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GABRIELITE, $Tl_2AgCu_2As_3S_7$, A NEW SPECIES OF THALLIUM SULFOSALT FROM LENGENBACH, BINNTAL, SWITZERLAND

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Abstract

Gabrielite, $Tl_2AgCu_2As_3S_7$, is a new species of sulfosalt mineral occurring in the famous Lengenbach locality, at Binntal, Canton Valais, Switzerland. It was discovered in association with numerous other As sulfosalts, such as hutchinsonite, hatchite, edenharterite, trechmannite, tennantite, and abundant realgar and rathite, generally occurring as idiomorphic crystals in cavities of a Triassic dolomitic rock. The whole region was transformed during the Alpine orogeny to the greenschist to garnet-amphibolite facies of metamorphism. Gabrielite occurs as pseudohexagonal crystals; its true symmetry is triclinic, space group $P\bar{1}$, $Z = 6$, a 12.138(3), b 12.196(3), c 15.944(4) Å, α 78.537(5), β 84.715(4), γ 60.470(4)°, V 2013(1) Å^3 (from the crystal-structure determination). The strongest powder-diffraction lines [d_{obs} in Å(I_{obs})(hkl)] are 15.63(100)(001), 3.143(90)($21\bar{4}$, $1\bar{1}4$), 3.531(80)(300), 2.911(70)(422, $2\bar{1}\bar{4}$), 2.520(60)($2\bar{2}3$), and 2.978(60)(324). Gabrielite is opaque with a grey to black color, metallic luster and blackish red streak. Individual crystals reach up to approximately 0.4 mm, and aggregates may exceed 1 mm across. Euhedral crystals are short prismatic to platy parallel to (001), with excellent cleavage along this face. The microhardness VHN,18 kg/mm^2 (load 10 g), corresponds to a Mohs hardness of 1½–2. The chemical composition (electron microprobe, mean result of 11 analyses, in wt%) is Tl 37.30, Cu 12.50, Ag 8.47, As 18.90, Sb 1.79, S 20.64, total 99.70 wt%, from which the empirical formula, $Tl_{5.95}(Ag_{2.56}Cu_{6.46})_{\Sigma 9.02}(As_{8.23}Sb_{0.48})_{\Sigma 8.71}S_{21}$, was derived. The simplified formula is $Tl_2AgCu_2As_3S_7$. The calculated density D_{calc} is 5.38 g/cm^3. In reflected light, gabrielite is white, has common red internal reflections, shows weak anisotropy and moderate reflectivity [%R_{max} in air (COM wavelengths in nm)]: 30.53(470), 29.10(546), 27.94(589), 26.35(650). The crystal structure is layered parallel to (001), which explains the habit and cleavage. The twin law with (100) as a twin plane is due to the local symmetry properties of the structural layers. A presence of metal–metal (Tl–Tl and Tl–Cu) and metal–semimetal (Tl–As) bonds in the structure explains the low hardness and metallic character. Substitutions of Cu for Ag in the tetrahedral coordinations, Ag for Cu in the trigonal–planar coordinations, and Sb for As in isolated AsS_3 coordination polyhedra and As_4S_8 rings lead to the structural formula $Tl_6(Ag,Cu)_3{}^{IV}(Cu,Ag)_6{}^{III}[(As,Sb)S_3]_3[(As,Sb)_2S_4]_3$; the empirical formula obtained from the microprobe data is $Tl_{5.95}(Ag_{2.56}Cu_{6.46})_{\Sigma 9.02}(As_{8.23}Sb_{0.48})_{\Sigma 8.71}S_{21}$. The name of the mineral honors Walter Gabriel, of Basel, Switzerland, well-known mineral photographer and expert of Lengenbach minerals.

Keywords: gabrielite, new mineral species, sulfosalt, thallium, Lengenbach, Switzerland.

Sommaire

La gabrielite, $Tl_2AgCu_2As_3S_7$, est une nouvelle espèce de sulfosel découverte à la carrière Lengenbach, localité bien connue à Binntal, Canton de Valais, en Suisse. Elle a été découverte en association avec de nombreux autres sulfosels d'arsenic, par exemple hutchinsonite, hatchite, édenharterite, trechmannite, tennantite, avec réalgar abondant et rathite, en général sous forme de cristaux idiomorphes dans des cavités d'une roche dolomitique triassique. La région entière a été transformée par métamorphisme alpin au faciès schistes verts ou amphibolite à grenat. La gabrielite se présente en cristaux pseudohexagonaux; par contre, sa vraie symétrie est triclinique, groupe d'espace $P\bar{1}$, $Z = 6$, a 12.138(3), b 12.196(3), c 15.944(4) Å, α 78.537(5), β 84.715(4), γ 60.470(4)°, V 2013(1) Å^3 (maille établie par affinement de la structure). Les raies les plus intenses du spectre de diffraction X (méthode des poudres) [d_{obs} en Å(I_{obs})(hkl)] sont 15.63(100)(001), 3.143(90)($21\bar{4}$, $1\bar{1}4$), 3.531(80)(300), 2.911(70)(422, $2\bar{1}\bar{4}$), 2.520(60)($2\bar{2}3$), et 2.978(60)(324). La gabrielite est opaque, grise à noire, avec un éclat métallique et une rayure rouge noirâtre.

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Les cristaux individuels atteignent en général environ 0.4 mm, et les aggrégats peuvent dépasser 1 mm. Les cristaux idiomorphes sont des prismes courts ou des plaquettes parallèles à (001) et au clivage le long de cette face. La microdureté VHN,18 kg/mm^2 (charge de 10 g), correspond à une dureté de Mohs de 1½–2. La composition chimique, établie avec une microsonde électronique (résultat moyen de 11 analyses, en %, base pondérale) est: Tl 37.30, Cu 12.50, Ag 8.47, As 18.90, Sb 1.79, S 20.64, total 99.70 wt%, ce qui mène à la formule empirique: $Tl_{5.95}(Ag_{2.56}Cu_{6.46})_{\Sigma 9.02}(As_{8.23}Sb_{0.48})_{\Sigma 8.71}S_{21}$. La formule simplifiée est $Tl_2AgCu_2As_3S_7$. La densité calculée D_{calc} est égale à 5.38 g/cm^3. En lumière réfléchie, la gabrielite est blanche; elle montre de nombreuses réflections internes, et possède une faible anisotropie et une réflectivité moyenne [%R_{max} mesurée dans l'air (longueurs d'onde de COM en nm)]: 30.53(470), 29.10(546), 27.94(589), 26.35(650). La structure cristalline possède des couches parallèles à (001), ce qui explique la morphologie et le clivage. La loi de macle, avec (100) comme plan de macle, découle des propriétés locales de symétrie de ces couches. La présence de liaisons de métal à métal (Tl–Tl et Tl–Cu) et de métal à semimétal (Tl–As) rend compte de la faible dureté et du caractère métallique. Les substitutions de Cu au Ag sur le site tétraédrique, de Ag au Cu sur le site trigonal–planaire, et du Sb au As dans les polyèdres isolés AsS_3 et les anneaux As_4S_8 mènent à la formule structurale $Tl_6(Ag,Cu)_3{}^{IV}(Cu,Ag)_6{}^{III}[(As,Sb)S_3]_3[(As,Sb)_2S_4]_3$; la formule empirique obtenue à partir des données de microsonde électronique est $Tl_{5.95}(Ag_{2.56}Cu_{6.46})_{\Sigma 9.02}(As_{8.23}Sb_{0.48})_{\Sigma 8.71}S_{21}$. Le nom de l'espèce honore Walter Gabriel, de Bâle, en Suisse, photographe de minéraux bien connu et expert des minéraux de Lengenbach.

(Traduit par la Rédaction)

Mots-clés: gabrielite, nouvelle espèce minérale, sulfosel, thallium, Lengenbach, Suisse.

INTRODUCTION

Gabrielite, ideally $Tl_2AgCu_2As_3S_7$, is a new species of sulfosalt mineral found at Lengenbach, in Binntal, Ct.Valais, Switzerland, a locality well known for its remarkable occurrence of numerous rare and unique sulfosalts. It has provided mineralogists and crystallographers all over the world with interesting research material for more than 200 years. The main mineralized zone is small, not exceeding a volume of about 50 × 30 × 30 meters within a dolomitic rock of presumably Triassic age. The host rock displays mostly a snow-white color, it is partly a compact, marble-like rock, yet, on the other hand, it may occur as a soft material of powdery consistency that can be pulverized even between the fingers ("sugary" dolomite). Undoubtedly, hydrothermal activity played an important role in the alteration of the host rock as well as in the formation of cavities and the crystallization of rare sulfosalt minerals within them. The arsenic necessary for the formation of the various As-rich sulfosalts in the dolomite presumably originated in the adjoining gneisses and migrated into the dolomite either during Alpine metamorphism, as postulated by the first author (Graeser 1965, 1998), or at a much earlier time, during the diagenesis of the dolomitic sediments (Hofmann & Knill 1996).

The name of the new mineral species honors Walter Gabriel, of Basel (b. 1943), a chemical technician who, in his spare time, has photographed thousands of Lengenbach specimens for books and publications. He has studied Lengenbach minerals for decades, and has gained considerable knowledge of these uncommon minerals.

The new mineral and its name have been approved by the Commission on New Minerals and Mineral Names, I.M.A. (IMA 2002–053). Type material is preserved at the Natural History Museum Basel and the Institute of Mineralogy, University of Basel (X-ray samples) under the catalogue number S113.

THE MINERALS OF LENGENBACH

The locality has yielded more than 120 different mineral species so far, and it is remarkable that almost half of these minerals are sulfides and sulfosalts. The sulfosalt minerals of Lengenbach belong almost exclusively to the group of arsenic sulfosalts, hosting additional elements like Pb, Tl, Cu, Ag; about 26 species of them were discovered here for the first time (type-locality). Fourteen Tl-sulfosalts have been discovered here during the past 20 years, of which 11 are new minerals. A recent description of the locality and its minerals is given in Graeser (1998).

Mineral collecting at Lengenbach dates back at least to the year 1797 (proven by a dated sample preserved in the Natural History Museum in Basel). Systematic exploitation of minerals for research by various syndicates and companies started around 1833, and has continued with interruptions caused by the two world wars up to the present time. Currently, works in Lengenbach are being carried out by the "Forschungsgemeinschaft Lengenbach", a syndicate consisting of various mineral collectors and mineralogists, under the supervision of the municipality of Binn.

OCCURRENCE

A sample (L21913) collected in 1989 by the syndicate working at that time in Lengenbach ("Arbeitsgemeinschaft Lengenbach"), contained some crystals with a peculiar shape that caught our attention. An X-ray powder-diffraction pattern demonstrated that it is different from any other Lengenbach mineral known so far. Unfortunately, the crystals on the sample were not suitable for a thorough study, either with respect to quality or to quantity. Further finds in 1990, and 1995, in particular, provided material more suitable for study, and the results are based mainly on this material.

With a Tl content of 37.4 wt%, gabrielite is among the minerals richest in Tl occurring at Lengenbach. Only imhofite, $Tl_3As_{7.67}S_{13}$, with 37.6 wt%, contains slightly more Tl. As in the case of the other minerals of Tl, gabrielite is only found in the zone with elevated Tl and As contents, in the southern part of the quarry, and consequently it is commonly accompanied by realgar, rathite, trechmannite, tennantite, hutchinsonite, hatchite, edenharterite and other Tl-rich sulfosalts. All these minerals crystallized in small irregularly shaped cavities in the dolomitic rock. The Tl-rich minerals, in particular, are the products of the latest stage of hydrothermal activity at Lengenbach.

PHYSICAL AND OPTICAL PROPERTIES

It was the very peculiar shape of some of the very small crystals (individual crystals not exceeding 0.3–0.4 mm), that piqued scientific interest in the mineral; the crystals show a conspicuous sixfold outline, though from the distinct striation parallel to the crystallographic *a* axis on the largest face, hexagonal symmetry can be ruled out (Fig. 1). In one case, an aggregate of loosely intergrown crystals attained a diameter of more than 1 mm. As is the case for most Tl-rich sulfosalts, it is not possible to unequivocally identify the mineral based on physical properties and visual inspection alone (Table 1). In its anhedral form, gabrielite is almost indistinguishable from other Lengenbach Tl-minerals, in particular sicherite, $TlAg_2(As,Sb_3)S_6$. Therefore, any material used for analytical work had to be verified first by X-ray diffraction (Gandolfi camera). However, the pseudohexagonal shape (where present), the perfect cleavage, the black metallic appearance and the reddish streak (clearly visible on broken edges) might give some indication of the kind of mineral. With a Mohs hardness of 1½–2 (according to VHN indentations measured on a Leitz Durimet microscope), gabrielite has the lowest hardness among all Lengenbach Tl-sulfosalts. In polished section under reflected light, gabrielite displays a white color with common red internal reflections. Anisotropy is weak; no bireflectance or pleochroism was observed. Reflectance values are moderate and close to those of most other Tl sulfosalts (Table 2).

TABLE 1. PHYSICAL AND OPTICAL PROPERTIES OF GABRIELITE

Morphology	pseudohexagonal, platy along (001)
Striations	parallel to *a* axis
Forms	{001} dominant, several not identified (*hk*0) faces
Color	dark grey to black, opaque
Luster	metallic
Streak	blackish red
Cleavage	excellent along (001)
Fracture	uneven to conchoidal
Twinning	common [along (100), suggested by structure determination]
Hardness	$VHN_{10} = 18$ (only two indentations)
	Mohs = 1½–2 (according to VHN values)
Density	5.38 g/cm^3 (calculated)

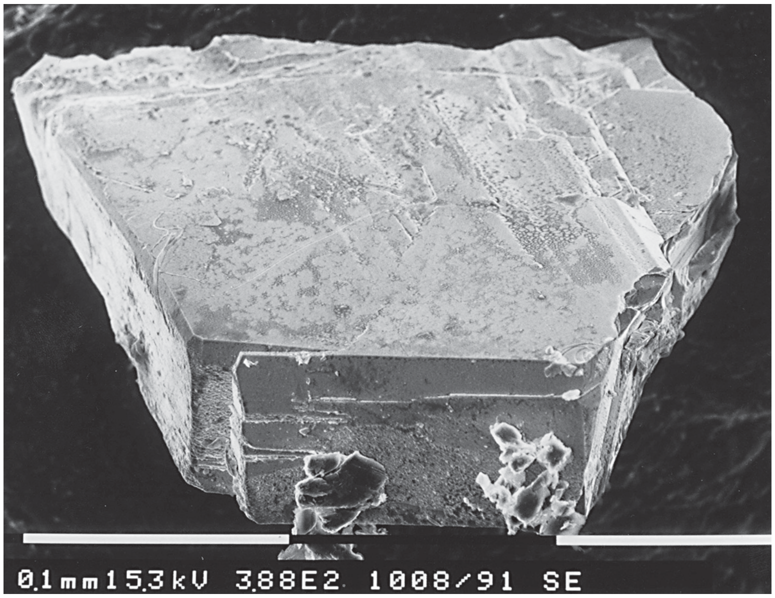

FIG. 1. Gabrielite, Lengenbach. SEM picture showing the dominant (001) face with striation along *a* axis and (presumable) twinning along (100).

CHEMICAL COMPOSITION

Preliminary qualitative chemical anlayses using a Hitachi S–2500 scanning electron microscope (SEM) revealed the presence of Tl, Cu, Ag, As, Sb, and S only. Quantitative analyses were performed using an electron microprobe (JEOL JXA–8600; LINK–eXL). Chemical data are reported in Table 3. From the observed ranges and the results of the crystal-structure analysis, it can be expected that Sb can substitute in even larger amounts for As. It is, however, difficult to predict whether an increased level of substitution would bring a structural change like in the case of the two Tl-sulfosalts edenharterite $TlPbAs_3S_6$ (Graeser & Schwander 1992) and jentschite $TlPbAs_2SbS_6$ (Graeser & Edenharter 1997), where the ordered distribution of Sb in the structure led to a lowering in symmetry from orthorhombic to monoclinic. A more probable reason for a change of symmetry could be a different proportion of Cu and Ag in the structure. The crystal-structure analysis pointed to an ordered partial substitution of these two elements on some structural sites as the main reason for the lowering of symmetry from an idealized monoclinic to the observed triclinic (Balić-Žunić *et al.* 2006). Although no close structural relations are observed between them, some of the Tl sulfosalts from Lengenbach show a conspicuous geometrical relationship (Table 4).

X-RAY DIFFRACTION

The mineral yielded excellent powder patterns (Debye–Scherrer and Gandolfi cameras) that are unlike those of any known mineral species. In combination with SEM data, it is obvious that the mineral represents a new complex sulfosalt phase with Tl, Cu, Ag, As, and Sb. Consequently, a preliminary single-crystal study was carried out using Weissenberg and precession techniques which, at first, did not lead to a satisfactory result. Preliminary studies led to (pseudo) monoclinic symmetry, presumably due to the presence of twinning phenomena; attempts to split the twins into unequivocal single crystals were unsuccessful. The true triclinic symmetry resulted from the subsequent structure-determination. The powder pattern was thereafter indexed on the basis of a triclinic cell. Cell parameters refined from the powder data are in good agreement with those obtained from the structure determination. An indexed powder-pattern is reported in Table 5, and unit-cell dimensions are listed in Table 6.

TABLE 2. THE REFLECTANCE OF GABRIELITE

Wavelength (nm)	R_{max} (%)	Wavelength (nm)	R_{max} (%)
400	32.39	560	28.78
420	31.32	580	28.23
440	30.84	**589**	27.94
460	30.68	600	27.67
470	30.53	620	27.23
480	30.49	640	26.75
500	30.17	**650**	26.35
520	29.76	660	26.03
540	29.35	680	25.49
546	29.10	700	24.89

* WTiC standard. The values were measured in air. Values in bold: COM wavelengths.

TABLE 3. CHEMICAL COMPOSITION OF GABRIELITE

	wt%*	range	st.dev.	probe standard	at.ratio†
Tl	37.30	37.05 – 37.48	0.11	Lorandite, Ti*L*α	5.95
Cu	12.60	12.35 – 12.80	0.13	Chalcopyrite, Cu*K*α	6.46
Ag	8.47	8.35 – 8.68	0.13	Metal (synthetic), Ag*L*α	2.56
As	18.90	18.64 – 19.06	0.15	Lorandite, As*K*α	8.23
Sb	1.79	1.44 – 2.25	0.30	Stibnite, Sb*L*α	0.48
S	20.64	20.40 – 20.91	0.14	Lorandite, S*K*α	21.00
Total	99.70		0.24		

* Mean result of 11 analyses. Acceleration voltage: 25 kV, beam current: 30 nA, correction procedure: ZAF.

† Basis: 21 S atoms: $Tl_{5.95}(Ag_{2.56}Cu_{6.46})_{\Sigma 9.02}(As_{8.23}Sb_{0.48})_{\Sigma 8.71}S_{21}$. Simplified formula: $Tl_2AgCu_2As_3S_7$.

TABLE 4. SELECTED Tl–As–Sb SULFOSALTS FROM LENGENBACH

Name	Formula	*a* (Å)	*b* (Å)	*c* (Å)	β (°)	*V* (Å³)	system	ref.
Imhofite	$Tl_3As_{7.67}S_{13}$	8.775	24.425	11.478	108.28	2331	monoclinic	1
Edenharterite	$TlPbAs_3S_6$	15.476	47.602	5.849	90.00	4292	orthorhombic	2
Jentschite	$TlPbAs_2SbS_6$	8.121	23.969	5.847	107.68	1084	monoclinic	3
Sicherite	$TlAg_2As_3S_6$	15.522	12.479	5.719	90.00	1108	orthorhombic	4
Gabrielite	$Tl_2AgCu_2As_3S_7$	15.944	12.196	12.138	84.71	2013	triclinic	5

References: 1) Burri *et al.* (1965), 2) Graeser & Schwander (1992), 3) Graeser & Edenharter (1997), 4) Graeser *et al.* (2001), 5) This study; *a* and *c* parameters interchanged.

TABLE 5. POWDER-DIFFRACTION DATA FOR GABRIELITE

h	*k*	*l*	d_{obs}	d_{calc}*	I_{obs}	I_{calc}†
0	0	1	15.631	15.630	100	100
1	2	$\bar{1}$	5.237	5.236	40	11
2	1	2	5.030	5.027	30	16
3	2	1	3.944	3.943	40	14
1	3	0		3.922		18
3	0	0	3.531	3.531	80	53
2	3	3	3.468	3.473	40	22
3	1	$\bar{2}$		3.459		21
0	3	$\bar{1}$	3.263	3.279	50	23
3	3	$\bar{1}$		3.253		15
2	1	$\bar{4}$	3.143	3.144	90	52
1	$\bar{1}$	4		3.140		49
2	4	1	3.049	3.050	40	18
1	2	5		3.035		32
3	2	4	2.978	2.974	60	55
4	2	2	2.911	2.917	70	17
2	$\bar{1}$	$\bar{4}$		2.907		51
1	4	3	2.767	2.766	10	15
2	4	$\bar{2}$	2.620	2.618	40	26
2	$\bar{2}$	3	2.520	2.519	60	35
3	$\bar{1}$	$\bar{4}$	2.417	2.416	20	10
1	$\bar{1}$	6	2.299	2.307	40	12
3	1	6	2.244	2.244	10	6
4	2	$\bar{5}$	2.076	2.074	20	10
3	6	0	1.991	1.991	30	8
1	6	1	1.904	1.902	20	11
3	$\bar{2}$	5	1.843	1.842	20	9
			1.794		30	
			1.723		20	
			1.633		20	
			1.571		20	

Notes: 114.6 mm Debye–Scherrer camera, Fe*K*α radiation.

* d_{calc} values for the cell *a* 12.152(7), *b* 12.203(7), *c* 15.965(5) Å, α 78.25(6), β 84.55(6), γ 60.65(7)° (refined from powder-diffraction data).

† Intensities calculated from structural data (Lazy Pulverix; Yvon *et al.* 1977).

DESCRIPTION OF THE STRUCTURE

The structure of gabrielite is the subject of a companion paper (Balić-Žunić *et al.* 2006). Here we mention only the main features that are responsible for the physical properties described.

The crystal structure is layered parallel to {001}, which explains both the platy habit and an excellent cleavage parallel to this form. There are three different types of layers present. Two of them are with internal trigonal symmetry, whereas the third has an internal monoclinic symmetry. The only symmetry elements that are shared by all three layers are centers of symmetry, resulting in an overall triclinic symmetry of the structure. This peculiar combination results in a crystal lattice very close to a monoclinic *C*-centered one, and is the reason for the pseudohexagonal outline of crystals, and for the nearly equal lengths of the *a* and *b* periods and a γ angle close to 60°. The pseudosymmetry of the crystal lattice is connected also to the twinning, with (100) as the twin plane. The local symmetry properties of layers suggest a possibility for polytypism. Both a monoclinic and a trigonal polytype can be expected, with a doubled and a tripled *c* axis periods, respectively.

An extraordinary property of the crystal structure is the presence of short metal–metal and metal–semimetal bonds (Tl–Cu of 3.03 to 3.08 Å, Tl–Tl of 3.09 Å, and Tl–As of only 3.11 Å), all of which are the shortest registered among sulfosalts so far. The metallic luster and a very low hardness may be linked to the influence of these metal–metal bonds in the structure.

The crystal structure contains isolated AsS_3 groups and As_4S_8 rings. Partial substitution of Sb for As is observed only in some of the As sites. The substitution is most probably limited by the size requirements and the bonding scheme, because Sb is found only in sites where the surrounding atoms enclose smaller volumes, and is not recorded in sites with short As–Cu distances. Cu and Ag atoms appear mixed in two types of coordinations with S, with Cu dominating in trigonal–planar, and Ag in tetrahedral ones. Owing to these preferences, it is probable that both elements are necessary to stabilize the structure, and no pure Cu or Ag variants are possible. The structural formula of gabrielite can be written as $\mathrm{Tl_6{}^{IV}(Ag,Cu)_3{}^{III}(Cu,Ag)_6[(As,Sb)S_3]_3[(As,Sb)_2S_4]_3}$, with IV and III denoting the tetrahedral and trigonal planar coordinations, and As (Sb) grouped according to the polymerization of the coordination polyhedra. The shortened form of the formula, with minor substitutions ignored, is $\mathrm{Tl_2AgCu_2AsS_3As_2S_4}$.

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Markus Ecker and Dr. Thomas Raber (members of the "Forschungsgemeinschaft Lengenbach") kindly provided their samples of gabrielite for the study. We are greatly obliged to R. Guggenheim, M. Düggelin and D. Mathys (SEM laboratory, University of Basel) for their help in acquiring the SEM pictures and performing the chemical analyses of the minerals). Eric Reusser (ETH Zürich) performed the EMP analyses of gabrielite. The first samples containing the new mineral species were put at our disposal by the "Arbeitsgemeinschaft Lengenbach", the syndicate exploiting the Lengenbach minerals until 1998. We thank Yves Moëlo, Nantes, and Franklin F. Foit, Jr., of Washington State University, and Associate Editor Nigel J. Cook for critical reviews and constructive comments, which greatly improved the manuscript.

TABLE 6. UNIT-CELL DATA FOR GABRIELITE

Crystal system, space group:		triclinic, $P\bar{1}$			
	CCD data	refined*		CCD data	refined*
a (Å)	12.138 (3)	12.152 (7)	α (°)	78.537 (5)	78.25 (6)
b (Å)	12.196 (3)	12.203 (7)	β (°)	84.715 (4)	84.55 (6)
c (Å)	15.944 (4)	15.965 (5)	γ (°)	60.470 (4)	60.65 (7)
V (Å³)	2013 (1)	2020 (2)			

* refined from powder-diffraction data.

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