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PŮVODNÍ PRÁCE/ORIGINAL PAPER

New data on sulphosalts from the hydrothermal siderite-type veins in the Spišsko-gemerské rudohorie Mts. (eastern Slovakia): 4. Tennantite-(Hg) from the Vyšný Klátov ore occurrence

Martin Števko[1, 2)]*, Jiří Sejkora[2)], Tomáš Mikuš[3)] and Dušan Peterec[4)]

[1)]Earth Science Institute v.v.i., Slovak Academy of Sciences, Dúbravská cesta 9, 840 05 Bratislava, Slovak Republic;
**e-mail: martin.stevko@savba.sk*
[2)]Department of Mineralogy and Petrology, National Museum, Cirkusová 1740, 193 00 Praha 9 - Horní Počernice, Czech Republic
[3)]Earth Science Institute v.v.i., Slovak Academy of Sciences, Ďumbierska 1, 974 01 Banská Bystrica, Slovak Republic
[4)]Rovníková 8, 040 12 Košice, Slovak Republic

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Abstract

A new occurrence of tennantite-(Hg) was recently confirmed at the Vyšný Klátov ore occurrence, Spišsko-gemerské rudohorie Mts., Košice-okolie Co., Košice Region, Slovakia. Tennantite-(Hg) occurs as lead-gray to black grains and aggregates up to 1 cm in size, associated with cinnabar, chalcopyrite, pyrite and hematite. Reflectance data of tennantite-(Hg) are given in this paper. The refined unit-cell parameters of tennantite-(Hg) from the Vyšný Klátov (for the cubic space group *I*-43*m*) are: *a* 22.523(7) Å and *V* 3105.4(1) Å^3. Empirical chemical formulae of the two studied samples of tennantite-(Hg) from the Vyšný Klátov ore occurrence, recalculated on the basis of ΣMe = 16 *apfu* are: $(Cu_{5.97}Ag_{0.03})_{\Sigma 6.00}[Cu_{3.99}(Hg_{1.95}Fe_{0.10})_{\Sigma 2.05}](As_{3.57}Sb_{0.39})_{\Sigma 3.96}S_{13.21}$ (sample VK1, n = 21) and $(Cu_{5.99}Ag_{0.01})_{\Sigma 6.00}[Cu_{4.05}(Hg_{1.91}Fe_{0.08})_{\Sigma 1.99}](As_{3.79}Sb_{0.15})_{\Sigma 3.94}S_{13.26}$ (sample VK3, n = 29).

Key words: *tennantite-(Hg), cinnabar, tennantite series, tetrahedrite group, sulphosalts, chemical composition, siderite veins, Vyšný Klátov, Spišsko-gemerské rudohorie Mts., Slovak Republic*

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Introduction

The Spišsko-gemerské rudohorie Mts. represent one of the most important accumulations of ore deposits in whole Carpathian mountain range. There are more than 1200 hydrothermal ore veins known within this relatively small area, with two major types of mineralization: siderite-type carbonate-quartz veins with sulphides (extensively exploited in Dobšiná, Štítnik, Rákoš, Rožňava, Drnava, Rudňany, Novoveská Huta, Hnilčík, Henclová, Prakovce, Gelnica, Slovinky, Medzev etc.) and quartz-stibnite veins (Betliar, Čučma, Štofova dolina, Helcmanovce, Poproč or Zlatá Idka). Furthermore, Sn-Mo-W bearing greisens or granite-related hydrothermal quartz veins, hydrothermal veins with U-REE mineralization as well as strata-bound VMS pyrite-Cu-Pb-Zn ore mineralization and hydrothermal-metasomatic bodies of siderite and magnesite±talc are present (Varček 1962; Chovan et al. 1994; Grecula et al. 1995; Rojkovič 1997). All of the above mentioned types of ore mineralization contain various sulphosalts mostly as accessory ore minerals.

Abundant presence of minerals of tetrahedrite-tennantite series (especially Fe, Zn and locally also Hg dominant members) is very typical feature of the siderite-type veins (e.g. Bernard 1958, 1961; Varček 1957, 1959, 1960; Novák 1959, 1967; Trdlička 1967; Háber 1980; Cambel et al. 1985; Peterec 1990; Miškovic 1991; Háber et al. 1993; Grecula et al. 1995; Antal 2002a, b; Pršek 2008; Pršek, Biroň 2007; Pršek, Lauko 2009; Števko et al. 2015; Mikuš et al. 2018; Peterec 2019; Števko, Sejkora 2020; Kyrc, Losos 2021). Various Bi sulphosalts are also quite common, especially minerals of the bismuthinite-aikinite series (e.g. Paděra et al. 1955; Kupčík et al. 1969; Hurný, Krištín 1978; Mumme, Žák 1983; Antal 1991; Beňka, Siman 1994; Pršek 2008; Števko et al. 2015; Mikuš et al. 2018, 2019; Števko et al. 2021a) and kobellite homologous series (e.g. Trdlička, Kupka 1957; Hak, Kupka 1958; Novák 1961; Trdlička et al. 1962; Václav 1964; Zábranský, Radzo 1966; Háber, Streško 1969; Háber 1980; Jeleň 1991; Pršek 2008; Pršek, Peterec 2008; Mikuš et al. 2018, 2019; Kúšik et al. 2021; Števko et al. 2021b, 2022). Other Bi sulphosalts like cosalite (Bernard 1964; Háber 1980), galenobismutite (Antal 1991; Pršek 2008), jaskólskiite (Pršek, Biroň 2007; Števko et al. 2021b), nuffieldite (Pršek et al. 2006; Števko et al. 2021a) or wittichenite (Háber 1978; Kozub et al. 2011) are rare. Chalcostibite is infrequent too (Sejkora et al. 2011; Mikuš et al. 2018). Unusual assemblage of Ag-Bi sulphosalts (matildite, gustavite and benjaminite) was recently described from the Kobaltová vein near Medzev by Mikuš et al. (2019). The most common Pb sulpho-

Fig. 1 *The main exploration pit at the Vyšný Klátov ore occurrence. Photo by M. Števko, November 2020.*

Fig. 2 *Steel-grey aggregate of tennantite-(Hg) associated with chalcopyrite. Field of view is 5.6 mm. Photo by L. Hrdlovič.*

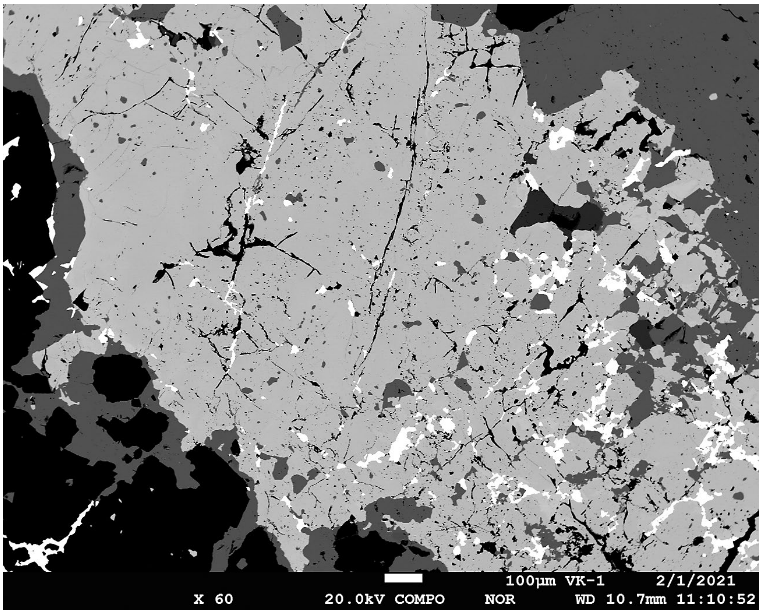

Fig. 3 *Aggregate of tennantite-(Hg) (light grey) associated with cinnabar (white) and chalcopyrite (dark grey) in dolomite (black). BSE image by T. Mikuš.*

salts at the siderite-type veins are bournonite, jamesonite (often Bi-rich) and boulangerite (e.g. Zimányi 1914; Novák 1962; Trdlička 1967; Kupčík et al. 1969; Varček 1971; Háber 1980; Miškovic 1990; Pršek, Biroň 2007; Pršek, Peterec 2008; Sejkora et al. 2011; Mikuš et al. 2018, 2019; Števko et al. 2021b, 2022), whereas berthierite and garavellite (Mikuš et al. 2018), meneghinite (Beňka, Siman 1994) or zinkenite and scainiite (Sejkora et al. 2011) are scarce. Rare Hg sulphosalts, marrucciite (Sejkora et al. 2011) and grumiplucite (Števko et al. 2015) were also recently identified. As-rich sulphosalts, seligmannite and jordanite, were described from the Zenderling deposit near Gelnica by Sejkora et al. (2011).

The occurrence of unusual Hg-rich tennantite at the Vyšný Klátov ore occurrence was first reported by Peterec (1990). Despite older notes of Hg-rich tennantite from the Arizona, USA (Faick 1958), Russia (Mozgova et al. 1978; Mozgova, Tsepin 1983) or Ireland (Steed 1983), tennantite-(Hg) was defined as a new mineral only recently from the famous Lengenbach quarry in Switzerland (Biagioni et al. 2021). Later it was reported also from the Nibao Carlin-type gold deposit in China (Wei et al. 2021) and by Ansermet et al. (2021) from the Chandolin Alp in Switzerland. This paper is presenting detailed mineralogical study of tennantite-(Hg) from the Vyšný Klátov ore occurrence, including reflectance and powder X-ray data as well as large dataset of quantitative chemical analyses obtained from the two samples.

Geological setting

The Vyšný Klátov ore occurrence is located 1.2 km NNW of the Vyšný Klátov village, Spišsko-gemerské rudohorie Mts., Košice-okolie Co., Košice Region, Slovakia. Ore samples with tennantite-(Hg) were collected at the dump of the main exploration pit (Fig. 1). GPS coordinates (WGS84) of this pit are: 48.756187° N and 21.117921° E, 572 m a.s.l.

The Vyšný Klátov ore occurrence was discovered during the geological mapping in the late 1980s by Dušan Peterec. It is represented by short system of NW - SE trending hydrothermal quartz-carbonate veins with cinnabar as a main ore mineral. Ore mineralization is developed at the contact of amphibolites and strongly hydrothermally altered serpentinites (fuchsite-rich listvenites) of the Lower Paleozoic Klátov Group and was explored by several shallow pits. The dominant gangue minerals are quartz, dolomite and siderite and ore minerals are represented by abundant cinnabar, chalcopyrite, pyrite, hematite and rare Hg-rich tennantite (Peterec 1990). We also identified millerite as microscopic, subhedral aggregates associated with chalcopyrite, pyrite and cinnabar.

Analytical methods

Reflectance spectra of tennantite-(Hg) were measured in air with a TIDAS MSP400 spectrophotometer attached to a Leica microscope (20× objective) using a WTiC (Zeiss No. 370) standard (Department of Mineralogy and Petrology, National Museum, Prague, Czech Republic).

X-ray powder diffraction data of tennantite-(Hg) were recorded at room temperature using a Bruker D8 Advance diffractometer (Department of Mineralogy and Petrology, National Museum, Prague, Czech Republic) equipped with solid-state LynxEye detector and secondary monochromator producing Cu*K*α radiation housed at the Department of Mineralogy and Petrology, National Museum, Prague, Czech Republic. The instrument was operating at 40 kV and 40 mA. In order to minimize the background, the powder samples were placed on the surface of a flat silicon wafer. The powder patterns were collected in the Bragg-Brentano geometry in the range 3 - 75° 2θ, step 0.01° and counting time of 20 s per step (total duration of experiment was ca. 3 days). The positions and intensities of diffractions were found and refined using the Pearson VII profile-shape function of the ZDS program package (Ondruš 1993) and the unit-cell parameters were refined by the least-squares program of Burnham (1962). The experimental powder pattern was indexed in line with the calculated values of intensities obtained from the crystal structure of tennantite-(Hg) (Biagioni et al. 2021), based on Lazy Pulverix program (Yvon et al. 1977).

Paragenetic and textural relationships as well as chemical zoning of ore minerals were studied in BSE mode using a JEOL JXA-8530F electron microprobe (Earth Science Institute, Slovak Academy of Sciences, Banská Bystrica, Slovak Republic).

The quantitative chemical analyses of tennantite-(Hg) were performed using a Cameca SX100 electron microprobe (Department of Mineralogy and Petrology, National Museum, Prague, Czech Republic) operating in the wave-dispersive (WDS) mode (25 kV, 20 nA and 0.7 μm wide beam). The following standards and X-ray lines were used to minimize line overlaps: Ag (Ag*Lα*), $\mathrm{Bi_2Se_3}$ (Bi*Mβ*), CdTe (Cd*Lα*), Co (Co*Kα*), $\mathrm{CuFeS_2}$ (Cu*Kα*, S*Kα*), $\mathrm{FeS_2}$ (Fe*Kα*), GaAs (Ga*Lα*), Ge (Ge*Lα*), HgTe (Hg*Lα*), InAs (In*Lα*), Mn (Mn*Kα*), NaCl (Cl*Kα*), NiAs (As*Lβ*), Ni (Ni*Kα*), PbS (Pb*Mα*), PbSe (Se*Lβ*), PbTe (Te*Lα*), $\mathrm{Sb_2S_3}$ (Sb*Lα*), Sn (Sn*Lα*), TlBrI (Tl*Lα*), and ZnS (Zn*Kα*). Contents of the above-listed elements, which are not included in the tables, were analysed quantitatively, but their contents were consistently below the detection limit (ca. 0.03 - 0.05 wt. % for individual elements). Raw intensities were converted to the concentrations of elements using automatic "PAP" matrix-correction procedure (Pouchou, Pichoir 1985).

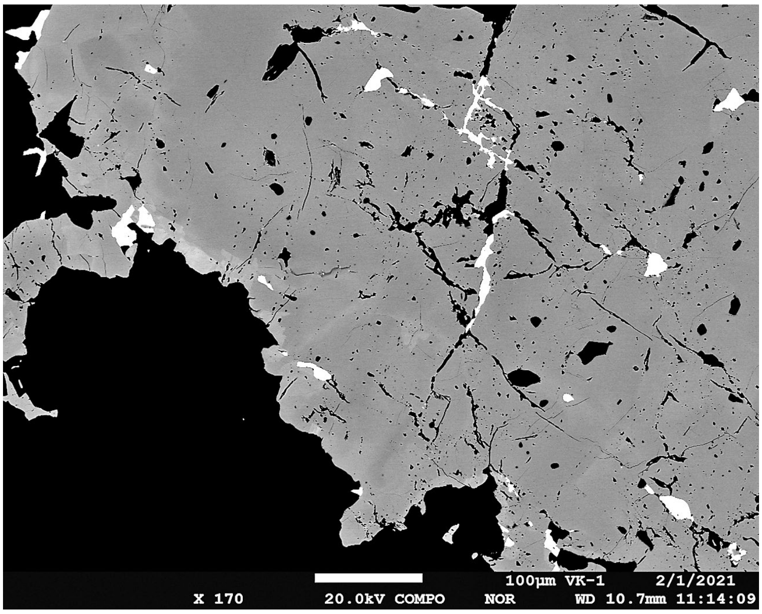

Fig. 4 *Weak chemical zoning of tennantite-(Hg), white inclusions and veinlets are cinnabar. BSE image by T. Mikuš.*

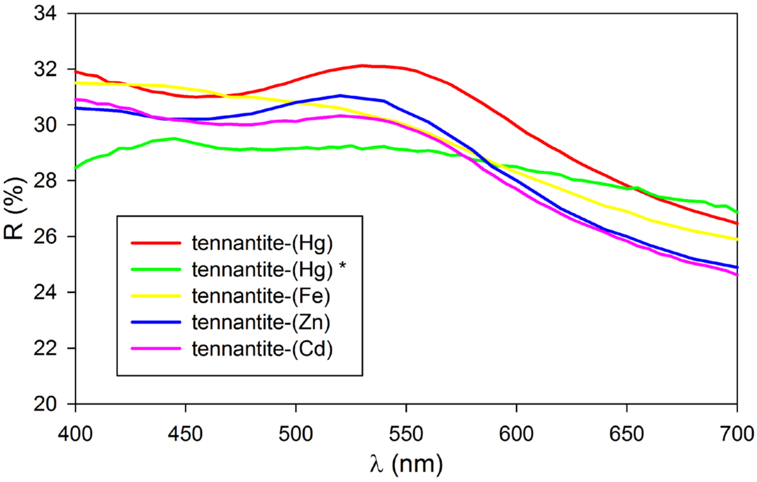

Fig. 5 *Reflectance curve for tennantite-(Hg) from Vyšný Klátov (this paper) and *Lengenbach (Biagioni et al. 2021). For the sake of comparison, the reflectance curves of other members of the tennantite series are shown: tennantite-(Fe), Wheal Jewel, Gwennap, Cornwall, U.K. (Criddle, Stanley 1993); tennantite-(Zn), Tsumeb, Namibia (Criddle, Stanley 1993); tennantite-(Cd), Berenguela mining district, Bolivia (Biagioni et al. 2022).*

Table 1 *Reflectance data for tennantite-(Hg) from the Vyšný Klátov*

λ (nm)	*R* (%)	λ (nm)	*R* (%)
400	31.9	560	31.8
420	31.5	580	31.0
440	31.1	589	30.5
460	31.0	600	30.0
470	31.0	620	29.0
480	31.2	640	28.2
500	31.6	650	27.8
520	32.0	660	27.5
540	32.1	680	26.9
546	32.1	700	26.5

Results

Tennantite-(Hg) is a rare mineral at the studied locality. It forms lead-grey to black, metallic, subhedral to anhedral grains and aggregates up to 1 cm in size (Fig. 2), enclosed in dolomite. Tennantite-(Hg) is often directly associated with cinnabar, chalcopyrite, pyrite and hematite (Fig. 3). Aggregates of tennantite-(Hg) show only a very weak chemical zoning in BSE (Fig. 4), caused by minor variation of As and Sb contents.

Figure 5 shows the reflectance curve of tennantite-(Hg) from Vyšný Klátov (sample VK3, Table 1) and its comparison with the published data for other members

Table 2 *X-ray powder diffraction data of tennantite-(Hg) from the Vyšný Klátov*

$I_{obs.}$	$d_{obs.}$	$d_{calc.}$	*h*	*k*	*l*	$I_{obs.}$	$d_{obs.}$	$d_{calc.}$	*h*	*k*	*l*
0.6	7.321	7.321	1	1	0	32.7	1.8303	1.8303	4	4	0
0.9	4.228	4.227	2	1	1	1.2	1.7758	1.7756	4	3	3
100.0	2.989	2.989	2	2	2	3.7	1.6797	1.6796	6	1	1
8.3	2.768	2.767	3	2	1	3.7	1.6797	1.6796	5	3	2
16.9	2.589	2.588	4	0	0	6.9	1.6377	1.6371	6	2	0
5.6	2.4404	2.4404	3	3	0	14.1	1.5609	1.5609	6	2	2
5.6	2.4404	2.4404	4	1	1	2.7	1.4943	1.4944	4	4	4
5.5	2.0307	2.0305	4	3	1	1.2	1.4642	1.4612	7	1	0
7.4	1.8903	1.8903	5	2	1	4.4	1.4090	1.4090	6	3	3

Table 3 *Representative WDS analyses of tennantite-(Hg) from the Vyšný Klátov (wt.%)*

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
sample	VK1	VK1	VK1	VK1	VK1	VK1	VK1	VK3	VK3	VK3	VK3	VK3	VK3	VK3	VK3
Cu	35.68	35.57	35.94	35.24	35.12	35.19	35.82	37.00	36.34	35.65	36.51	36.39	36.49	36.08	35.56
Ag	0.12	0.17	0.12	0.17	0.19	0.24	0.11	0.08	0.12	0.00	0.00	0.00	0.00	0.00	0.09
Hg	22.42	21.99	21.98	22.40	21.80	22.00	21.70	22.00	21.83	21.80	21.47	21.88	21.87	21.75	21.83
Fe	0.34	0.29	0.27	0.36	0.27	0.25	0.34	0.46	0.29	0.19	0.24	0.30	0.26	0.23	0.19
As	16.16	14.67	16.02	14.00	12.49	12.50	15.94	15.48	16.43	13.05	16.72	16.81	16.60	14.65	13.22
Sb	1.31	3.24	1.52	4.04	6.69	6.52	1.55	0.77	0.68	5.69	0.23	0.26	0.47	2.79	5.73
S	24.06	23.85	24.10	23.74	23.47	23.34	24.02	24.02	23.96	23.75	24.12	24.17	24.31	23.84	23.61
total	100.08	99.79	99.95	99.95	100.03	100.05	99.48	99.81	99.65	100.13	99.29	99.81	100.00	99.34	100.23
$Cu_{A+B\text{-}site}$	9.906	9.966	9.972	9.921	9.939	9.948	9.971	10.195	10.034	10.040	10.092	10.029	10.059	10.108	9.991
Ag	0.020	0.028	0.020	0.028	0.032	0.040	0.018	0.013	0.020	0.000	0.000	0.000	0.000	0.000	0.015
Hg	1.972	1.952	1.932	1.998	1.955	1.970	1.914	1.921	1.910	1.945	1.880	1.910	1.910	1.930	1.943
Fe	0.107	0.093	0.086	0.116	0.088	0.081	0.109	0.144	0.091	0.061	0.075	0.094	0.082	0.073	0.061
Σ C site	2.079	2.046	2.018	2.114	2.042	2.052	2.023	2.065	2.001	2.006	1.955	2.004	1.992	2.004	2.004
Sb	0.190	0.474	0.220	0.594	0.989	0.962	0.225	0.110	0.098	0.836	0.033	0.037	0.068	0.408	0.840
As	3.805	3.485	3.770	3.343	2.998	2.998	3.763	3.617	3.848	3.117	3.920	3.929	3.881	3.481	3.150
Σ D site	3.995	3.960	3.990	3.937	3.987	3.959	3.988	3.727	3.946	3.954	3.953	3.967	3.949	3.889	3.990
S	13.238	13.245	13.249	13.244	13.161	13.074	13.253	13.115	13.111	13.256	13.212	13.201	13.281	13.235	13.146

calculated empirical formulae are based on ΣMe = 16 *apfu*

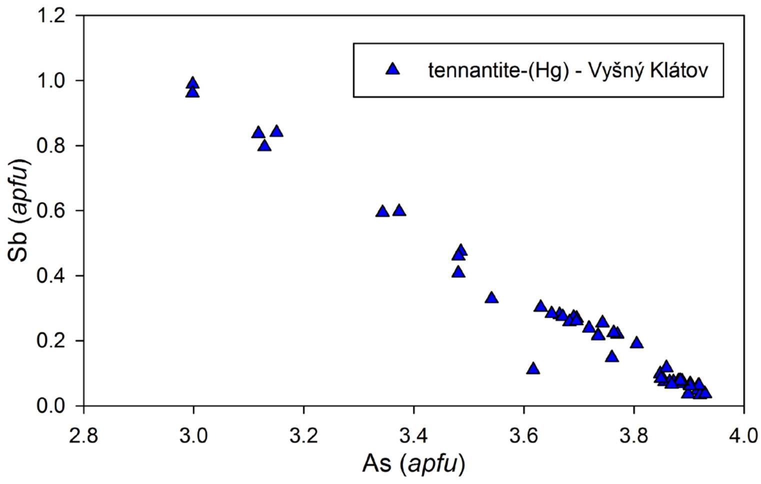

Fig. 6 *Variation of As vs. Sb contents (apfu) in tennantite-(Hg) from the Vyšný Klátov.*

of the tennantite series. The slightly different character of the curve of tennantite-(Hg) from Lengenbach (Biagioni et al. 2021) is probably caused by minimal dimensions of the measured sample and not quite perfect surface of studied polished section.

The obtained experimental X-ray powder diffraction data set of tennantite-(Hg) from the Vyšný Klátov (sample VK3) given in Table 2 agrees well with data for minerals of the tennantite series. Its refined unit-cell parameters a = 10.3536(3) Å, V = 1109.88(9) Å^3 are in very good agreement with the predicted value a = 10.358 Å calculated by equation of Johnson et al. (1987). Johnson et al. (1987) proposed a relation between the unit-cell parameter and chemistry of minerals of the tetrahedrite group: a (Å) = 10.379 + 0.082(Ag) - 0.01(Ag^2) - 0.009(Cu*) + 0.066(Hg) - 0.038(As) + 0.144(Bi), where Cu* = 2.0 - (Fe+Zn+Hg+Cd), the coefficient of the Hg term is corrected according to Di Benedetto et al. (2002). The increased value of unit-cell parameter a = 10.455(7) Å of tennantite-(Hg) from the Lengenbach quarry (Biagioni et al. 2021) probably caused by the elevated amounts of Ag detected in studied sample, its predicted value based on equation of Johnson et al. (1987) is a = 10.404 Å.

Representative quantitative chemical analyses of two samples of tennantite-(Hg) from the Vyšný Klátov and the corresponding empirical formulae are shown in Table 3. All 50 WDS analyses are available as supplementary file. Both studied samples of tennantite-(Hg) from the Vyšný Klátov ore occurrence are containing only minor amounts of Ag (up to 0.04 *apfu*) and Fe (up to 0.14 *apfu*). The overall Hg content in tennantite-(Hg) is ranging from 1.88 to 2.00 *apfu*. The extent of AsSb$_{-1}$ substitution is limited to 0.99 *apfu* of Sb (Fig. 6). Empirical chemical formulae of the studied samples of tennantite-(Hg) from the Vyšný Klátov ore occurrence, recalculated on the basis of ΣMe = 16 *apfu* are: $(Cu_{5.97}Ag_{0.03})_{\Sigma 6.00}[Cu_{3.99}(Hg_{1.95}Fe_{0.10})_{\Sigma 2.05}](As_{3.57}Sb_{0.39})_{\Sigma 3.96}S_{13.21}$ (sample VK1, n = 21) and $(Cu_{5.99}Ag_{0.01})_{\Sigma 6.00}[Cu_{4.05}(Hg_{1.91}Fe_{0.08})_{\Sigma 1.99}](As_{3.79}Sb_{0.15})_{\Sigma 3.94}S_{13.26}$ (sample VK3, n = 29).

Conclusions

A presence of Hg-dominant member of the tennantite series, tennantite-(Hg), was confirmed at the Vyšný Klátov ore occurrence in Slovakia. It is the first occurrence of this species in the Western Carpathians and one of only a few known occurrences of this rare member of the tetrahedrite group in Nature.

Acknowledgements

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