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SULPHOSALTS OF CUPROBISMUTITE HOMOLOGOUS SERIES FROM BANSKÁ HODRUŠA AND ČIERNA LEHOTA (WESTERN CARPATHIANS, SLOVAKIA)

S. Jeleň[1], T. Mikuš[1], J. Pršek[2]

[1]*Geological Institute of Slovak Academy of Sciences*
Ďumbierska 1, 974 01 Banská Bystrica, Slovakia
[2]*Department of Mineralogy and Petrology, Faculty of Natural Sciences,*
Comenius University
Mlynská dolina, 84215 Bratislava, Slovakia

Relatively rare kupčíkite was found together with other members of cuprobismutite homologous series – hodrushite and cuprobismutite. They occur in hydrothermal Ni-Bi-As mineralization in Čierna Lehota and at base and precious metal mineralization deposit Banská Hodruša, Rozália Vein. Kupčíkite is associated with cuprobismutite, hodrushite and members of bismuthinite-aikinite series, emplectite, paděraite, gold of higher fineness. It was studied chemical composition of all associated Bi-sulphosalts.

Key words: Bi-sulphosalts, kupčíkite, chemical composition, hydrothermal mineralization, gold, Western Carpathians, Slovakia.

Hodrushite – Cu-Bi sulphosalt of the cuprobismutite homologous series – is the first latter-day described mineral from Slovakia, found at Rozália Vein by M. Koděra. It was identified and detailed characterized by V. Kupčík and E. Makovický and named after village Banská Hodruša. It forms needles up to 1 mm in size, rarely 5 mm of gray-black colour. Luster is metallic. It is brittle, without distinct cleavage, monoclinic, space group A2/*m* and cell dimensions: a = 27,21; b = 3,93; c = 17,58; β = 92°09′ [6]. Its density is about 6,35. It forms mostly aggregates and irregular overgrowth with aikinite, wittichenite, matildite, emplectite and other Bi minerals in the quartz-hematite-chalcopyrite accumulations. Later was this mineral found in ores of polymetallic deposit Kajragač, Kuramin Mts., Uzbekistan, in Pioche (Nevada, USA), at the locality Čierna Lehota [17] and at the scheelite deposit Felbertal, Austria [21]. The next occurrences are known from Romania (Paulus mine, Ocna de Fier by Baita Bihor and Valea Seaca [1]), from USA, Nevada (Black Metal mine), Colorado (Alice mine, Campbell mine, Bisbee mine) and also from Peru–Julcani ore field [22].

Sulphosalts of cuprobismutite homologous series include Cu-Bi sulphosalts. General formula for minerals of cuprobismutite group was postulated by [10] as

$$Cu_4Me_{2(N-1)+1}{}^{\text{quasi-octahedral}}Bi_4{}^{\text{square-pyramidal}}S_{2N+8}\ (Z=2),$$

which could be simplify written as $Cu_8Bi_{4N-2}S_{16+4N}$. Members of cuprobismutite homologous series are: N = 1 kupčíkite ($Cu_8Bi_{10}S_{20}$) discovered in the Felbertal D. Topa et al [21], N = 1,5 hodrushite $Cu_8Bi_{12}S_{22}$ M. Koděra et al. [6] and N = 2 cuprobismutite $Cu_8Bi_{14}S_{24}$ T. Ozawa and W. Nowacki [15]. Some authors assigns also paděraite $Ag_{1,2}Cu_{5,8}Pb_{1,3}Bi_{11,5}S_{22}$ to this series [14]. However, paděrait belongs to special paděraite homologous series derived from cuprobismutite homologous series according to latest result [1]. Structure of cuprobismutite homologous series sulphosalts could be explained as overgrowth of galena structure $(331)_{PbS}$ with layer of trigonal coordinated metals [10]. All published analyses show except main elements Cu, Bi, S, also Pb, Ag and Fe, rarely also Se, Zn, Mn, Te. Sulphosalts of cuprobismutite homologous series are relatively rare in the nature. They occur mainly in Cu skarns in Romania [1, 2, 8, 14] and also in miscellaneous hydrothermal mineralisation [3, 16, 17, 20, 21]. Sulphosalts of this homologous series was found in Slovakia only in Hodruša and Čierna Lehota [6, 8, 17]. Comparison of these two localities is the subject of this paper.

Banská Hodruša, Rozália Vein. The largest and most famous epithermal gold-silver-polymetalic mineralization is situated in the Štiavnica-Hodruša ore field in the central upwelled part of the vast polygenetic Štiavnica stratovolcano (fig. 1) extending 100 sq. km.

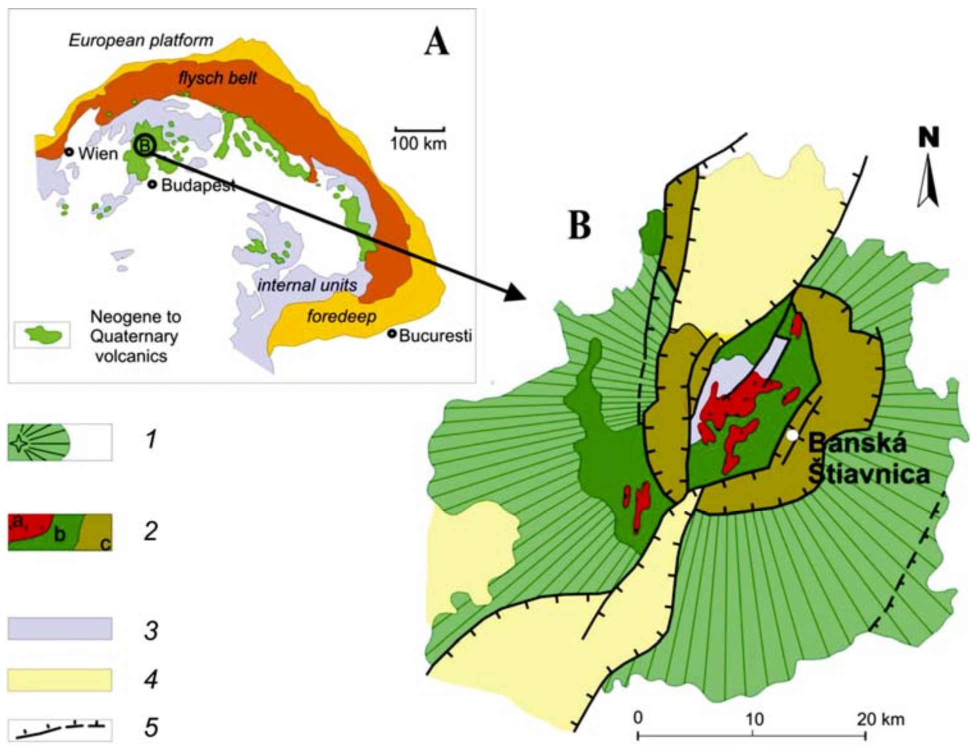


Fig. 1. Position of the Western Carpathian neovolcanites (*A*) and scheme of the Štiavnica stratovolcano (*B*), adapted from [9]:

1 – centre and cone of andesite stratovolcano; *2* – central zone of stratovolcano: *a* – intrusions; *b* – propylitized andesites; *c* – caldera and graben filling; *3* – prevolcanic basement; *4* – postvolcanic sediments; *5* – faults.

More than 120 ore veins occur mainly in propylitized pyroxene andesites, diorites and quartz diorite porphyries. The length of main ore veins is up to 8 km. Rozália Vein belongs to main polymetalic veins of “štiavnica type” [4]. The depth of ore mineralization reaches 500 m. Ores are concentrated into 6 vertical lenses (shots). Whole Rozália vein belong to cooper zone (Cu prevailing over Pb and Zn; fig. 2).

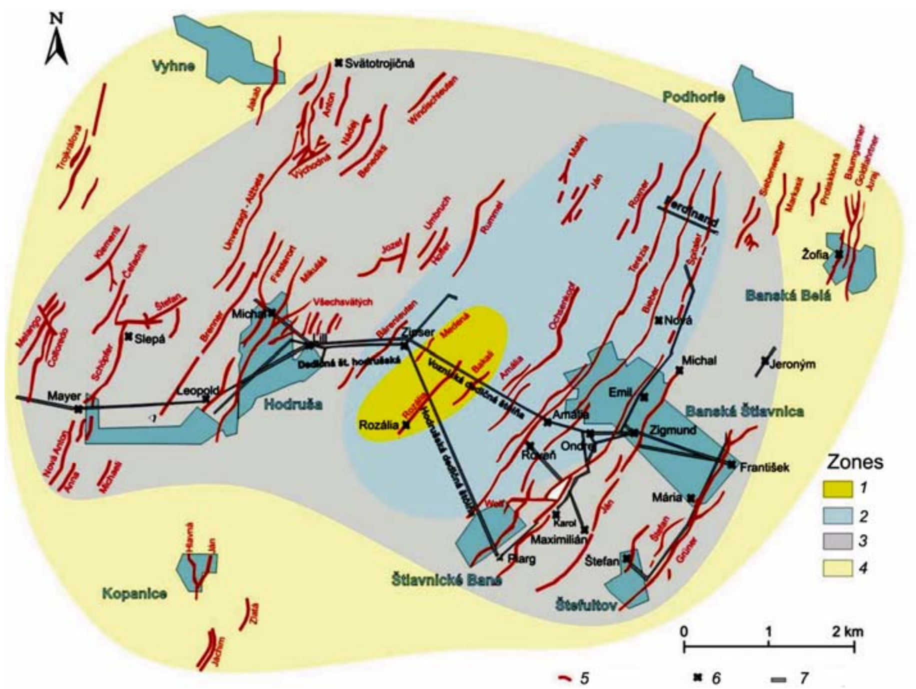
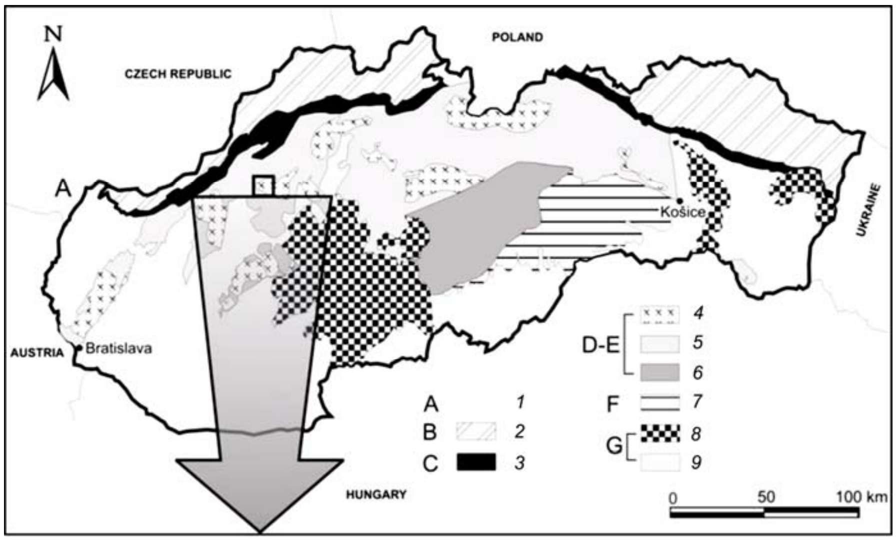


Fig. 2. Horizontal zoning of the Štiavnica-Hodruša ore field:
1–4 – zones: *1* – Cu±Bi, W, *2* – Pb-Zn-Cu, *3* – Ag-Au, *4* – Au-Ag; *5* – ore veins; *6* – adits; *7* – mines.

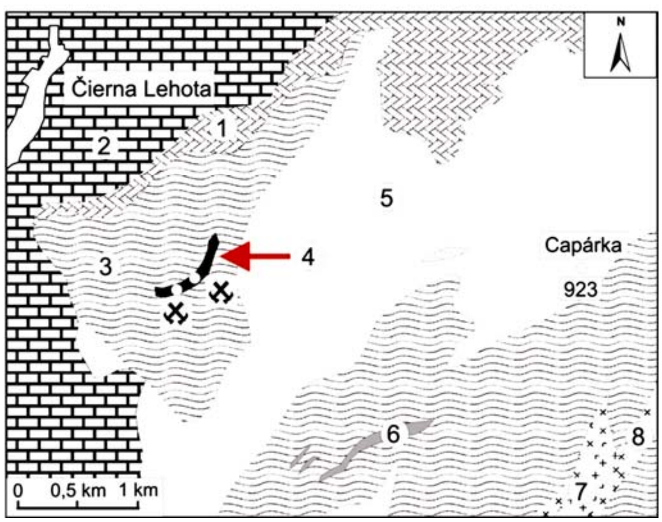
Succession of mineralization was detailed characterized by M. Koděra [4]. Mineralization of Rozália Vein (deeper parts) is characterized by presence of bismuth minerals. Typomorphous mineral is hodrushite which occurs in the quartz-hematite-chalcopyrite aggregates. Hodrushite together with aikinite and wittichenite belong to most prevalent Bi-sulphosalt of quartz-hematite-chalcopyrite aggregates of the fourth mineralization stage. Gold of higher purity (860) is associated together with Bi-sulphosalts: emplectite $CuBiS_2$, matildite $AgBiS_2$, hodrushite $Cu_8Bi_{12}S_{22}$ and paděraite $Cu_{6.41}Ag_{0.73}Pb_{1.24}Bi_{10..99}S_{21.72}$. In the upper parts of the Rozália Vein are characteristic CuBi- and CuPbBi-sulphosalts (gladite $CuPbBi_5S_9$, krupkaite $CuPbBi_3S_6$, hammarite $Cu_2Pb_2Bi_4S_9$, lindströmite $Cu_3Pb_3Bi_7S_{15}$, friedrichite $Cu_5Pb_5Bi_7S_{18}$ and new mineral phase similar to mummeite $(Cu,Ag)_4PbBi_6S_{12}$). Bismuthinite Bi_2S_3 and solid-solution of PbS with matildite myrmecites originated in the lower parts of the Rozália Vein (9^{th} to 14^{th} deep horizon) by the higher temperature. Ag-sulphosats (polybázite, pearceite and Ag-tetrahedrite) occur in the southern subsurface part of Rozália Vein associated with carbonates and quartz.

Čierna Lehota. Ni-Bi-As assemblages described in the Čierna Lehota occur in the Tatric tectonic unit of the crystalline basement of Suchý Mts (Western Carpathians). The investigated mineralization is situated approximately 2 km SSE from the village of Čierna Lehota (fig. 3). Mineralization is situated in a narrow zone (20 m, SSW–NNE direction) of black shales and intensely graphitized rocks (graphitic-biotitic gneisses, graphitic metaquartzites). This zone occurs in the quartz-biotitic paragneisses. Ni-Bi-As mineralization is developed in the environment of metamorphic syngenetic pyrite–pyrrhotite (Py–

Po) mineralization in the black shales. Mineralogy of the area was studied by S. Mikoláš et al. [12], T. Mikuš et al. [13] and J. Pršek et al. [17]. These authors described following minerals: pararammelsbergite, rammelsbergite, löllingite, pyrhotite, pyrite, tennantite, Bi sulphosatls, calcite, hematite, magnetite galenite, Cu and Ni secondary minerals. Mineralization was developed in several stages: Fe-Ni arzenides with quartz and dolomite; Bi sulphosalts with quartz and sulphides; Pb-Zn stage with carbonates; Fe oxides with calcite. Sulphosalts of the cuprobismutite homologues occurs in the third stage.



A



B

Fig. 3. Schematic geological map of Slovakia and a part of crystalline basement of the Suchý.

A: *1* – Carpathian Foredeep; *2* – flysch zone; *3* – klippen belt; *4* – Tatric unit (core mountains); *5* – Upper Palaeozoic to Palaeogene sequences; *6* – Veronic unit; *7* – Gemeric unit; *8* – Tertiary to Quaternary volcanites (mostly Neogene); *9* – Neogene to Quaternary basins;

B: *1* – Mesosoic sedimentary cover; *2* – Krížna nappe; *3* – biotitic mica shists; *4* – graphitic black shales; *5* – ribbed migmatites; *6* – amphibolites; *7* – leucocratic granites; *8* – granodiorites.

Sulphosalts were analysed using wavelenght dispersion analysis (WDS) by JEOL Superprobe 733 microprobe (Kobenhavn Universitet), Camebax (CNIGRI, Moscow) and Cameca SX 100 (ŠGUDŠ, Bratislava) under following conditions: 20 kV and 10–20 nA, beam diameter 5 μm. The following natural (*n*) and synthetic (*s*) standards were used: pure Ag (AgLα), pure Bi (BiLα), pure Sb (SbLα), $_{s}PbS$ (PbLα), $_{s}Bi_2S_3$ (BiLα), $_{n}CuFeS_2$ (CuKα, FeKα, SKα), $_{s}CdSe$ (SeKα) and $_{n}CuSbS_2$ (CuKα, SKα).

Cuprobismutite homologues. ***Banská Hodruša.*** *Kupčíkite* forms several irregular grains associated with matildite and wittichenite in the chalcopyrite and hematite aggregates of quartz found at 14th deep horizon of Rozália Vein. The other occurrence is at 6th deep horizon where kupčíkite is associated with members of bismuthinite-aikinite series and other Bi-sulphosalts. Thin lamellas of kupčikíte are overgrowth with emplectite, hammarite and forms aggregates with krupkaite, hodrushite and paděraite (fig. 4). Analyses of kupčíkite are presented in the table 1.

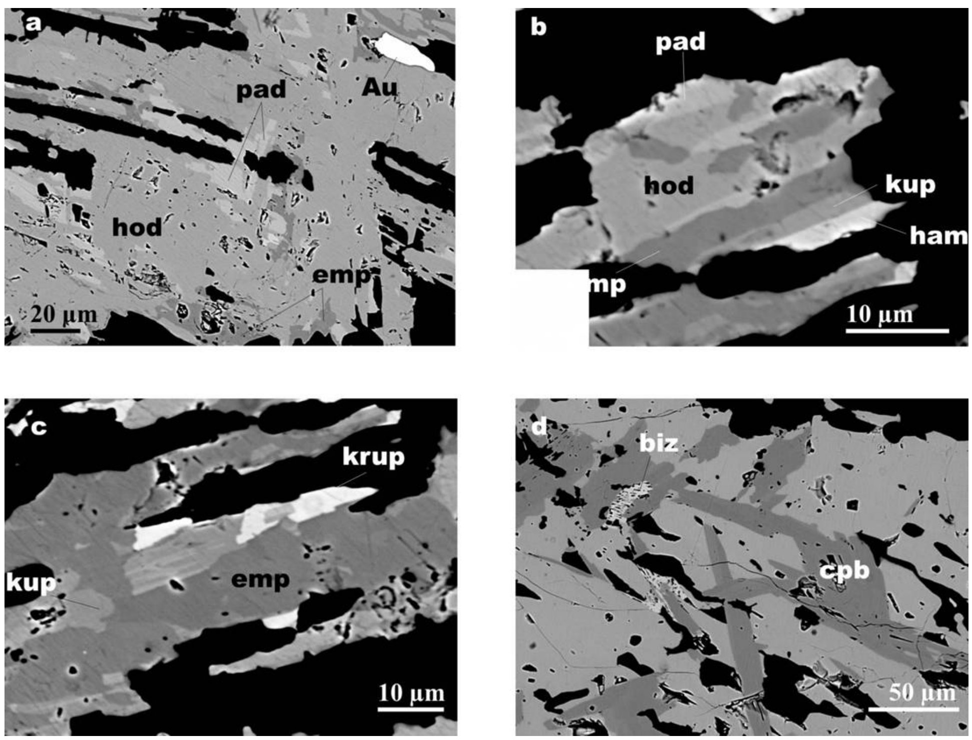


Fig. 4**.** Aggregate of sulfosalts Bi – kupčíkite (kup), hodrushite (hod), emplectite (emp), paděraite (pad), hammarite (ham), krupkaite (krup), needle-like cuprobismutite (cpb), bizmuthinite (biz) and gold (Au) in quartz, hematite and chalcopyrite (black), BSE. Banská Hodruša.

Content of Fe is uniform (1,67–3,83 wt. %) and contents of Pb (up to 0,94) and Ag (up to 1,08 wt. %) are low. Chemically determined *N* varies between values 0,86 to 1,24. General formula of the kupčíkite could be expressed as

$$(Cu_{6,82}Fe_{0,36}Cd_{0,02})_{7,96}(Bi_{9,78}As_{0,31}Sb_{0,03}Ag_{0,02})_{10,14}(S_{19,76}Se_{0,15})_{19,91.}$$

Hodrushite is typomorphous mineral of bismuth from the Rozália Vein. It is dominant Bi-sulphosalt of Cu zone. It occurs associated with wittichenite, aikinite, emplectite, kupčíkite, paděraite, matildite and with other members of bismuthinite-aikinite series and also with gold (fineness 900–910) in the quartz-hematite-chalcopyrite aggregates. It generally forms needles grains up to 1 mm up to size, irregular grains or fine-grained aggregates with other Bi-sulphosalts. Chemical composition of hodrushite is presented in the table 1 and fig. 5.

Table 1

Electron microprobe analysis of minerals cuprobismutite homologous series from Banská Hodruša

Components	Minerals and samples							
	Cpb*	Cpb	Kup		Hod			
	Ro-10A	5-50	a-15	a-23	1-4	1-9	1-18	LIK15
S	16,38	18,33	19,13	19,50	18,63	18,82	19,09	19,20
Fe	0,45	0,06	3,83	1,92	0,60	0,71	1,35	0,84
Bi	65,37	63,01	61,37	62,92	63,55	64,08	63,03	64,30
Pb	–	0,69	0,28	0,00	0,14	0,25	0,16	0,16
Ag	3,55	3,57	1,08	0,07	1,06	0,97	1,17	1,33
Sb	–	0,00	0,00	0,12	0,00	0,00	0,06	0,00
As	–	1,28	0,89	0,71	0,22	0,56	1,14	1,10
Cu	10,44	11,26	12,75	13,34	13,34	13,21	13,15	12,99
Se	0,27	0,45	0,34	0,36	0,40	0,32	0,32	0,39
Cd	–	0,17	0,06	0,06	0,11	0,08	0,12	0,22
Total	96,46	98,82	99,73	99,01	98,05	99,02	99,58	100,53
N	2,51	2,50	0,86	1,06	1,39	1,45	1,36	1,59

* Cpb – cuprobismutite; Kup – kupčíkite; Hod – hodrushite.

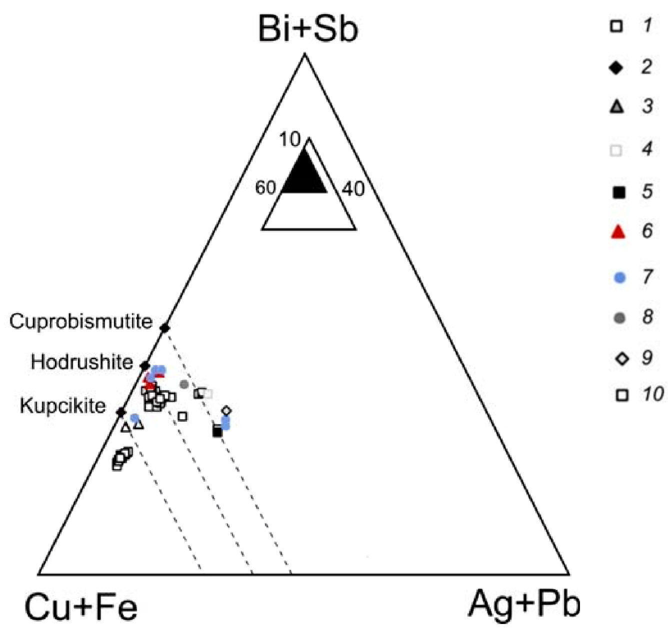


Fig. 5. Chemical composition of cuprobismutite homologous, at. %:
1 – our information; *2* – theoretical data; *3* – from [2]; *4* – from [18]; *5* – from [3]; *6* – from

[8]; *7* – from [7]; *8* – from Žák et al., 1994; *9* – from [16]; *10* – from [17].

Hodrushite composition shows uniform low content of Ag (0,97–1,45 wt. %), Fe (0,60–1,35), Pb (0,14–0,25), As (0,22–1,14) and Se (0,15–0,61 wt. %). Chemically determined *N* varies between values 1,36 to 1,68. General formula of the hodrushite could be expressed as

$$(Cu_{7,68}Fe_{0,47}Cd_{0,03})_{8,18}(Bi_{11,33}As_{0,28}Pb_{0,04}Ag_{0,33})_{11,98}(S_{12,69}Se_{0,15})_{21,84.}$$

Relatively rare c*uprobismutite* was found in aggregates Bi-sulphosalts in quartz-hematite-chalcopyrite accumulations at 6th horizon in the Rozália vein. It forms needles grains and irregular aggregates with bismuthinite, hodrushite, minerals of bismuthinite-aikinite series (fig. 6, 7) and Ag-Cu-Pb-Bi phase (kitaibelite?, fig. 8). Chemical composition of cuprobismutite is presented in the table 1. Cuprobismutite composition shows uniform content of Ag (2,00–3,57 wt. %), Fe (0,06–0,51), Pb (up to 1,03 wt. %). Chemically determined *N* varies between values 1,73 to 2,51. General formula of the cuprobismutite could be expressed as

$$(Cu_{7,08}Fe_{0,24}Cd_{0,10})_{7,42}(Bi_{12,47}As_{0,55}Pb_{0,21}Ag_{1,36})_{14,59}(S_{23,70}Se_{0,29})_{23,99.}$$

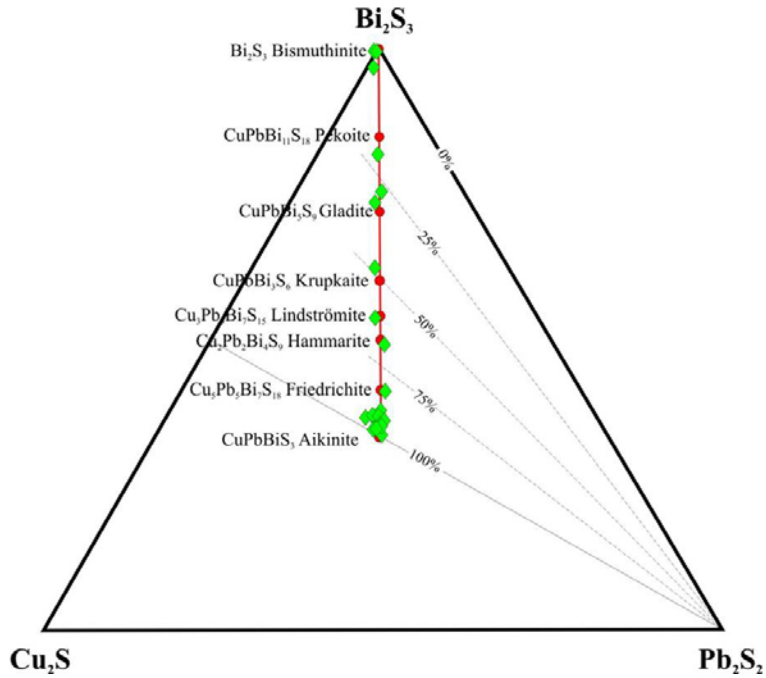


Fig. 6. Chemical composition of sulfosalts bismuthinite–aikinite series, at. %.

Čierna Lehota. *Kupčíkite* ($N = 1$) occurs only in one sample as a small μm-scale grain intergrowth with hodrushite (fig. 9,*a*). Kupčíkite was formed by reaction of hodrushite with the solutions depositing Fe rich tennantite. Microprobe analyses are presented in the table 2. Kupčíkite contains essential but important amounts of Fe and Pb, as well as minor Ag. Content of Fe is very uniform and values ranges around 1,25 at. %. Content of Ag reaches 0,13 at. % (typical for kupčíkite) and content of Pb is relatively stable; its average value is around 1,1 at. %. Chemically determined N varies between values 0,73 and 0,88. General formula of the kupčíkite from Čierna Lehota could be expressed as

$$(Cu_{7.88}Fe_{0.47})_{8.35}(Ag_{0.03}Pb_{0.41}Sb_{0.02}Bi_{9.18})_{9.64}S_{19.12}.$$

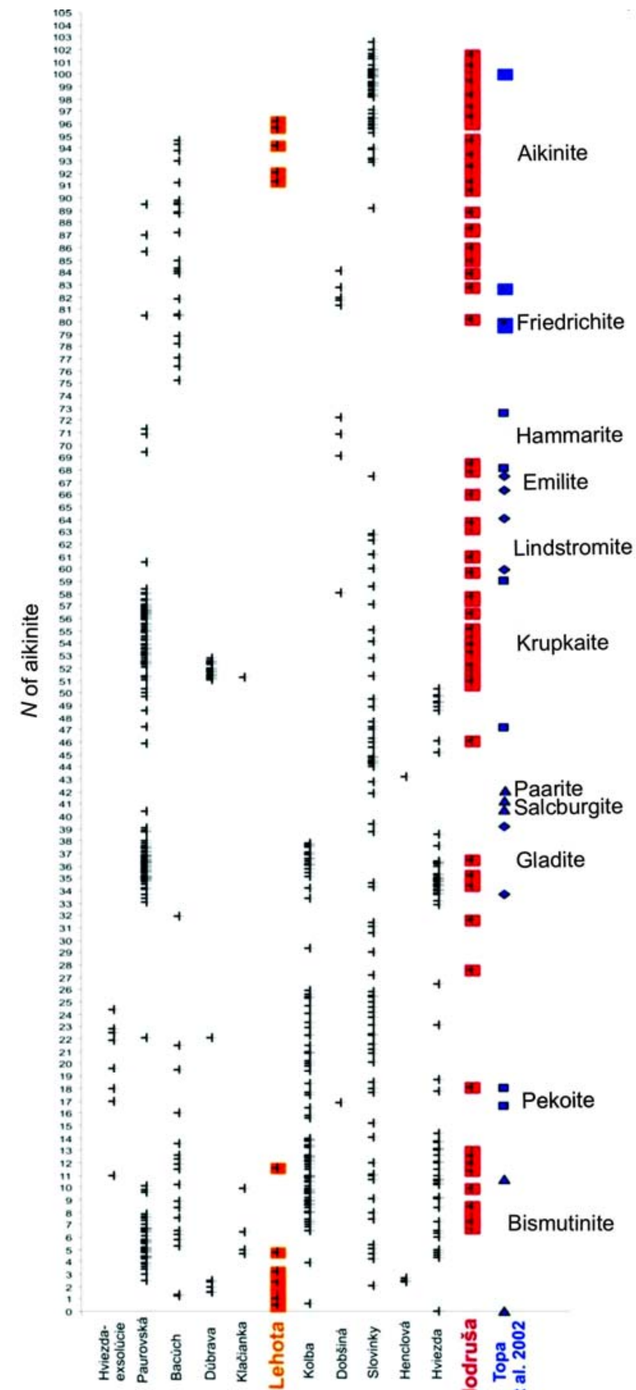


Fig. 7. Sulfosalts of the bismuthinite–aikinite series from different Slovak localities.

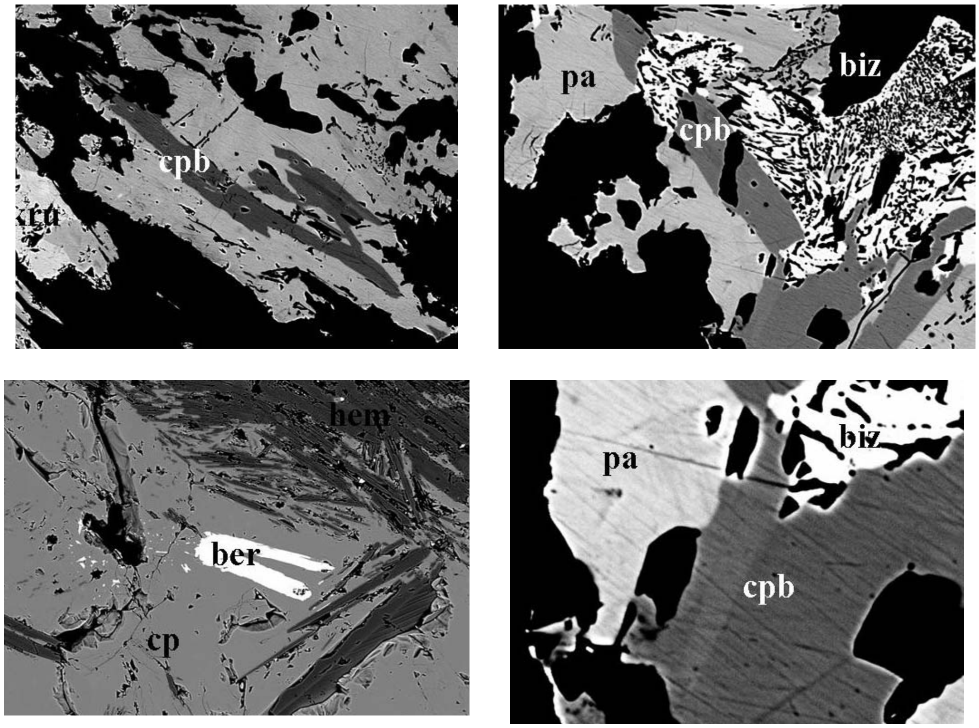
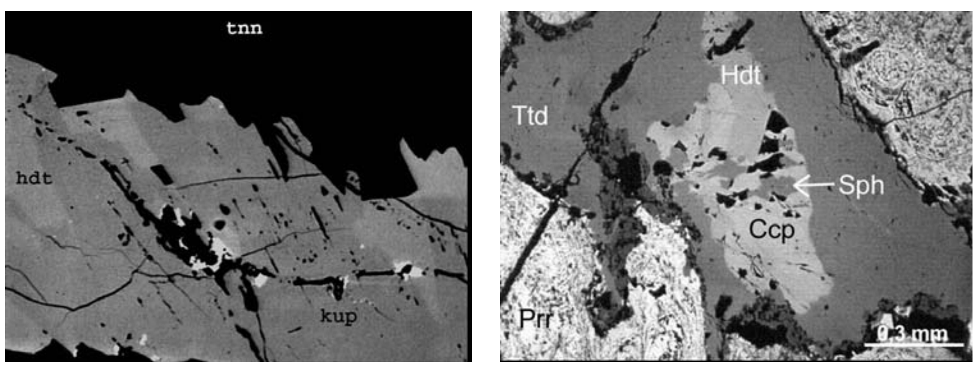


Fig. 8. Aggregates of the intergrowth Bi sulfosalts cuprobizmutite (cpb), krupkaite (kru), berryite (ber), pavonite (pa) and bismuthinite (biz), BSE. Banská Hodruša, Rozália mine.



a *b*

Fig. 9. Intergrowth kupčíkite (kup) and hodrushite (hdt) with tennantite, Čierna Lehota (*a*), and prismatic grain of hodrushite (hdt) intergrowth with sphalerite (sph) and chalcopyrite (ccp) included in tennantite (tn) and pararammelsbergite (prr), BSE, Čierna Lehota (*b*).

Hodrushite (N = 1; 2) is the most commonly encountered bismuth sulphosalt in the locality. Domains (20 μm to 1,5 mm in size) usually forms grains or platy laths in association with chalcopyrite, Bi-tennantite and other Bi sulphosalts disseminated in quartz, carbonates and Ni diarsenides (see fig. 9,*b*). It is partly decomposed into a mixture of native bismuth, bismuthinite and bismuth oxides. Microprobe analyses are presented in the table 2. Our analyses contain small content of Ag, Pb, and Fe. Values for Pb do not exceed 0,3 at. %. Content of iron is relatively uniform around the value of 0,9 at. % and content of silver is up to 1,7 at. %. Chemically calculated *N* has values from 1,25 to 1,65. The results of 75 spot analyses show formula

$$(Cu_{7.79}Fe_{0.38})_{8.17}(Bi_{11.34}Ag_{0.43}Pb_{0.04}Sb_{0.02})_{11.83}S_{22.13.}$$

Table 2

Electron microprobe analysis of minerals cuprobismutite homologous series from Čierna Lehota

Com-po-nents	Minerals								
	Cuprobismutite			Kupčíkite		Hodrushite			
	1	2	3	4	5	6	7	8	9
S	18,75	19,22	19,50	19,02	19,09	19,51	19,43	18,87	19,40
Fe	0,18	0,63	0,58	0,86	0,85	0,56	0,69	0,56	0,53
Bi	59,61	64,21	64,63	61,85	60,60	65,46	65,65	64,64	65,14
Pb	6,73	0,13	0,10	2,98	2,67	0,41	0,47	0,02	0,32
Ag	2,28	3,78	3,87	1,25	0,09	1,47	1,28	1,30	1,83
Sb	0,05	0,01	0,06	0,01	0,01	0,01	0,01	0,07	0,01
Cu	11,99	11,68	11,67	15,67	15,75	13,52	13,38	13,39	13,38
Total	99,60	99,56	100,41	100,63	99,08	100,94	101,07	98,84	100,60
N	2,03	2,02	2,07	0,88	0,81	1,45	1,46	1,42	1,50

Cuprobismutite ($N = 2$) occurs as irregular platy mm-scale grains associated with bismuthinite and matildite. It was also found as a small μm-scale grain enclosed in hodrushite. Microprobe analyses are presented in the table 2.

Cuprobismutite $(Cu_{7.42}Fe_{0.47})_{7.87}(Bi_{12.56}Ag_{1.55}Pb_{0.02})_{14.13}S_{24.68}$ associated with matildite and bismuthinite has higher content of silver up to 3,7 at.%. Content of Fe is 1 at. % – means from 6 analyses. Lead is present only in minor amounts. Chemical calculated *N* is from 2,02 to 2,19. Cuprobismutite $(Cu_{7.67}Fe_{0.14})_{7.81}(Bi_{11.95}Ag_{0.85}Sb_{0.02}Pb_{1.37})_{14.19}S_{24.13}$ occurring with hodrushite is enriched in Pb up to 3 at. % and up to 1,9 at. % of Ag in the structure.

Consequently, sulphosalts of cuprobismutite homologous series are relatively rare in the nature (see fig. 5). Cuprobismutite is known only from several localities and kupčíkite is known up to now from Felbertal deposit and Čierna Lehota occurrences. Recalculation of the published analyses believed to be hodrushite using the above formula shows that $N = 1$ phase might have been encountered in the nature together with other members of cuprobismutite series. It might have occurred as some analyses published by [2], which contain Ag (up to 0,90 at. %), Fe (up to 0,87) and Pb (up to 0,21 at. %) and order *N* is 0,97 and 1,12; by [19] with $N = 1,17$ and contents of Ag – 0.96 at. % and Pb – 0,35 at. % but without Fe and by [8] with $N = 1,11$ and contents of Fe– 1,79 at. %, Ag – 0,79 at. %, and without Pb. The elevated contents of Ag make these determinations as kupčíkite uncertain because D. Topa et al. [20] found that the crystal chemistry of kupčíkite requires higher cation charge than the Cu^{+}–Bi combination yields. This means that for kupčíkite very low Ag contents substituting for Bi are mandatory unless (hypothetically) balanced by unusually high contents of Fe. Our analyses from Hodruša and Čierna Lehota support this hypothesis. If the Ag content is low also Fe content is low and vice versa. Analyses from other published localities show similar chemical composition [11], published analyses of hodrushite from type locality with $N = 1,25$ and 0,70 at. % of Fe, and with a problematic minor Pb content (not included in the calculation of *N*). V. Kovalenker [7] published analyses of hodrushite from Russia with contents of Ag up to 0,82 at. %, Fe up to 1,24 at. %, Pb up to 0,64 at. % and *N* from 1,45 to 1,63, which is similar to our results. Analyses from type locality Banská Hodruša [8] show the content of Fe up to 1,74 at. %, Ag up to 0,82 at. %, Pb up to 0,17 at. % and the calculated *N* is from 1,51 to 1,69. Chemically calculated *N* of our analyses has values from 1,25 to 1,73 and contain small content of Ag, Pb, and Fe.

Published analyses of cuprobismutite generally show higher content of Ag in agreement with the required charge balance [20]. M. Tarkian and V. Breskovska [18] published cuprobismutite with content of Ag up to 3,5 at. %, Pb up to 0,26 at. % and calculated *N* from 1,97 to 2,16. Dobbe's cuprobismutite has content of Ag 4,79 at. %, Fe 1,28 at. % and Pb 0,3 at. % with *N* 2,02 [3]. V. Kovalenker [7] has 0,45 at. % of Fe, 1,86 at. % of Ag and 0,64 at. % of Pb and calculated *N* = 2,07. Also Pattrick's cuprobismutite has high content of Ag (3,26 at. %), Fe (2,42) and Pb (1,41 at. %), and calculated *N* is 2,31.

Noteworthy conclusion is that high content of Fe without Ag is typical for kupčíkite, high content of Ag is typical for cuprobismutite but hodrushite needs both Fe and Ag.

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СУЛЬФОСОЛІ ГОМОЛОГІЧНОГО РЯДУ КУПРОБІСМУТИТУ З РАЙОНІВ БАНСЬКА ГОДРУША ТА ЧЕРНА ЛЕГОТА (ЗАХІДНІ КАРПАТИ, СЛОВАЧЧИНА)

С. Єлень[1], Т. Мікуш[1], Й. Пшек[2]

[1]*Геологічний інститут Словацької Академії наук*
97401 Словаччина, м. Банська Бистриця, вул. Думб'єрська, 1
[2]*Університет Коменіус, факультет природничих наук,*
кафедра мінералогії та петрології
84215 Словаччина, м. Братислава, Млинська Долина

Порівняно рідкісний мінерал купчикіт виявлено разом з іншими членами гомологічного ряду купробісмутиту – годрушитом і купробісмутитом. Вони містяться в складі Ni-Bi-As гідротермальної мінералізації в районі Черна Легота, а також на родовищі кольорових і дорогоцінних металів Банська Годруша (жила Розалія). Купчикіт асоціює з купробісмутитом, годрушитом та членами ряду бісмутиніт–айкініт, емплектитом, падераїтом і високопробним золотом. Досліджено хімічний склад всіх бісмутових сульфосолей, які асоціюють.

Ключові слова: сульфосолі бісмуту, купчикіт, хімічний склад, гідротермальна мінералізація, золото, Західні Карпати, Словаччина.

СУЛЬФОСОЛИ ГОМОЛОГИЧЕСКОГО РЯДА КУПРОВИСМУТИТА ИЗ РАЙОНОВ БАНСКА ГОДРУША И ЧЕРНА ЛЕГОТА (ЗАПАДНЫЕ КАРПАТЫ, СЛОВАКИЯ)

С. Елень[1], Т. Микуш[1], И. Пшек[2]

[1]*Геологический институт Словацкой Академии наук*
97401 Словакия, г. Банска Быстрица, ул. Думбьерская, 1
[2]*Университет Комениус, факультет естественных наук,*
кафедра минералогии и петрологии
84215 Словакия, г. Братислава, Млынска Долина

Сравнительно редкий минерал купчикит обнаружен вместе с другими членами гомологического ряда купровисмутита – годрушитом и купровисмутитом. Их выявлено в составе Ni-Bi-As гидротермальной минерализации в районе Черна Легота, а также на месторождении цветных и благородных металлов Банска Годруша (жила Розалия). Купчикит ассоциирует с купровисмутитом, годрушитом и членами ряда висмутинит–айкинит, эмплектитом, падераитом и высокопробным золотом. Исследован химический состав всех ассоциирующих висмутовых сульфосолей.

Ключевые слова: сульфосоли висмута, купчикит, химический состав, гидротермальная минерализация, золото, Западные Карпаты, Словакия.

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