

Klajite from Recsk (Hungary), the first Mn-Cu arsenate mineral

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Abstract: Klajite (IMA no. 2010-004) was found in ore samples from the Lahóca Hill, Recsk, Mátra Mountains (northern Hungary). It belongs to the Lahóca epithermal high-sulphidation Cu-Au-As ore deposit, a classical occurrence of enargite. The primary ore consists of enargite, luzonite, pyrite, tennantite, chalcopyrite, galena, and sphalerite. Klajite occurs in the cavities of enargite and quartz, in close association with other secondary minerals: gypsum, jarosite, and an unknown Ca-Cu arsenate. Klajite is a secondary mineral formed by the decomposition of enargite. It forms irregular or sheaf-like aggregates, up to 0.5 mm in diameter, made up of lath-like to thin tabular crystals, typically 0.05–0.2 mm in length. The mineral is translucent, greenish yellow to yellowish green in colour with white streak and vitreous lustre. Its Mohs hardness is about 2–3, the calculated density is 3.213 g/cm³. Klajite has a perfect cleavage parallel to {010}, and is extremely brittle; the fracture is uneven. Optically it is biaxial negative or positive with $\alpha = 1.595(30)$, $\beta = \text{n.d.}$ and $\gamma = 1.665(20)$. It is weakly pleochroic, from colourless to pale green. The chemical composition obtained after correction of electron-microprobe analysis is: MnO 5.67, CuO 32.03, CaO 0.41, As₂O₅ 44.40, H₂O (calc.) 17.49, total 100.00 wt%. The empirical formula is $(\text{Mn}_{0.82}\text{Cu}_{0.10}\text{Ca}_{0.08})_{\Sigma} = 1.00 \text{ Cu}_{4.05}\text{As}_{3.98}\text{O}_{14}(\text{OH})_2 \cdot 9\text{H}_2\text{O}$, and the simplified formula is $\text{MnCu}_4(\text{AsO}_4)_2(\text{AsO}_3\text{OH})_2 \cdot 9\text{H}_2\text{O}$. The strongest seven lines in the X-ray powder diffraction pattern are [d_{hkl} in Å (I_{obs} %, hkl)] 10.39 (100, 001), 2.916 (64, 202), 2.708 (29, 113), 3.616 (28, 021), 3.050 (28, 022 and 211), 3.956 (27, 020) and 3.110 (24, 122). According to X-ray powder diffraction, klajite is structurally analogous to lindackerite-group minerals; it is triclinic, space group $P\bar{1}$, $a = 6.441(3)$, $b = 7.983(4)$, $c = 10.562(3)$ Å, $\alpha = 85.28(4)^\circ$, $\beta = 80.63(5)^\circ$, $\gamma = 84.80(4)^\circ$, $V = 532.4(3)$ Å³, $Z = 1$. Klajite is named after Sándor Klaj (bom. 1948), a Hungarian mineral collector.

Key-words: klajite, new mineral, lindackerite group, X-ray powder diffraction data, electron-microprobe analysis, copper manganese arsenate hydroxide hydrate, Recsk, Hungary.

1. Introduction

During a field trip to the Mátra Mountains (Hungary) in 1995, Sándor Klaj collected enargite ore samples from the old pit-heap of the Lejtakna area, Lahóca Hill at Recsk. In the vugs of the collected samples he observed yellowish green submillimetric aggregates. All the samples were sent to the Herman Ottó Museum for identification. Analysis by energy dispersive spectrometry (EDS) performed by the senior author revealed the mineral to be a Mn–Cu arsenate. The second author ascertained by X-ray powder diffraction that the mineral was a new member of the lindackerite group. When all investigations enabled by the very limited amount of material available were completed, the mineral was submitted to the Commission on New Minerals, Nomenclature and Classification of the International Mineralogical Association (IMA CNMNC) and the mineral and its name klajite (клайт in Russian) were approved under IMA no. 2010-004. The new mineral was presented at the 20th General Meeting of IMA held in Budapest (Szakáll *et al.*, 2010).

Type materials are deposited in the collections of the Herman Ottó Museum, Miskolc (holotype, catalogue number 2010.1) and of the Hungarian Natural History Museum, Budapest (cotype, specimen number Gyn./1842).

The mineral was named in honour of Sándor Klaj (b. 1948), a prominent figure of the Hungarian mineral collecting community since 1990. He has found more than 50 mineral species new to the territory of Hungary and provided the samples used for the present study.

2. Occurrence and mineralogy of the ore deposit

The Lahóca Hill area, between Recsk and Paráds (Mátra Mountains, northern Hungary) was mined for copper and gold from 1852 until 1980. The Paleogene volcanic unit that hosts the high-sulphidation Cu-Au-As epithermal ore consists of a quartz-bearing andesite crystal tuff and diatreme-breccia complex. The major host-rock of the mineralization is an andesitic crystal tuff. Targets of the old underground

mining were the copper-rich enargite-luzonite ore bodies hosted by the complex. These ore bodies consisted mainly of hydrothermal breccia with massive enargite-luzonite. The Lahóca central brecciated zone contains the major part of the mineralization with its upper boundary contact “blue shale”. This is a clayey rock with a high content of dispersed pyrite and small silicified fragments. Due to the clayey character it formed an impermeable cap above the stratovolcanic sequence. The highest gold grades were historically found in the “blue shale” zone. This was also proved by the latest explorations (Földessy *et al.*, 2008; Molnár *et al.*, 2008). To date eleven large ore bodies (stocks) were known in the Lahóca Hill. The Lahóca ore deposit was the first in Europe where enargite was described (Pettkó, 1863).

Specimens containing the new mineral were collected from the pit-heap of the Lejtakna (“inclined adit”) area, in the northern part of the Lahóca Hill (Fig. 1). The ore of the Lejtakna area was discovered in 1969 when borehole Rm-48 was drilled (Baksa, 1975). This mineralization is part of the Lahóca epithermal high-sulphidation Cu-Au-As ore deposit. Here, late pipe breccias containing reworked clasts of enargite ores with hydrothermal quartz cement were the host environment for the Lejtakna ore body. This ore body has lower gold and higher silver content than the main Lahóca ore bodies (Fig. 2).

The primary ore consists mainly of enargite, luzonite and pyrite, less frequently tennantite, chalcopyrite, galena, and sphalerite (Baksa, 1975; Földessy *et al.*, 2008). The researchers distinguished three mineralization phases: (1) early galena-sphalerite-pyrite; (2) enargite-luzonite; (3) early galena-sphalerite-pyrite; (2) enargite-luzonite; (3)



Fig. 1. Topographic map of the Lejtakna area and its surroundings. TL = type locality.

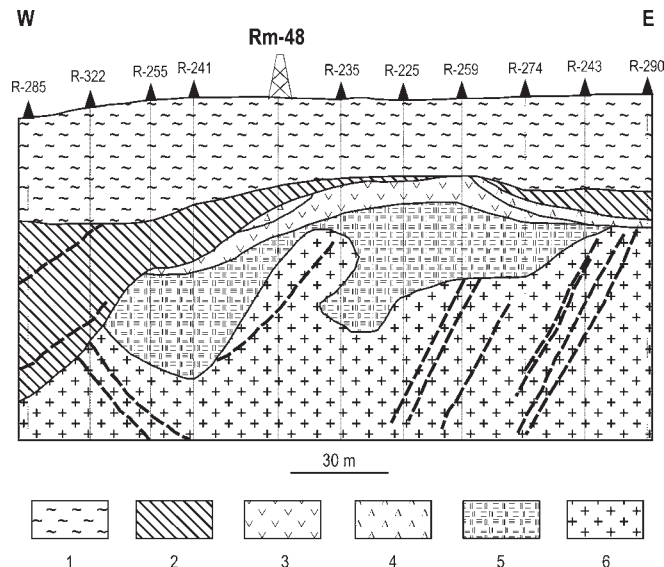


Fig. 2. Geological cross section of the Lejtakna area (after Földessy *et al.*, 2008). Legend: (1) clay, clayey marl; (2) pre-ore subvolcanic andesite; (3) andesite, pyroclastites; (4) mineralized contact breccia; (5) pipe breccia; (6) andesite.

late pyrite-native gold. The main copper-bearing minerals are enargite and luzonite. Enargite forms irregular, subhedral to euhedral elongated crystals (up to 1 cm), however, it was also identified in fine to coarse-grained masses. The corroded contours of enargite and luzonite grains indicate that they were formed in the earlier phase of mineralization.

In the near-surface position, along with cracks of the ores, sulphides are commonly altered to sulphates, phosphates and arsenates (Szakáll *et al.*, 2005). Among the arsenates and phosphates, sampleite, crandallite and scorodite were identified. Klajite occurs in small cavities of enargite ore or appears in little patches in the strongly altered enargite (Fig. 3).

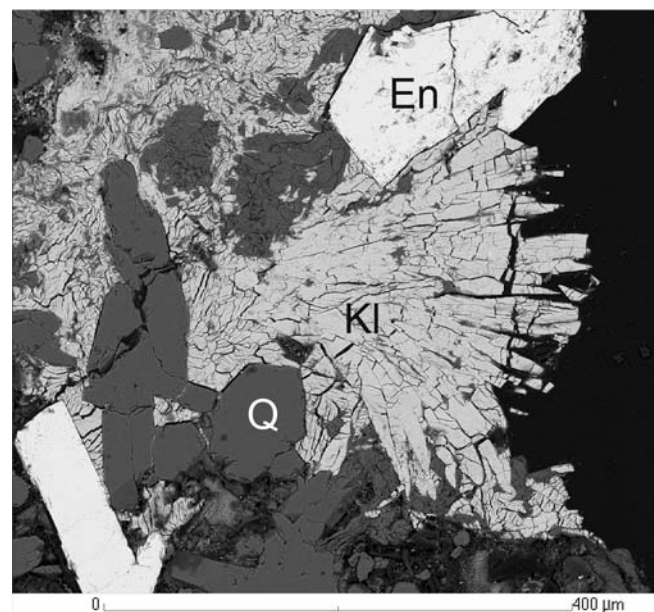


Fig. 3. Back-scattered electron image showing the association of klajite (Kl), enargite (En) and quartz (Q).

A sky-blue, unknown Ca-Cu arsenate was found in close association with klajite. Occasionally some gypsum and jarosite appear as finally precipitated secondary minerals. Klajite formed by the decomposition of enargite.

3. Appearance and physical properties

Klajite forms irregular or sheaf-like aggregates up to 0.5 mm in diameter, which are made up of lath-like to thin tabular crystals, typically 0.05–0.2 mm in length. Twinning was not observed. The mineral is greenish yellow to yellowish green, translucent, with white streak and vitreous lustre. Its Mohs hardness is about 2–3. Density could not be measured directly due to the very limited amount of material available as well as the fact that the mineral decomposes in heavy liquids. The calculated density is 3.213 g/cm^3 for the empirical formula. Klajite has a perfect cleavage parallel to $\{010\}$, and is very brittle. Its fracture is uneven. The mineral is not fluorescent under either long- or short-wave ultraviolet light and it is insoluble in water (Fig. 4 and 5).

Most of the optical properties could not be determined because of the size and extreme brittleness of the crystals. Klajite crystals break into pieces during the preparation or disintegrate in the measuring liquids. Optically klajite is biaxial negative or positive with $\alpha = 1.595(30)$, $\beta = \text{n.d.}$ and $\gamma = 1.665(20)$, $n = 1.601(10)$. It is weakly pleochroic, from colourless to pale green.

4. X-ray crystallography

Single-crystal X-ray analysis was not possible by lack of klajite crystals with appropriate quality, because of multiple intergrowths. A powder X-ray diffraction pattern was obtained using Ni-filtered $\text{CuK}\alpha$ radiation and a 114.6 mm



Fig. 4. Photomicrograph of greenish yellow sheaf-like aggregate of klajite. Length of the aggregate is 0.5 mm. Photo courtesy of László Tóth.

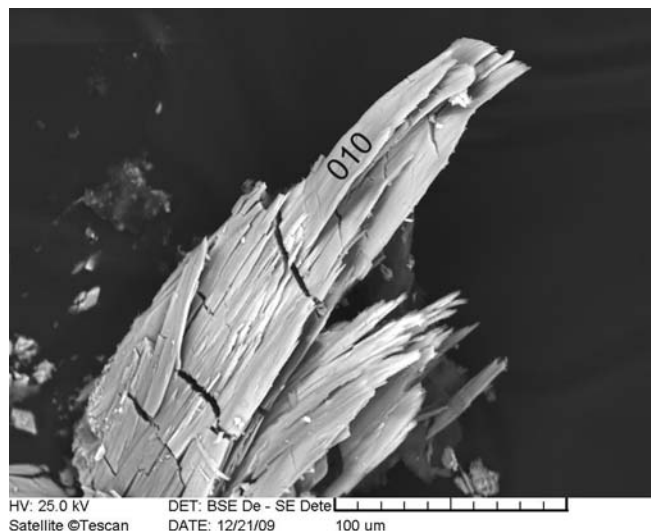


Fig. 5. SEM image of klajite. Thin tabular crystals with relatively well-developed $\{010\}$ form (based on analogy with lindackerite-group phases). The image pictures the extreme brittleness of klajite.

Gandolfi camera. Operating conditions were as follows: accelerating voltage 40 kV, tube current 25 mA, and exposition time 48 hours. The measured klajite aggregate (0.2 mm in diameter) was intergrown with quartz which served as internal standard for film shrinkage. The Gandolfi film was scanned using a UMAX PowerLook 3000 scanner with resolution of 3000 dpi. Values of interplanar spacings (d) were measured manually on contrasted scanned film using Corel PhotoPaint software. First the I_{rel} values were estimated visually then these were measured by two independent methods: (1) using ImageJ plus PowderX softwares and (2) using QSpectr software (see Zelensky *et al.*, 2009). Both methods produced the same values (Table 1).

According to the X-ray powder diffraction pattern, klajite is structurally analogous to lindackerite-group minerals. We used the crystal structure data of lindackerite determined by Hybler *et al.* (2003) for calculating the X-ray powder pattern of klajite, where the M site is occupied by Mn while Cu1 and Cu2 sites are occupied by Cu (site notations according to Hybler *et al.*, 2003). Klajite is triclinic, space group $P\bar{1}$. The cell parameters obtained from the observed powder data: $a = 6.441(3)$, $b = 7.983(4)$, $c = 10.562(3) \text{ \AA}$, $\alpha = 85.28(4)^\circ$, $\beta = 80.63(5)^\circ$, $\gamma = 84.80(4)^\circ$, $V = 532.4(3) \text{ \AA}^3$, $Z = 1$, and $a:b:c = 0.8068:1:1.3231$.

5. Chemical composition

A preliminary, qualitative energy-dispersive analysis of type klajite was performed using a JEOL JXA8600 electron-microprobe. The result showed that the mineral is rich in Mn, Cu and As, and contains some Ca as well. No other elements with atomic number >10 were detected.

Table 1. X-ray powder diffraction data of klajite from Recsk.

$I_{\text{rel.}} (\%)$	$d_{\text{meas.}} (\text{\AA})$	$d_{\text{calc.}} (\text{\AA})$	h	k	l	$I_{\text{rel.}} (\%)$	$d_{\text{meas.}} (\text{\AA})$	$d_{\text{calc.}} (\text{\AA})$	h	k	l
100	10.39	10.396	0	0	1	8	1.975	1.977	-3	-1	1
22	7.95	7.931	0	1	0			1.973	0	4	1
7	6.11	6.107	0	-1	1	8	1.954	1.956	1	-3	3
13	5.16	5.151	1	1	0			1.955	2	-3	0
6	4.77	4.771	1	-1	0	5	1.932	1.936	1	4	0
10	4.47	4.468	1	-1	1			1.933	1	3	4
27	3.956	3.966	0	2	0	6	1.900	1.898	-2	2	3
12	3.786	3.793	0	2	1			1.897	0	4	2
28	3.616	3.623	0	-2	1	10	1.858	1.857	-2	-3	2
17	3.479	3.487	1	2	0	9	1.816	1.817	1	-4	1
16	3.264	3.263	1	0	3	8	1.780	1.782	3	1	4
		3.263	0	2	2	8	1.739	1.738	2	-3	3
18	3.167	3.172	2	0	1			1.735	1	1	6
		3.169	-1	-2	1	6	1.698	1.697	3	-1	4
		3.168	2	0	0	6	1.676	1.680	-2	-4	1
24	3.110	3.115	1	2	2			1.674	3	-2	3
28	3.050	3.053	0	-2	2	8	1.619	1.621	1	-4	3
		3.052	2	1	1	7	1.606	1.609	-1	0	6
64	2.916	2.915	2	0	2			1.604	4	0	1
14	2.860	2.849	2	-1	1	5	1.587	1.586	0	5	0
5	2.781	2.785	1	-2	2			1.586	4	0	2
		2.776	-2	-1	1	9	1.546	1.547	0	5	2
29	2.708	2.707	-1	1	3			1.547	3	-1	5
16	2.604	2.607	2	2	1			1.544	1	-2	6
		2.605	0	3	1	5	1.478	1.481	0	5	3
12	2.543	2.545	2	0	3			1.479	2	5	1
17	2.517	2.521	0	-3	1	8	1.459	1.461	-4	-2	1
		2.515	1	3	1			1.459	2	3	6
14	2.380	2.385	2	-2	0			1.457	-2	4	3
		2.383	1	3	2			1.457	-3	-4	1
13	2.292	2.296	-1	-2	3	4	1.422	1.421	2	5	3
		2.294	0	-3	2			1.420	4	2	4
8	2.257	2.261	2	2	3	6	1.386	1.387	-3	-4	2
9	2.084	2.082	3	1	0	7	1.375	1.376	2	-5	0
		2.079	0	0	5			1.375	-1	-5	3
10	2.057	2.059	3	1	2	5	1.363	1.362	-2	5	1
		2.055	1	1	5	4	1.344	1.342	-1	-4	5
		2.054	1	-2	4			1.342	-4	2	2
5	2.013	2.016	3	-1	1	7	1.321	1.322	3	1	7
		2.010	2	2	4			1.319	4	-3	1
		2.009	-2	-3	1						
		2.009	-3	0	1						

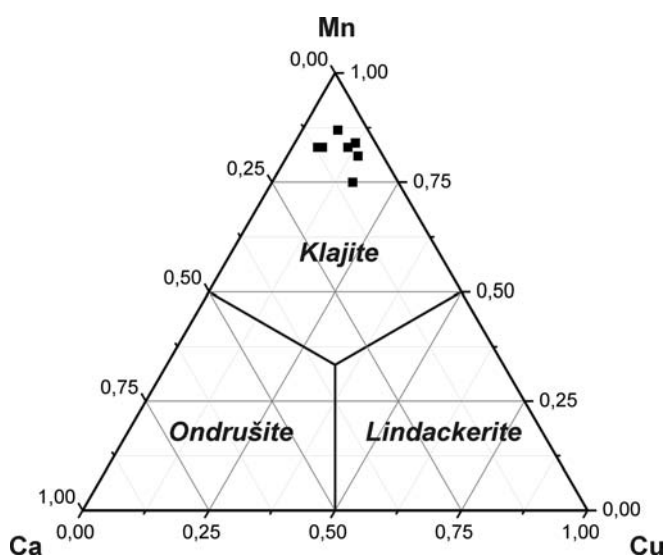
Quantitative chemical analyses were performed using an ARL-SEMQ electron microprobe operated in wavelength-dispersive mode. Operating conditions were as follows: excitation voltage 15 kV, probe current 20 nA, a final beam diameter of 6 μm , peak count-times 10 s and background count-times 5 s. Raw intensity data were corrected using a $\Phi(\rho Z)$ matrix correction.

The results of individual spot analyses and the cation numbers for 15 oxygens are shown in Table 2. Considering klajite, the dominant element in the M site is always Mn (0.75–0.87 *apfu*), with minor amounts of Cu (0.05–0.16 *apfu*) and Ca (0.04–0.11 *apfu*). The compositional variations of klajite can be represented in a ternary diagram, the apices of which are the ideal compositions of klajite ($M = \text{Mn}$), lindackerite ($M = \text{Cu}$) and ondušite ($M = \text{Ca}$) (Fig. 6).

Under the electron beam, klajite is unstable and partly dehydrates, as shown by analytical totals (90.24–97.89 wt%) that are higher than the theoretical anhydrous sum (82.49 wt%). Hence, we were not able to derive the water contents from the difference of the analytical sum to 100 wt%. As the water content could not be determined directly due to the very limited amount of material available, elements present in klajite were recalculated considering the calculated water contents in order to make individual analytical data comparable. This recalculation decreased the values of standard deviations. In Table 3 the mean values and ranges of recalculated analyses as well as standard deviations and probe standards are listed. For the recalculations we used $9\text{H}_2\text{O}$ because the determined water contents of the lindackerite-group minerals range from 9 to 10 molecules per formula unit according to the published data

Table 2. Individual electron-microprobe spot-analyses and formulae of klajite (wt%).

	(1)	(2)	(3)	(4)	(5)	(6)	(7)
MnO	6.95	6.28	6.26	5.85	6.29	6.88	6.35
CuO	36.50	34.78	34.54	36.26	35.46	38.55	37.13
CaO	0.37	0.67	0.69	0.55	0.33	0.26	0.34
As ₂ O ₅	52.15	48.78	48.75	50.18	48.48	52.20	50.22
Total	95.97	90.51	90.24	92.84	90.56	97.89	94.04
Cation numbers on the basis of 30 charges							
Mn	0.87	0.83	0.83	0.75	0.83	0.84	0.81
Cu	0.05	0.06	0.05	0.16	0.11	0.12	0.14
Ca	0.06	0.11	0.12	0.09	0.06	0.04	0.05
ΣM	0.98	1.00	1.00	1.00	1.00	1.00	1.00
Cu	4.00	4.04	4.03	4.01	4.08	4.10	4.09
As	4.01	3.98	3.99	3.99	3.97	3.96	3.96

Fig. 6. Ternary diagram showing the cation contents in *M* site for klajite. Filled squares indicate the individual spot analyses.

(e.g., Sarp & Dominik, 1995; Sejkora *et al.*, 2010). The mean composition of klajite gives the following empirical formula, calculated on the basis of 30 negative charges of the anhydrous material: $(\text{Mn}_{0.82}\text{Cu}_{0.10}\text{Ca}_{0.08})_{\Sigma} = 1.00 \text{ Cu}_{4.05}\text{As}_{3.98}\text{O}_{14}(\text{OH})_2 \cdot 9\text{H}_2\text{O}$. The simplified formula is $\text{MnCu}_4\text{As}_4\text{O}_{14}(\text{OH})_2 \cdot 9\text{H}_2\text{O}$ and implies (in wt%): MnO 6.89, CuO 30.92, As₂O₅ 44.68 and H₂O 17.51, total = 100.00. The formula may also be expressed as $\text{MnCu}_4(\text{AsO}_4)_2(\text{AsO}_3\text{OH})_2 \cdot 9\text{H}_2\text{O}$ in order to emphasize the similarity to the lindackerite-group formula. We remark here that klajite is the first known Mn+Cu arsenate mineral reported so far.

6. Discussion of related minerals

The members of the lindackerite group are hydrous arsenate minerals with the general formula $\text{MCu}_4(\text{AsO}_4)_2(\text{AsO}_3\text{OH})_2 \cdot 9\text{--}10\text{H}_2\text{O}$, where *M* = Cu

(lindackerite, Hybler *et al.*, 2003), Co (pradetite, Burke *et al.*, 2007), Zn (veselovskýite, Sejkora *et al.*, 2010) and Mn (klajite, this study). The first species of the group, lindackerite, was described by Vogl (1853) from Jáchymov (St. Joachimsthal), NW Bohemia, Czech Republic; 139 years later (January 1992) pradetite was approved by IMA CNMMN (IMA no. 91-046) but its publication was delayed, pending the work on holotype lindackerite (Burke *et al.*, 2007). In the meantime Sarp & Dominik (1995) redefined “lindackerite” (IMA 91-046) on a sample from the Cap Garonne mine, Le Pradet, Var, France, implying the discreditation of pradetite as a Co-bearing variety of lindackerite. However, Hybler *et al.* (2003) revealed the importance of isomorphous substitutions in the *M* site for the nomenclature of lindackerite-type minerals and, as a consequence, a revalidation proposal was approved by IMA CNMNC for pradetite, the Co analogue of lindackerite. The group was further extended by Sejkora *et al.* (2010) who described the Zn analogue of lindackerite, veselovskýite.

It is worth mentioning that the atomic arrangement in lindackerite is very similar to that of chudobaite-group minerals (*i.e.*, chudobaite and geigerite as well as “pseudolindackerite” of Ondruš *et al.*, 1997, which was recently described as ondrušite by Sejkora *et al.*, 2011). The most important differences are the distribution of water molecules around the *M* position as well as their occupancy (for details see Hybler *et al.*, 2003). In the absence of structure determination, Sejkora *et al.* (2010) suggested the water content to be appropriate for discrimination between lindackerite- and chudobaite-group minerals, with water content of 9 and 10 molecules in a formula unit, respectively. Unfortunately, neither measured nor calculated water contents are available for klajite. However, the published (measured or calculated) water contents (see Sejkora *et al.*, 2010, 2011) do not obey strictly the above described rule, so this discrimination is rather informal.

In a most recent paper, Sejkora *et al.* (2011) put all the above mentioned species into the lindackerite group which was divided in two series, namely lindackerite-pradetite-veselovskýite and ondrušite-geigerite-chudobaite. Comparative data of all the known members of the

Table 3. Recalculated electron-microprobe data in weight per cent.

Constituent	Mean	Range	Stand. dev.	Probe standard
MnO	5.67	5.17–6.00	0.23	Spessartine
CuO	32.03	31.33–32.60	0.46	Cu ₉₄ Sn ₆
CaO	0.41	0.22–0.65	0.16	Clinopyroxene A1P
As ₂ O ₅	44.40	44.07–44.82	0.26	As ₂ S ₃
H ₂ O*	17.49	17.44–17.52	0.03	
Total	100.00			

Note: *Calculated from the stoichiometry: 9H₂O + 2(OH).

Table 4. Comparative data for the lindackerite-group minerals in which Cu occupies dominantly both Cu1 and Cu2 sites.

Mineral name	Lindackerite	Pradetite	Veselovskýite	Ondrušite	Klajite
Type locality	Jáchymov CZE	Cap Garonne FRA	Jáchymov CZE	Jáchymov CZE	Recsk HUN
Dominant cation in M site	Cu	Co	Zn	Ca	Mn
Crystal system	Triclinic	Triclinic	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$ or $P1$	$P\bar{1}$ or $P1$	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$
<i>a</i> (Å)	6.453(2)	6.434(5)	6.4022(4)	6.432(1)	6.441(3)
<i>b</i> (Å)	8.035(2)	8.059(5)	8.0118(4)	7.986(1)	7.983(4)
<i>c</i> (Å)	10.368(4)	10.399(8)	10.3665(4)	10.827(1)	10.562(3)
α (°)	86.17(3)	85.70(6)	85.491(3)	85.75(1)	85.28(4)
β (°)	79.60(3)	79.41(7)	79.377(4)	81.25(1)	80.63(5)
γ (°)	84.83(3)	84.87(7)	84.704(5)	85.04(1)	84.80(4)
<i>V</i> (Å ³)	525.9(2)	526.9(7)	519.34(4)	546.6(1)	532.4(3)
<i>Z</i>	1	1	1	1	1
<i>D</i> _{calc} (g/cm ³)	3.37(1)	3.31(1)	3.28	3.12	3.213
Colour	Apple-green	Green, light green, ash grey	Colourless, greyish white	White, whitish green, grey-green, apple-green	Greenish yellow to yellowish green

lindackerite group with Cu dominancy in Cu1 and Cu2 sites are shown in Table 4.

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