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Fülöppite, a new Hungarian mineral of the plagionite-semseyite group.

BY

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[Reprinted from the Mineralogical Magazine, December, 1929, Vol. XXII, No. 127, pp. 179-184.]

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Fülöppite, a new Hungarian mineral of the plagionite-semseyite group. 1

By I. DE FINÁLY,
Royal Hungarian Geological Survey,
and Sándor Koch, Ph.D.
Hungarian National Museum, Budapest.

[Read June 11, 1929.]

In the Autumn of 1928 Dr. Béla Fülöpp and Mr. G. Kupás sent for examination an unknown mineral, which had been found in no. III level on the main lode of the Kereszthegy mine at Nagybánya, comitat Szatmár [now Baia Mare, Satul-Mare, Romania]. The small crystals show a close resemblance to those of plagionite from Wolfsberg in the Harz Mountains, but the results of our detailed examination prove that we have here a new mineral. For this we propose the name fülöppite in honour of Dr. B. Fülöpp, an enthusiastic collector of minerals and a generous patron of the Magyar Nemzeti Múzeum (Hungarian National Museum).

The crystals are usually about a millimetre across, and some up to 2 mm., but only the smaller crystals are suitable for goniometric measurement. Calculated from the measured angles

$$(100)$$
: $(111) = 59^{\circ}$ 34', (001) : $(111) = 41^{\circ}$ 48', and (111) : $(1\overline{1}1)$ 59° 20'

the axial elements of the holohedral monoclinic crystals are

$$a:b:c=1.1087:1:0.7011, \beta=85^{\circ}15\frac{1}{2}$$

Eight forms, c(001), a(100), $d(\overline{1}01)$, e(112), p(111), $t(\overline{2}23)$,

¹ Read at the Hungarian Academy of Sciences, Class III (Natural History), on April 22, 1929. The mineralogical and crystallographic work was done by Dr. S. Koch in the Mineralogical-Petrographical Institute of the Pázmány Péter University, Budapest, and the chemical portion by I. de Finály in the Chemical Laboratory of the Royal Hungarian Geological Survey.

 $o(\overline{1}11)$, $s(\overline{2}21)$, were observed on the crystals. Other measured angles are :

	Ob-	Cal-		Ob-	Cal-
	served.	culated.	>	served.	culated.
ac = (100):(001)	85° 12′	$85^{\circ}\ 15\frac{1}{2}'$	$tt'=(\bar{2}23):(\bar{2}\bar{2}3)$	47° 0′	$47^{\circ} 2\frac{1}{2}'$
$a'c = (\bar{1}00):(001)$	94 50	$94 \ 44\frac{1}{2}$	$oc = (\bar{1}11):(001)$	44 45	44 47
$dc = (\bar{1}01):(001)$	33 35	33 37	$oo' = (\overline{1}11) : (\overline{1}\overline{1}1)$	63 30	63 $5\frac{1}{2}$
ec = (112):(001)	24 37	24 38	$od = (\overline{1}11) : (\overline{1}01)$	31 49	$31 \ \ 32\frac{3}{4}$
$ee' = (112): (1\bar{1}2)$	36 20	36 2	$op = (\bar{1}11) : (111)$	54 16	54 44
$tc = (\bar{2}23):(001)$	32 58	$33 1\frac{1}{2}$	$sc = (\bar{2}21):(001)$	64 10	64 33

The calculated angles for each of the eight forms to the three axial planes are given in the following table:

			Angle to				
			a (100).	b (010).	\widehat{c} (001).		
c(001)		•••	85° 15½′	90° 0′			
a(100)	•••			90 0	85° 15½′		
$d(\overline{1}01)$			$118 \ 52\frac{1}{2}$	90 0	33 37		
[abs. (110)]	•••	47 51	42 9	$85 \ 42\frac{1}{2}$		
e (112)			$69 \ 18\frac{1}{2}$	71 59	24 38		
p(111)		•••	59 34	60 20	41 48		
$t(\bar{2}23)$		•••	$107 \ 29\frac{1}{2}$	66 29	$33 1\frac{1}{2}$		
o (111)			114 18	58 27	44 47		
$s(\bar{2}21)$			$124 \ 33\frac{1}{2}$	47 54	64 33		

The negative hemi-pyramids are the predominating forms, and give rise to a short-prismatic habit of the crystals. On this account the crystals have been drawn (except fig. 3) showing the back view with $a'(\bar{1}00)$ in front. Crystals of thick-tabular habit with the basal plane c(001) largely developed are of rare occurrence; on such crystals the positive and negative hemi-pyramids are equally developed (fig. 3).

The larger crystals are always short-prismatic in habit, and usually show only the forms c(001), a(100), and $o(\overline{1}11)$ (fig. 1). The predominating faces of $o(\overline{1}11)$ are slightly concave and are heavily striated parallel to the intersection edge o/c. The well-developed faces of a(100) are slightly convex and striated parallel to the intersection edge a/c. The small triangular faces of c(001) are bright, but generally somewhat curved and give poor reflections. On some of the larger crystals the positive hemi-pyramid p(111) occurs as narrow striated faces (fig. 2).

The smaller crystals are richer in faces, which are generally brighter, though still sometimes slightly curved and more or less striated. Most of these crystals are also of the short-prismatic habit (fig. 4) and only a few of the thick-tabular habit (fig. 3). On seven

of the smaller crystals measured on the goniometer the forms c a e p t o are present on all; the rarer form d ($\overline{1}01$) was present on three of these crystals, and s ($\overline{2}21$) on only one. The best reflections are given by the small rhomb-shaped faces of c (001) and by the positive hemi-pyramids p (111) and e (112). The narrow faces of

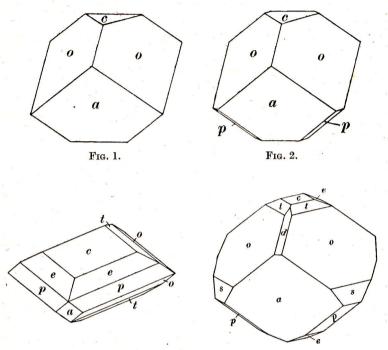


Fig. 3. Fig. 4. Crystals of fülöppite. $[a'(\overline{1}00)]$ in front in figs. 1, 2, and 4.]

 $d(\overline{1}01)$ also give good reflections. a(100) is curved and striated. The worst reflections are those from the curved and striated negative hemi-pyramids.

No twinned crystals or parallel overgrowths were observed. Groups consist of individual crystals irregularly grown over one another.

The mineral is brittle with uneven fracture and shows no cleavage. The smaller crystal faces show a lead-grey colour with bright metallic lustre, and the striated faces a steel-grey with feeble lustre. The mineral sometimes shows a steel-blue, or less often a bronzy, tarnish. The streak is reddish-grey and the hardness rather greater than 2.

The mineral is readily fusible before the blowpipe, and gives yellow and white deposits on the charcoal. It melts easily in the open tube, yielding sulphurous vapours and a deposit of Sb₂S₃. It is not attacked by concentrated hydrochloric nor by concentrated nitric acid.

A small crystal was embedded in sealing-wax and polished for metallographic examination. It was very soft and polished easily to a brilliant white surface. This surface was treated with various reagents. Negative results were obtained with HNO₃ (1:1), HCl (1:1), KCN (20%), FeCl₃ (20%), and HgCl₂ (5%). With KOH (40%) acting for 60 seconds a slight brown tarnish was formed, which was easily wiped off with a wet rag, the mineral not being attacked. With aqua regia a brownish-yellow, later bluish, tarnish is developed in 20 seconds; this can be rubbed off, the surface then being grey. After longer action (50–60 seconds) the mineral becomes dark grey with here and there minute yellowish specks.

The specific gravity was determined in a small pyknometer on the carefully selected crystals used for the chemical analysis and weighing 0.3052 gram. The result, 5.24, when corrected is $D_4^{20}=5.23$.

The qualitative analysis showed the presence of lead, antimony, and sulphur. Lead and antimony were estimated in one portion (0·1606 gram) of the material which was decomposed in a current of chlorine. The lead chloride was washed out with warm water and the lead precipitated and weighed as PbSO₄. Antimony was precipitated by hydrogen sulphide and after heating in a current of carbon dioxide weighed as Sb₂S₃. Sulphur was estimated in a separate portion (0·0368 gram) according to the method of Fresenius, with the precautions and corrections prescribed by Winkler. The following results were obtained:

				Atomic	ratios.	Calculated for Pb ₂ Sb ₆ S ₁₁ .
S	•••	•••	24.10%	0.7524	11.01	23.56
Sb		•••	47.50	0.3904	5.71	48.78
Pb		• • •	28-29	0-1367	2	27.66
SiO_2	• • •	•••	0.19			
			100.08			100.00

¹ F. P. Treadwell, Kurzes Lehrbuch anal. Chem., 1922, vol. 2, p. 307.

² L. W. Winkler, Zeits. angew. Chem., 1922, vol. 35, p. 662.

³ F. P. Treadwell, loc. cit., vol. 2, p. 180.
⁴ F. P. Treadwell, loc. cit., vol. 2, p. 306.

⁵ L. W. Winkler, Zeits. angew. Chem., 1917, vol. 30, p. 251; 1920, vol. 33, pp. 59, 159.

Although the antimony is rather low, the chemical composition of fülöppite may therefore be expressed by the formula

$$Pb_2Sb_6S_{11}$$
 or $2PbS.3Sb_2S_3$.

Fülöppite differs from the other members of the plagionite-semseyite group in containing antimony sulphide in molecular excess over lead sulphide, and it is one of the few known acid sulphantimonites.

The data obtained give some support to the suggestion made by L. J. Spencer¹ that these minerals form a morphotropic series. With increasing lead there is an increase through the series in the length of the c-axis whilst the a-axis remains practically unchanged.

			a : b : c.	β.
Fülöppite,2	$3PbS.4Sb_2S_3$		1.1087:1:0.7011	85° 15‡′
Plagionite,3	$5 PbS.4 Sb_2S_3$		$1 \cdot 1305 : 1 : 0 \cdot 8422$	72 45
Heteromorphite,	$7 PbS.4 Sb_2S_3$		~ .	_
Semseyite,4	$9Pb8.4Sb_2S_3$	***	$1 \cdot 1356 : 1 : 1 \cdot 0218$	74 14

Fülöppite usually forms crystals of short prismatic or rhombohedral habit; plagionite forms small crystals of tabular habit; and semseyite assumes a tabular or prismatic habit.

Another lead antimony mineral is present in intimate association with the fülöppite. This forms radiating or globular groups of fine needles of a steel-grey to black colour with feeble metallic lustre and sometimes a blue tarnish. Some hairs of plumosite are perhaps also present. The acicular mineral is brittle with uneven fracture, dark grey streak, and hardness about 2. The very thin needles yielded no results on the goniometer. Specific gravity 5.23, corrected $D_4^{20} = 5.22$. Analysis gave the following results:

			A	tomic ratios.		Calculated for Pb ₃ Sb ₈ S ₁₅ .
\mathbf{s}	•••		23.10 %	0.7306	15.27	23.17
Sb		•••	46.17	0.3844	8.04	46.91
$\mathbf{P}\mathbf{b}$		•••	29.33	0.1436	3	29.92
\mathbf{Fe}	•••		0.08	_	-	_
SiO_2	•••		0.94		_	-
			$\overline{99.62}$	*		100.00

Formula Pb₂Sb₈S₁₅ or 3PbS.4Sb₂S₃. The small excess of sulphur shown in the atomic ratios can be explained by the presence of a few

¹ L. J. Spencer, Min. Mag., 1899, vol. 12, p. 55.

² The analysis is actually between 3PbS.4Sb₂S₃ and 2PbS.3Sb₂S₃., as may be seen from the calculated values for the formula given below.

³ Axial ratios of F. Zambonini, Rivista Min. Crist. Ital., 1912, vol. 41, p. 21.

⁴ Axial ratios of G. F. H. Smith. Min. Mag., 1919, vol. 18, p. 357.

minute crystals of sulphur adhering to the needles. This mineral, with practically the same chemical composition as fülöppite, is also an acid sulphantimonite of lead. The analyses of both give percentages of lead and antimony between the limits required of the formulae 2PbS.3Sb₂S₃ and 3PbS.4Pb₂S₃. With the presence of some plumosite (or even possibly stibnite 1), as noted above, the formula is open to some doubt. From the acicular habit of the crystals the mineral may be presumed to be orthorhombic, and it is provisionally referred to keeleyite, a mineral recently described by S. G. Gordon 2 from Oruro, Bolivia, to which he gave the formula 2PbS.3Sb₂S₃. Later examinations 3 of keeleyite suggest, however, that it is identical with zinckenite PbS.Sb₂S₃.

Other minerals present on the fülöppite specimens are dark brown blende, small quartz crystals, a few small brown rhombohedra of dolomite, and a few minute crystals of sulphur.

¹ L. J. Spencer, Note on 'feather-ore' . . . Min. Mag., 1907, vol. 14, p. 207.

² S. G. Gordon, Proc. Acad. Nat. Sci. Philadelphia, 1922, vol. 74, p. 101 [Min. Abstr., 2–11].

³ E. V. Shannon and M. N. Short, Amer. Min., 1927, vol. 12, p. 405; E. T. Wherry, ibid., 1928, vol. 13, p. 29 [Min. Abstr., 3-453].

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