Simpsonite and stibiotantalite from Benson pegmatite mine, Mtoko, Southern Rhodesia.

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Summary. A new occurrence of the tantalum minerals simpsonite and stibiotantalite is described from a lithium pegmatite at Mtoko, Southern Rhodesia. Optical, chemical, and powder X-ray data are given and the formula of simpsonite is considered; a new formula is proposed.

SIMPSONITE and stibiotantalite, two of the rarer tantalum minerals, are found in close association in one of a number of pegmatites that comprise the Benson mine at Mtoko, in the north-eastern part of Southern Rhodesia. Simpsonite has been previously described from four pegmatite areas only: Tabba Tabba in Western Australia (Bowley, 1939), Alto do Giz and Onça mines in north-eastern Brazil (Kerr and Holmes, 1945; Pough, 1945), Bikita in south-eastern Southern Rhodesia (Macgregor, 1946), and the North Kola Peninsula (Sosedko and Denisov, 1958). Stibiotantalite has been recorded from Greenbushes, Western Australia (Goyder, 1892), San Diego, California (Penfield and Ford, 1906), Topsham, Maine (Palache and Gonyer, 1940), Varuträsk, Sweden (Ödman, 1941), Alto do Giz, Brazil (Pough, 1945), Kazakhstan (Barsanov and Ginzburg, 1946), Alto Ligonha, Mozambique (Bandy, 1951), North Kola Peninsula (Sosedko, 1958), and Gunnison, Colorado (Heinrich, 1960).

Several of the forty pegmatites that occur in the Benson mine have stages containing a suite of minerals typical of other important pegmatite fields in Southern Rhodesia; this includes amblygonite, spodumene, lepidolite, topaz, pollucite, multicoloured beryl, apatite, zircon, cassiterite, bismutite, columbite–tantalite, manganotantalite, and microlite.

In general very little is known about the simpsonite- and stibiotantalite-bearing pegmatites, their mineralogy, and their internal structure. According to Bowley (1939), the original simpsonite from Tabba Tabba 'occurs as flat, tabular crystals in a quartz-biotite pegmatite between a quartz blow and a feldspar pegmatite'. The simpsonite from north-eastern Brazil is associated with a number of tantalum minerals, e.g. tantalite, manganotantalite, microlite, and stibiotantalite. From the description by Pough (1945), simpsonite and stibiotantalite were not seen in place, but the pegmatites appear to be typically lithium-bearing, with late stage mineralization and replacement phenomena. The first find of simpsonite in Southern Rhodesia was from one of the Bikita pegmatites; here the simpsonite is partly altered to microlite and is closely associated with another mineral, which, judging from the description by Macgregor, may well have been stibiotantalite.

The simpsonite from the north Kola Peninsula is associated with cleavelandite, rose-coloured mica, caesium beryl, spodumene, montebrasite, pollucite, petalite, eucryptite, rose and greenish and colourless tourmaline, manganotantalite, microlite, and stibiotantalite. The Kola simpsonite is sometimes rimmed with a dark brown halo consisting of manganotantalite with scattered microlite crystals.

Stibiotantalite was originally found in the cassiterite concentrates from the Greenbushes tinfield south of Perth in Western Australia, and was known as 'pale' or 'resin' tin by the prospectors (Miles *et al.*, 1945). Some was also recovered, together with tantalite and cassiterite, from the Enterprise lode, a greisenized pegmatite. All the samples from this lode, and many of those from the alluvium, show stibiotantalite as a replacement product of earlier tantalite. The replacement usually starts at the margins and proceeds inwards to various degrees. Polished sections of the tantalite–stibiotantalite from Greenbushes, examined by Edwards (Miles *et al.*, 1945) show that, although the tantalite crystallized first, there has been a subsequent crystallization of stibiotantalite with little marked replacement.

Stibiotantalite in fine crystals has been recovered from Mesa Grande, San Diego County, California, where it is associated with pink beryl, lepidolite, pink tourmaline, and some cassiterite. At the Varuträsk lithium pegmatite in northern Sweden, stibiotantalite has been partly replaced by an antimonian microlite and native antimony.

Stibiotantalite recorded from the Brown Derby pegmatite in Gunnison County, Colorado, is associated with pink tourmaline, lepidolite, columbite, euxenite, and microlite.

Excellent stibiotantalite crystals have been recovered from Mina Muiane and other mines in the Alto Ligonha region of Mozambique. Fragments of stibiotantalite have been observed by the present writers in the heavy mineral concentrates at the Muiane mine, together with tantalite, manganotantalite, microlite, and native bismuth.

The simpsonite–stibiotantalite-bearing pegmatite described in the present paper is known as Benson no. 3. This has a dyke-like form, is 900 ft long, 25–35 ft wide and is exposed in open cuttings to a depth of some 40 ft. The pegmatite is almost vertical and has the following zonal pattern:

Hanging wall zone	quartz–muscovite–albite	5 ft
Intermediate zone	quartz–lepidolite–albite	5–8 ft
Core	quartz	5 ft
	spodumene and amblygonite in core footwall	0–1 ft
Late stages	cleavelandite with purple and colourless fine-grained lithium mica	0–3 ft
Intermediate zone	quartz–lepidolite–albite	1–3 ft
Footwall zone	quartz–muscovite–albite	2–3 ft

The country rocks are metavolcanic amphibolites that have undergone metasomatic alteration to holmquistite- and biotite-bearing rocks (von Knorring and Hornung, 1961*a*).

Accessory minerals show a marked zonal distribution, with spessartine garnet occurring in both the hanging and footwall zones, whilst cassiterite is limited to the footwall. Tantalite is found in the intermediate zones both above and below the core, though dominantly below. Microlite and manganotantalite are associated with cleavelandite and purple fine-grained lepidolite, both situated below the quartz core. Simpsonite and stibiotantalite are almost entirely confined to bands of a very fine-grained, massive colourless mica containing small amounts of spodumene, quartz, topaz, and albite. The spodumene associated with the quartz core contains no accessory minerals except occasional white beryl crystals. Pink-coloured beryl occurs in the intermediate zone.

The sequence of tantalum minerals at the Benson mines appears to be as follows:

> columbite–tantalite; manganotantalite; simpsonite and stibiotantalite; microlite.

This shows a general agreement with Cameron *et al.* (1949) and Ginzburg (1956). In some lithium pegmatites the last four minerals are intimately associated and in particular it has been observed that manganotantalite and microlite, on the one hand, and simpsonite and stibiotantalite, on the other, are frequently intergrown. In the north Kola occurrence all four minerals are found, sometimes intergrown.

Simpsonite and stibiotantalite were observed in the heavy mineral concentrates when Benson 3 was being mined. The proportion of these minerals, in comparison with microlite and manganotantalite, was very small indeed. The dark grey stibiotantalite is more easily recognized in hand specimen than simpsonite and was observed in places in the grey, fine-grained mica rock, in irregular crystals up to 15 mm long and 10 mm across. During examination of the mica specimens in ultra-violet light, fluorescent simpsonite grains were also noticed, and it was possible to separate a sufficient amount of both minerals for chemical analysis.

The simpsonite usually occurs in minute rounded grains, but occasionally larger grains and aggregates up to 10 mm across have been found. The general appearance of isolated simpsonite is that of quartz, though possibly whiter in colour and with a greasier lustre. The cleavage is very poorly developed. A few small crystals with hexagonal outlines and striated prism faces have been recovered. In thin sections the simpsonite is seen in irregular grains rather like anhedral garnet, with enclosed grains of mica (fig. 1). A marked concentration of small spodumene crystals is often observed around the simpsonite. No replacement by other tantalum minerals has been observed.

Simpsonite is noticeably harder than quartz. It has a measured specific gravity of 6·68–6·72 and a rather strong bluish fluorescence similar to that of scheelite. Chemical analysis and powder data are given in tables I and II.

Stibiotantalite occurs in rounded grains, sometimes closely associated with simpsonite, but mostly completely isolated. Larger stibiotantalite grains almost invariably have tiny simpsonite inclusions and marginal microlite replacing the stibiotantalite (figs. 2 and 3). The cell edge of this microlite is 10·42 Å, corresponding to that of the antimonian microlite described by Ödman (1941). The colour of isolated grains of stibiotantalite varies from clear transparent honey-yellow to greenish yellow, grey, and vitreous black. The darker grains have a sub-metallic lustre. The mineral has a pronounced cleavage and a hardness of about 5.

One of the intriguing features of simpsonite is the uncertainty of its chemical formula. The original simpsonite was partly replaced by other tantalum minerals and after recomputation Bowley (1930) suggested the formula $4Ta_2O_5.5Al_2O_3.CaO.2H_2O$. Kerr and Holmes (1945) proposed the formula $2Ta_2O_5.3Al_2O_3$ on the basis of their analyses of Brazilian and Bikita simpsonites. Macgregor (1946) meanwhile arrived at the formula $3Ta_2O_5.4Al_2O_3$, based on analysis of simpsonite from Bikita and also by recalculating the original simpsonite analyses from Tabba Tabba. Sosedko and Denisov (1958), in their work on simpsonite from north Kola Peninsula, agreed with the formula proposed by Macgregor. A chemical analysis of the Mtoko simpsonite is given in table I.

The material for this analysis was prepared by using heavy liquids (acetylene tetrabromide and Clerici solution) followed by repeated

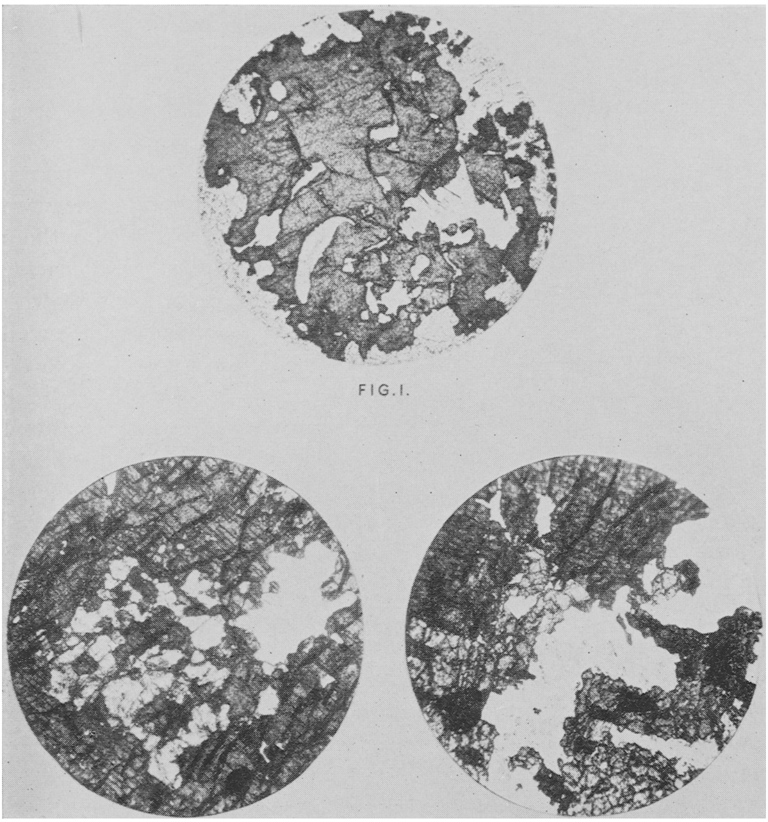

FIG. 2. FIG. 3.

FIGS 1–3: FIG. 1 (*top*). An aggregate of simpsonite in fine-grained lithium mica. ×35. FIG. 2 (*bottom left*). Simpsonite grains in stibiotantalite. ×35. FIG. 3 (*bottom right*). Stibiotantalite (dark grey) replaced by microlite (grey). Most of the white mineral is lithium mica. ×35.

hand-picking in white and ultra-violet light. The major contamination in the final product was a colourless lithium mica; there were also traces of albite. The chemical composition of the mica was known (Hornung, 1961) and the simpsonite analysis could therefore be recalculated.

The present analysis indicates that an appreciable amount of water and some fluorine are present even after correcting the analysis for mica. A thorough examination of the mineral revealed no alteration products and repeated determinations of the water showed consistent

TABLE I. Chemical composition and physical properties of simpsonite and stibiotantalite from Benson mine, Mtoko, Southern Rhodesia.

	1.	1′	1*a*.	2.	2*a*.
Ta_2O_5	70·48	71·71	Ta 2·78	Ta_2O_5 56·98	Ta 0·95
Nb_2O_5	2·10	2·14	Nb 0·14	Nb_2O_5 1·94	Nb 0·05
SiO_2	0·85	—	Sn 0·06	Sb_2O_3 38·97	Sb 0·98
TiO_2	tr.	—	Al 4·00	Bi_2O_3 0·91	Bi 0·02
SnO_2	1·00	1·02	OH 1·12	Fe_2O_3 } Mn_2O_3 } 0·20	O 4·00
Al_2O_3	23·89	23·85	F 0·08		
Fe_2O_3	tr.	—	O 12·80		
CaO	tr.	—		99·00	
Li_2O	0·04	—			
Na_2O	0·07				
K_2O	0·16	—	simpsonite:	stibiotantalite:	
H_2O+	1·20	1·18	Sp. gr. 6·70	Sp. gr. 7·30	
H_2O-	0·06	—	ω 2·036	Refr. ind. $>$ 2·11	
F	0·25	0·17	ϵ 2·004	$2V_\gamma \sim$ 70°	
	100·10	100·07			
Less O for F	0·10	0·07			
	100·00	100·00			

1. Simpsonite from Benson mine, Mtoko, Southern Rhodesia. Anal.: O. von Knorring.

1′. Anal. 1, corrected for mica impurities and recalculated to 100 %.

1*a*. Atomic ratios of simpsonite to 14 (O,OH,F).

2. Stibiotantalite (ass. with simpsonite from Benson mine). Anal.: O. von Knorring.

2*a*. Atomic ratios of stibiotantalite to 4 oxygen.

results. Unfortunately, there is insufficient data on the water content in simpsonites from other localitites for comparison. A relatively large amount of tin was also observed. Previously, tin has been attributed to cassiterite present as impurity; in the Benson material the tin is believed to replace tantalum. This may be a characteristic feature of simpsonite from pegmatites where tin is available, e.g. Tabba Tabba and Bikita. On the other hand, no tin was observed in the simpsonite from north Kola (Sosedko and Denisov, 1958).

The chemical analysis and powder data of stibiotantalite compare favourably with available analyses of this mineral and conform with the established formula $SbTaO_4$. Small amounts of niobium and bismuth take the place of tantalum and antimony respectively (tables I and II).

TABLE II. X-ray powder data for simpsonite (1) and stibiotantalite (2) from Benson mine, Mtoko, Southern Rhodesia. Fe-$K\alpha$ radiation (simpsonite), Cu-$K\alpha$ radiation (stibiotantalite), camera diameter 9 cm.

1.				2.					
d, Å.	*I.*	*d*, Å.	*I.*	*d*, Å.	*I.*	*d*, Å.	*I.*	*d*, Å.	*I.*
6·47	m	1·361	w	5·92	w	1·519	w	1·030	w
4·53	m	1·315	w	4·52	w	1·495	w	1·017	w
3·69	s	1·277	w	3·55	m	1·454	w	0·993	w
3·20	m	1·230	w	3·13	vs	1·398	w	0·983	w
2·880	m	1·209	w	2·96	m	1·350	w	0·965	w
2·615	m	1·188	w	2·678	m	1·305	w	0·956	w
2·427	w	1·169	w	2·452	w	1·267	w	0·944	w
2·135	s	1·147	w	2·259	w	1·248	w	0·936	w
1·920	w	1·129	w	2·040	w	1·231	w	0·927	w
1·845	w	1·115	w	1·987	w	1·208	w	0·914	w
1·773	s	1·088	w	1·817	m	1·182	w	0·908	w
1·651	vw	1·081	w	1·735	m	1·153	w	0·896	w
1·598	vw	1·066	m	1·715	m	1·136	w	0·854	w
1·550	m	1·051	m	1·665	vw	1·124	w	0·845	w
1·503	m			1·626	m	1·081	w	0·834	w
1·467	s			1·589	w	1·075	w		
1·394	w			1·561	w	1·044	w		

From their X-ray studies Kerr and Holmes (1945) have deduced for the unit cell of simpsonite *a* 7·3766 Å, *c* 4·5141 Å. Sosedko and Denisov (1958) obtained very similar results for their simpsonite from north Kola. The cell volume of simpsonite, which is hexagonal, is accordingly 123 Å^3, and by taking this value together with the oxygen data obtained from the chemical analysis and the measured density, one arrives at 14 (O,OH) atoms in the unit cell and an ideal formula $Ta_3Al_4O_{13}OH$. The calculated unit-cell weight of simpsonite (based on the cell volume and a specific gravity of 6·70) is 858, comparing favourably with the analytical result of 855. Neither of the formulae previously suggested support a unit-cell weight that agrees with the observed value, whilst the formula $Ta_3Al_4O_{13}OH$ (formula weight 875) shows good agreement and at the same time accommodates the water clearly present in the Benson simpsonite.

Geochemical considerations.

In a previous paper von Knorring and Hornung (1961*b*) have discussed the enrichment and separation of coherent pairs of elements during successive stages of pegmatite crystallization. Available analytical data for various tantalum minerals from the Benson mine (Hornung, 1961) show the following enrichment of tantalum as compared with niobium in the later-formed minerals:

		Ta_2O_5/Nb_2O_5
Columbite–tantalite	Benson 4	1·3
Manganotantalite	Benson 4	4·0
Microlite (yellow)	Benson 2	15
Stibiotantalite	Benson 3	29
Simpsonite	Benson 3	33

The amount of tin in the Benson tantalum minerals varies in the following manner:

		% SnO_2.
Columbite–tantalite	Benson 4	tr.
Manganotantalite	Benson 4	0·20
Simpsonite	Benson 3	1·02
Microlite (yellow)	Benson 2	1·50

According to Solodov (1959) there is a marked increase of tin in the later phases of pegmatite crystallization; this is in close agreement with the present data on the distribution of tin in the tantalum minerals.

The Benson 3 pegmatite, where simpsonite and stibiotantalite are found, appears to be a very late differentiate and characteristically low in potash feldspar, a common feature of many rare-metal pegmatites. Potassium is accommodated largely in the lithium-bearing micas whilst an appreciable amount has migrated outward to form biotite in the halo of metasomatized country-rock. Sodium is fixed largely as albite. This migration may well have been facilitated by contemporaneous tectonic and chemical processes. Lithium, sodium, fluorine, water, and other volatiles are considered to have been the major operative participants during the formation of this pegmatite.

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