Macphersonite, a new mineral from Leadhills, Scotland, and Saint-Prix, France a polymorph of leadhillite and susannite

A. LIVINGSTONE

Department of Geology, Royal Scottish Museum, Chambers Street, Edinburgh EH1 1 JF, Scotland

AND

H. SARP

Département de minéralogie et de pétrographie, Muséum D'Histoire Naturelle, Case Postale 284, Genève 6, Switzerland

ABSTRACT. Macphersonite is white, resinous to adamantine, hardness (Mohs) $2\frac{1}{2}$ -3, density 6.50-6.55 gm/cm^3 and possesses a perfect cleavage on {010}. Optically it is negative with $2V_{\alpha}$ 35-36°, $\alpha = 1.87$, $\beta =$ 2.00 and $\gamma = 2.01$, $\alpha = b$, $\beta = c$, and $\gamma = a$, dispersion r > v. Polysynthetic twinning, with either coarse or fine lamellae, is common, as are contact twins. Crystals are orthorhombic, tabular on b with a 10.37, b 23.10 and c 9.25 Å, cell volume 2215.8 Å³ and space group Pcab; Z is 8 formula units. The seven strongest lines in the X-ray powder pattern are 3.274 (50) 052; 3.234 (100) 251; 2.654 (90) 351, 203; 2.598 (30) 172, 400; 2.310 (30) 004, 371, 0.10.0; 2.182 (30) 263; and 2.033 (30) 234, 452, 154, 1.10.2. Electronprobe-microanalysis-determined chemistry leads to the empirical formula (Pb_{4.08}, Cu_{0.10}, Cd_{0.07})_{24.25}S_{0.90} $C_{2.18}O_{10.55}(OH)_{1.58}$ which yields the ideal formula $Pb_4(SO_4)(CO_3)_2(OH)_2$ and hence macphersonite is a polymorph of leadhillite and susannite. The infrared spectrum shows basic similarities to leadhillite and susannite spectra with additional diagnostic absorption bands. Macphersonite shows an identical thermogravimetric behaviour to that of leadhillite. It is associated, in varying combinations, with leadhillite, susannite, cerussite, caledonite, pyromorphite, scotlandite, galena, and the 'lead hydroxyapatite' of Temple (1955). The new mineral mimics leadhillite.

WITHIN the Heddle collection, at the Royal Scottish Museum, a small quartz specimen (approximately $2 \times 4 \times 5$ cm) from Leadhills Dod, Leadhills, Lanarkshire, Scotland, bore the label 'Leadhillite? Anglesite?' In order to resolve the dilemma an X-ray powder photograph of the mineral in question, which in colour and morphology neither resembles leadhillite nor anglesite, became essential. The resulting powder photograph could not be matched with that of any known mineral or synthetic inorganic analogue, and further work revealed the mineral to be a new polymorph of leadhillite and susannite.

Subsequently, a submission to the IMA resulted in the new species, and name, being approved. During the closing stages of the voting period a second submission for an identical phase from the Argentolle Mine, near Saint-Prix, Saône-et-Loire, France, reached the IMA. Consequently, both authors considered a joint publication to be in the best interests of mineralogy. With regard to the Saint-Prix material this was collected, as leadhillite, at the Mine by Jean-Louis Passaqui, from Dijon, and arrived at the Geneva Museum in the care of Eric Asselborn, a mineral collector.

The mineral is named after Dr Harry Gordon Macpherson, Keeper of Minerals at the Royal Scottish Museum, Edinburgh, Scotland.

Physical and optical properties. Macphersonite from Leadhills is very pale amber en masse though colourless in thin flakes whereas that from the Argentolle mine is colourless to white. The lustre is adamantine on fresh surfaces otherwise resinous, fracture uneven, and Mohs hardness $2\frac{1}{2}$ -3. Dilute hydrochloric and nitric acids are dissolvents that generate effervescence and a complete rapid dissolution; in the latter acid, a curdy white $PbSO_4$ precipitate forms. Using a Berman balance on a 29.15 mg Leadhills fragment a density of 6.55 gm/cm³ resulted whereas the measured Argentolle macphersonite density is 6.50 gm/cm³. Densities calculated from empirical formulae are 6.65 and 6.60 gm/cm³ respectively. Cleavage is perfect on $\{010\}$ and the mineral is tabular on b. A very strong, vivid yellow fluorescence under long and short

wave ultraviolet is displayed by Leadhills macphersonite although this fluorescence is characteristic of several Leadhills minerals. The Argentolle macphersonite does not fluoresce. Very slight morphological differences exist between the two types for the rare Leadhills macphersonite (fig. 1) is a 'composite crystal' ($2 \times 3 \times 7$ mm) displaying interpenetrant growth which contrasts with the abundant pseudohexagonal, tabular to micaceous crystals (2-5 mm) from the Argentolle Mine. The morpho-



FIG. 1. Dumb-bell shaped composite crystal $(2 \times 3 \times 7 \text{ mm})$ of macphersonite projecting from cavity, showing interpenetrant growth. The cleavage runs parallel to the crystal length. Leadhills.

logies of a cleavage fragment (Leadhills) and an Argentolle crystal are shown in fig. 2, together with the optic orientation.

Detailed optical examination suggests orthorhombic symmetry with a 2 V_x of 35-36° and hence optically negative. Due to reactivity with high refractive index liquids the optical constants for the Argentolle macphersonite are calculated values. In this case α (1.87) is derived from the axial angle and both β (1.98) and γ (2.00) are calculated from reflectivity measurements made on- orientated polished sections. These values are in very good agreement with those actually determined for Leadhills macphersonite where $\alpha = 1.87$, $\beta = 2.00$, and $\gamma = 2.01$. Optic orientation is $\alpha = b$, $\beta = c$, and $\gamma = a$. Moderate dispersion was observed for Leadhills macphersonite with r > v and thus being opposite to strong leadhillite dispersion.

In keeping with leadhillite, twinning is common in macphersonite also. Leadhills macphersonite displays lamellar twins with both broad and fine lamellae, the composition plane being parallel to $\{102\}$. Twinned crystals of Argentolle macphersonite may attain 1 cm; contact twins (see fig. 2) are common and lamellar twins uncommon.

X-ray crystallography. X-ray powder diffraction data for both types are detailed in Table I. An 114.6 mm diameter asymmetric Straumanis camera was used for Leadhills macphersonite and Guinier-Hägg and Gandolfi cameras for the Argentolle mineral. The powder data in Table I are indexed on



FIG. 2. (1) Cleavage fragment of macphersonite, Leadhills. (2) Crystal drawing from a perfect crystal, and optic orientation of macphersonite. Argentolle Mine. (3) Pseudohexagonal contact twins. Argentolle Mine.

Leadhills'	*			Saint-Prix [†]					
d _{obs}	I _{est}	hkl	d _{calc}	$d_{\rm obs}$	I _{est}	hkl	d _{calc}		
11.45p	5	020	11.55	11.50	20	020	11.55		
	_			7.75	< 5	120	7.716		
	—		—	5.942	10	121	5.923		
5.74	2	040	5.775	5.790	5	040	5.775		
—		—		5.189	< 5	200	5.185		
—	_	_	—	5.044	< 5	140	5.050		
_	-	—		4.641	10	002	4.627		
4.50	10		4.522	4.533	25	012	4.535		
4.50	10	201	4.523	—	_	< <u>-</u>	 <		
4.43	10	$\begin{cases} 211 \\ 141 \end{cases}$	{ 4.441 { 4.432	4.430	10	$\left\{ \begin{array}{c} 211\\ 141 \end{array} \right.$	{ 4.441		
		(141	1 4.432	4.289	5	022	4.432 4.294		
	_			4.289	10	221	4.212		
		∫ 122	3.968			(122	§ 3.968		
3.94	10	{ 032	3.968	3.965	15	$\begin{cases} 122\\ 032 \end{cases}$	3.968		
_				3.910	25	231	3.900		
3.88	10	240	3.860						
_	_	_		3.849	5	060	3.850		
						042	3.613		
_	~	_	_	3.602	< 5	160	3.611		
3.57	2	241	3.561	3.569	5	241	3.561		
_	—	—	_	3.457	5	202	3.451		
3.41	5	ر 212	3.414 ر	3.415	10	$\begin{cases} 212 \\ 142 \end{cases}$	$\begin{cases} 3.414 \\ 3.412 \end{cases}$		
5.41	5	142	₹ 3.412	5.415	10				
			_	3.313	< 5	{ 320	{ 3.312		
	00	0.50	A A (A)			222	(3.307		
3.25	90	052	3.269	3.274	50	052	3.269		
3.22	100	251	3.232	3.234	100	251	3.232		
_	_	_		2.988	5	331	2.985		
				2.968	5	$\begin{cases} 340 \\ 242 \end{cases}$	2.966		
				2.908	5	062	2.959		
2.924	2	261	2.932	2.935	25	261	2.932		
2.874p	$2\overline{0}$	080	2.888	2.888	20	080	2.888		
		_		2.842	5	162	2.845		
2 7/0	6	(252	\$ 2.765			1 252	0.04		
2.760	5	$\begin{cases} 232 \\ 133 \end{cases}$	{ 2.765 2.759	2.758	10	133	{ 2.765 2.759		
2.644	20	j 351	£ 2.652	2.654	90	5 351	j 2.652		
2.044	20	203	2.652	2.034	90	203	2.650		
				2.598	30	$\begin{cases} 172 \\ 400 \end{cases}$	{ 2.601		
				2.570	50	1 400	2.593		
2.586	10	223	2.583	_	_				
				2.537	< 5	420	2.530		
				2 400		233	2.506		
			_	2.498	< 5	342	2.497		
		(101	(2440			(401 ∢421	2.496 2.440		
2.433	5	$\begin{cases} 421 \\ 281 \end{cases}$	${2.440 \\ 2.434}$	2.434	5	281	2.440		
		(272	{ 2.385			<pre>(281 (272)</pre>	<pre>{ 2.434 } 2.385</pre>		
2.368	5	{ 182	2.383	2.388	5	$\begin{cases} 272 \\ 182 \end{cases}$	2.383		
		(004	{ 2.313			(004	{ 2.313		
2.304p	80	371	2.313	2.310	30	371	2.313		
P	00	0.10.0	2.311	2.510	50	0.10.0	2.310		
		0.10.0	. 2.710			0.10.0	. 2.510		

 TABLE I. Comparison of observed and calculated powder diffraction data for macphersonite from Leadhills (Scotland) and Saint-Prix (France)

Leadhills	*			Saint-Prix†					
d _{obs}	I _{est}	hkl	d _{calc}	dobs	I _{est}	hkl	d _{calc}		
2.260	2	$\big\{ {\begin{array}{*{20}c} 024 \\ 402 \end{array}} \big.$	{ 2.268 2.262	2.265	20	{ 024 402 { 422	{ 2.268 2.262 2.219		
_	-	_	_	2.213	10	$ \left\{\begin{array}{c} 380\\ 124\\ 034\\ 282 \end{array}\right\} $	2.216 2.215		
2.209	2	333	2.205	_		、202)			
 2.171	10	432	2.170	2.182	30	263	2.183		
—		—	—	2.138	< 5	{ 044 343 { 204	{ 2.147 2.138 2.112		
	—	_		2.108	5	2.10.0 442 214 144	2.110 2.106 2.103 2.102		
_	_	_	_	2.060	< 5	$\left\{\begin{array}{c} 054\\ 0.10.2\\ 273\\ 183\\ 353\\ 292 \end{array}\right\}$	$\left\{\begin{array}{c} 2.068\\ 2.067\\ 2.066\\ 2.065\\ 2.060\end{array}\right.$		
2.056	10	2.10.1 (234	2.057 (2.037	_	—	(234	(2.037		
2.028	10	452 154 1.10.2	2.031 2.028 2.027	2.033	30	452 154 1.10.2	2.031 2.028 2.027		
1.996 1.950 1.936 1.820 1.738 1.723 1.653 1.653 1.632 1.612 1.538 1.458	2 5 5 20 10 30 2 2 5 30			Plus mor	e than 2	:0 weak lin	nes.		
1.282 1.271 1.235 1.154	10 5 5 5								

TABLE I (cont.)

p = lines showing preferred orientation.

* Data obtained from 114.6 mm diameter camera, Cu-Ka, Ni filter.

† Data obtained from Guinier-Hägg and Gandolfi; cameras, Cu-Kα, Ni filter.

the basis of a 10.37, b 23.10, and c 9.25 Å (cell volume 2215.8 Å³) obtained from a perfect Argentolle crystal using precession X-ray photographs. The Argentolle parameters demonstrate a relationship to those derived by Mrose and Christian (1969) for leadhillite and susannite. In the direction per-

pendicular to the perfect cleavage of all three polymorphs Argentolle b (23.10 Å) is twice leadhillite and susannite c values of 11.56 and 11.54 Å respectively. The Argentolle a parameter is exactly half leadhillite b. Precession and de Jong photographs on Argentolle material, for 0kl, h0l and hk0 show the following extinction conditions; hkl no condition, $0kl \ l = 2n$, $hk0 \ k = 2n$, $h0l \ h = 2n$, $h00 \ h = 2n$, $00l \ l = 2n$ and $0k0 \ k = 2n$. These conditions lead to the space group *Pcab*. Both leadhillite and susannite have Z = 8 formula units in the unit cell, as does macphersonite.

Chemistry and thermal analysis. Leadhills macphersonite analyses were undertaken using a Cambridge Instruments Geoscan V utilizing galena, copper and cadmium metals as standards. Results for ten separate cleavage fragments (single spot analysis per fragment) are given in Table II together with those for H_2O^+ and CO_2 determined on a 2.16 mg sample using thermogravimetric and evolved gas analysis. The empirical formula, calculated on the basis of 12 oxygen atoms from the average analysis, is (Pb_{4.08},Cu_{0.10}, $Cd_{0.07})_{\Sigma 4.25}S_{0.90}C_{2.18}O_{10.55}(OH)_{1.58}$, which leads to the ideal formula $Pb_4(SO_4)(CO_3)_2(OH)_2$. This formula is identical to that of leadhillite and susannite and both these minerals were analysed for comparative purposes using the same instrument and standards. All three polymorphs yielded a consistent and identical composition although no traces of Cu and Cd were detected in leadhillite or susannite. Empirically, sulphur is slightly lower and CO_2 slightly higher than the ideal values. Low sulphur cannot be attributed to wavelength shift due to valency differences between standard and unknown for the optimum 2θ value for S (due to SO_4) was ascertained before measurement on the unknown commenced.

Argentolle macphersonite conforms to the above chemistry for analysis on approximately 2 gm using gravimetric and thermogravimetric techniques yielded PbO 83.59, SO₃ 7.65, CO₂ 8.47, H₂O⁺ 1.93 (total 101.64%) which corresponds to Pb_{4.06}S_{1.03} $C_{2.08}O_{10.15}(OH)_{2.32}$.

Leadhills macphersonite CO_2 and H_2O^+

evolution profiles were very similar to those of leadhillite (Morgan, pers. comm.), i.e. simultaneous evolution of H_2O and approximately half the CO_2 at 300 °C in a well-defined, sharp reaction, followed by gradual release of the remaining CO_2 . The thermogravimetric trace (0.94 mg sample) revealed an initial sharp, weight loss between 250-320 °C followed by a slower loss up to 520 °C. This agrees with the evolved gas analysis results and is identical thermogravimetric behaviour to that of leadhillite.

Infrared spectra. Mrose and Christian (1969) in their abstract of unpublished data concluded that leadhillite and susannite could be differentiated by their infrared spectra; this is confirmed for leadhillite and susannite from the Leadhills-Wanlockhead area. Macphersonite produces an infrared spectrum which displays the basic similarities to the above minerals although additional, sharp absorptions are present which serve to distinguish it from the dimorphs. The Leadhills and Saint-Prix macphersonite samples produce identical spectra. Detailed infrared data for the three polymorphs will form the subject of a separate note (see p. 295).

Assemblage. The two assemblages in which macphersonite occurs are virtually identical. The Leadhills mineral is associated with leadhillite, susannite, cerussite, caledonite, yellow pyromorphite, and a milky-white mineral which produced an X-ray powder pattern akin to 'lead-hydroxyapatite' of Temple (1955). Argentolle associates are susannite, cerussite, pyromorphite, galena, quartz, and scotlandite.

The above common assemblages and abundance of leadhillite, combined with the macphersonite physical characteristics, strongly suggest the mineral may masquerade as leadhillite. It is surprising that the polymorph has not been described previously. Accordingly, leadhillite and macphersonite may mimic each other and a fairly rigorous

	1	2	3	4	5	6	7	8	9	10	Ave.
PbO SO ₃ CuO CdO CO ₂ H_2O^+	84.4 6.4 0.06 0.2	84.1 6.9 0.08 0.1	82.9 6.4 0.06 0.1	84.2 6.4 0.05 0.1	83.5 6.4 0.05 0.1	83.5 6.5 0.01 0.1	83.4 6.5 0.08 0.1	82.2 6.7 0.13 0.1	83.6 6.7 0.12 0.1	82.0 6.7 0.15 0.0	83.4 6.6 0.1 0.1 8.8 1.3
Total											100.3

TABLE II. Chemical composition of macphersonite from Leadhills Dod, Leadhills, Lanarkshire, Scotland

Anals. 1-10 electronprobe microanalyses (single spot analysis per grain) of macphersonite (Royal Scottish Museum specimen number 721.34).

Ave. Average of anals. 1-10 and including CO_2 and H_2O^+ determined by thermogravimetric and evolved gas analysis.

test will now be essential in order to distinguish the species with certainty. Apart from a larger 2V in macphersonite, either an X-ray powder photograph or an infrared spectrum will be required to identify either *leadhillite* or *macphersonite* in the future. The picture is complicated further for all three polymorphs may occur on the same specimen and leadhillite and susannite produce the same X-ray powder diffraction pattern.

Type material from Leadhills is lodged in the Royal Scottish Museum and registered under number 721.34, and Argentolle macphersonite is registered under number 435/80 within the Department of Mineralogy and Petrography, Museum of Natural History, Geneva. Acknowledgements. One of the authors (A.L.) is greatly indebted to Dr J. D. Russell of the Macaulay Institute for Soil Research for the infrared investigations. Thanks are also due to Dr D. J. Morgan of the Geochemical Division, Institute of Geological Sciences, for thermal studies. Preparation of fig. 2 by Mr P. J. Davidson and polished material for electronprobe-microanalysis by Mr R. J. Reekie, both of the Royal Scottish Museum, is gratefully acknowledged.

REFERENCES

Mrose, M. E., and Christian, R. P. (1969) Abstract, *Can. Mineral.* **10**, 141.

Temple, A. K. (1955) Trans. R. Soc. Edinb. 63, 85.

[Manuscript received 16 June 1983]

[Note added in proof. The full crystal structure of leadhillite and macphersonite has been recently determined from single crystal X-ray diffraction studies (Smith and Vickers, 1984, in preparation) and studies on susannite are in progress.]