim proslojcima grauvaka, subgrauvaka i drugih tipova pješčenjaka.

Orudnjenje je konkordantno uloženo u sedimentnu seriju. Opće pružanje sedimenata i mineralizacije je NW—SE s padom ka SW. Rudne pojave imaju oblike slojevitih leća promjenljive debljine i kvalitete. U rudištima Katarina i Svinica postoje dva orudnjena horizonta, a u Gradskom potoku tri. U Katarini donji, bogatiji horizont praćen je na dužini od 240 m, gornji na dužini od 120 m. U rudištu Svinica donja zona je bila praćena na dužini od 140 m, a gornja samo na 80 m. Rudište Gradski potok je znatno veće i bogatije. Istraženo je do dubine od 147 m vertikalne visine s oknom Gradina koje je istražni prostor povezalo s pet horizonata i izvoznim potkopom Donji Breunner. Orudnjena površina imala je oblik kosog trapezoida nagnutog pod kutem od 45—62° ka jugozapadu, s gornjom stranicom dužine 575 m i donjom dugom 322 m. Bočne stranice trapezoida su ograničene s dva vrlo jaka rasjeda suprotnog smjera pada. Iza tih prelomnica nije nađen nastavak orudnjenja. Mineralizacija je bila najbogatija u prva tri horizonta, prema dubini kvalitet je slabio, ruda je sadržavala sve manje bakarne rude, a sve više kvarca i pirita. Najbogatiji je bio u stratigrafskom smislu najdonji nivo orudnjenja, debeo do 3 m, koji je u donjem salbandu sadržavao 0,3 do 0,5 m debeo masivni, kompaktni halkopirit prelazeći prema gore u glavni dio izgrađen od siderita s 5—15 % kvarca te uprskanja gnijezda i žilica halkopirita, pirita, malo sfalerita, tetraedrita i vrlo rijetko galenita. Donji horizont bio je odijeljen od srednjeg, tanjeg (1—2 m), slabije orudnjenog s 1,4 m debelim proslojkom šejla. Nakon ponovnog uloška od 1,4 m šejla razvio se gornji rudonosni horizont oko 1 m

debeo s čestim istanjivanjem, slabo orudnjen sulfidima. U sva tri horizonta uočeni su vrlo brojni epigenetski tektonski poremećaji. Oksidaciona zona rudišta bila je predmet eksploatacije na kvalitetan limonit zbog proizvodnje sirovog željeza u topionici u selu Bešlincu, a primarna zona ili na željezo u dijelovima koji su sadržavali siderit s vrlo malo sulfida ili na bakarnu rudu u bogatijim dijelovima ležišta kojih je bilo do 50 % prostiranja ležišta u gornjim horizontima odnosno 15—30 % u donjim dijelovima ležišta.

U periodu najintenzivnije eksploatacije bakarne rude (od 1838 do 1870 te od 1901 do 1913 god.) prosječan sadržaj rovne rude bio je 6,55 % Cu, a sideritne rude za proizvodnju željeza 37—38 % Fe i 0,4—0,6 % Cu.

Detaljnim mikroskopskim istraživanjem utvrđena je ova parageneza: (a) hipogeni minerali: siderit I, pirit, kvarc, gersdorfit, milerit, sfalerit I, halkopirit I, halkopirit III, sfalerit II, bornit, tetraedrit, bravoit, galenit i kalcit; (b) hipergeni minerali: getit, lepidokrokit, elementarni bakar, elementarno srebro, halkozin, kovelin, kuprit, malahit i azurit; (c) metamorfni minerali: siderit II, hematit.

U prvoj fazi mineralizacije koja je ujedno i glavna faza, kristalizirao je srednje do krupnozrnati siderit s niskim sadržajem mangana. U drugoj fazi prva podfaza je karakterizirana procesom silifikacije siderita, odlaganjem pirita i gersdorfita te vrlo malih količina sfalerita s izdvajanjem haskopirita I u središnjim dijelovima zrna sfalerita. U slijedećoj podfazi kristalizira halkopirit II, glavni rudni mineral rudišta bakra u tom području, s malo sfalerita II, bornita, milerita i tetraedrita. Na kraju procesa mineralizacije javljaju se male količine srebronosnog galenita. U toku narednih orogenih faza dolazi do naprezanja u kristalima, kataklaziranja, djelomične rekristalizacije te hematitizacije siderita (red spar ili Branuspat). U mlađem tercijeru razvili su se u najgornjim dijelovima sva tri ležišta snažni procesi oksidacije i u manjoj mjeri procesi cementacije.