

*Pyrosmalite in the Broken Hill lode, New South Wales.*<sup>1</sup>

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*Summary.* Pyrosmalite is a widespread, though very limited, constituent of the Broken Hill lode, and is formed by the hydration and chlorination of the manganese-iron silicates. It is altered in contact with galena and eventually disintegrates into a mat of very fine fibres. The pyrosmalite varies substantially in ratio of Mn/Fe, and two specimens from the rare vughs and fractures gave on analysis ratios of 1.96 and 0.71. The refractive indices decrease slightly with increasing amounts of manganese and water, with a simultaneous small increase in the unit-cell dimensions. The name pyrosmalite is retained for the mineral species in accord with its earlier usage and its recent limitation to the iron-rich variety of an isomorphous series is not endorsed.

THE rare manganese-iron silicate pyrosmalite was first found in the iron mines of Nordmark and Dannemora, Sweden. Published analyses of material from these localities, summarized by Zambonini (1901), reveal a variation in the proportions of manganese and iron; both, however, are major constituents. Frondel and Bauer (1953) have described a manganiferous pyrosmalite from Sterling Hill, New Jersey, and show the clear distinction by X-ray powder pattern of pyrosmalite from the similar minerals friedelite and schallerite.

Pyrosmalite was first recognized at Broken Hill by M. A. Mawby in 1943 (personal communication) in specimens from the No. 12 level Zinc Corporation mine, No. 10 level Broken Hill South mine, and No. 17 level North Broken Hill mine. The identification was confirmed by partial analyses of the samples from the Zinc Corporation and North Broken Hill mines, carried out in the Zinc Corporation assay office. The list of Broken Hill minerals in Woodward (1952), prepared by M. A. Mawby, is the first record of this mineral in the Broken Hill lode.

Pyrosmalite was independently recognized during a petrological study of drill cores from the No. 11 level Junction Mine, North Broken Hill Ltd. Since then it has been found in thin section as a frequent but minor constituent of the manganese silicate bodies within the Broken Hill lode. It occurs in thin section as colourless to very pale brown ragged plates

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up to 1.5 mm. across which occasionally show a roughly hexagonal outline in near basal sections; a perfect basal cleavage is perpendicular to the optic axis in the uniaxial negative interference figure. The refractive indices of the mineral were determined in sodium light on the analysed samples and on particles excavated from polished sections. The extreme values obtained were:  $\omega$  1.664 and 1.682, both  $\pm 0.002$ ,  $\epsilon$   $1.634 \pm 0.002$  and  $1.650 \pm 0.003$ .

Crystals of pyrosmalite occur in a vugh bordering calcite within a pocket of blende that bulged from an inch-thick seam of blende through bustamite and manganhedenbergite, in the east leg of the ore body at the 1370-foot level, Broken Hill South mine. The crystals are well formed hexagonal plates up to 3 cm. in width and up to 1.5 cm. thick; many are less than  $\frac{1}{2}$  cm. across. The pyrosmalite is coated by quartz crystals, which partially line the vugh; both pyrosmalite and quartz are partially covered with a thin black film comparable in composition with sturtite (Hodge-Smith, 1930), which gives an X-ray diffraction pattern consisting of three diffuse lines at spacings of 4.37, 2.62, and 1.56 Å. The crystals are pale brown with a pearly lustre on the basal pinacoids, in contrast to the prism faces which vary from rose-pink to darker reddish-brown, and have a vitreous lustre.

Mawby's specimen of pyrosmalite, from about 75 feet above the 1700-foot level, North Broken Hill mine, is part of a large crystal 1 cm. thick along a joint or fracture plane through rhodonite and garnet. It is pale brown in colour and has the basal cleavage parallel to the fracture and a prismatic parting perpendicular to this cleavage.

*Crystallography.* The tabular crystals in the vugh from the Broken Hill South mine consist of the basal pinacoid in combination with the hexagonal prism (10 $\bar{1}$ 0); an intergrowth of such crystals is shown in fig. 1. Rhombohedral faces are rare, as narrow facets truncating the junction of prism and pinacoid. These crystals are of similar morphology to some from the Swedish occurrences (Flink, 1917). The blocky texture of the prism and pinacoid, and the re-entrant angles on some prism faces, reveal that most larger crystals are a sub-parallel intergrowth of smaller crystals. The internal anastomosing network seen within the crystals in transmitted light is visible in fig. 1.

An X-ray Laue photograph taken with the beam incident along the *c*-axis reveals that the crystals are trigonal, with a Laue symmetry of  $\bar{3}m$ ; the symmetry planes are at 30° to the prism faces. A weak single-crystal oscillation photograph about the intersection of prism and pinacoid determines the repetition period of this direction as 13.3 Å.

The X-ray powder pattern, table I, indexes very well for a unit cell of dimensions  $a$  13.36,  $c$  7.16 Å., corresponding to that found for pyrosmalite by Frondel and Bauer (1953) and slightly below the dimensions given by Gossner and Mussgnug (1931).

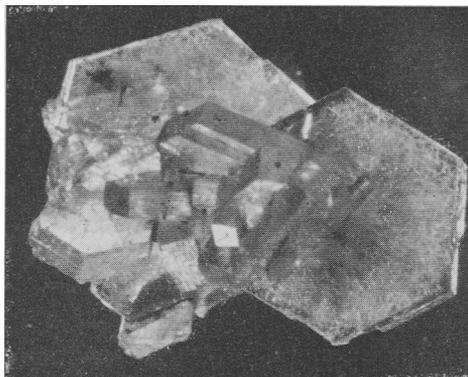


FIG. 1. Cluster of crystals of pyrosmalite from vugh, 1370-foot level, section J2i, Broken Hill South mine. Transmitted dark ground illumination combined with oblique incident illumination.  $\times 8$ .

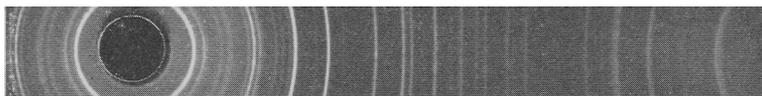


FIG. 2. X-ray powder pattern of pyrosmalite, about 75 feet above 1700-foot level, No. 11 section, North Broken Hill mine. Fe radiation, Mn filter.

† The possible space-groups are  $P321$ ,  $P3m1$ , or  $P\bar{3}m1$ , in agreement with that found by Gossner and Mussgnug assuming holohedral symmetry. The indexing given in table I reveals more possible diffracting planes than are given by Frondel and Bauer. It was found that the five sets of lattice planes of spacing greater than 2.47 Å. that do not give visible diffractions on the powder pattern are all of type  $3.k.\overline{k+3}.l$  with  $k$  positive.

The X-ray powder patterns obtained from Broken Hill pyrosmalites of different composition are identical except for slight variation in position of some of the back-diffraction lines. One pattern is listed in table I. Relative to other lines the basal  $000l$  diffractions have only a half to

TABLE I. X-ray powder pattern of pyrosmalite, North Broken Hill mine, New South Wales ( $a = 13.36$ ,  $c = 7.16$  Å., Fe radiation, Mn filter).

As the symmetry is trigonal many of the indices refer to more than one form.

<i>I.</i>	<i>d</i> (meas.).	<i>hkl.</i>	<i>d</i> (calc.).	<i>I.</i>	(meas.).	<i>hkl.</i>	<i>d</i> (calc.).
2	11.5 Å.	1010	11.57 Å.			{ 3363	1.628 Å.
8	7.16	0001	7.16	4	1.629 Å.	{ 4481	1.626
$\frac{1}{2}$	6.65	1120	6.68			{ 5381	1.610
$\frac{3}{2}$	6.06	1011	6.09	tr.	1.608	{ 7071	1.610
$\frac{1}{2}$	5.74	2020	5.78			{ 6172	1.583
2	4.87	1121	4.88	tr.	1.588	{ 5163	1.567
1	4.50	2021	4.50			{ 6281	1.566
2	4.35	2130	4.37	tr.	1.566	{ 3144	1.563
2	3.73	2131	3.73			{ 4044	1.522
5	3.58	0002	3.58	5	1.521	{ 6282	1.464
		{ 1012	3.42			{ 5273	1.463
4	3.41	{ 3031	3.40	$\frac{1}{2}$ br.	1.459	{ 4154	1.460
$\frac{1}{2}$	3.34	2240	3.34			{ 6390	1.458
tr.	3.15	1122	3.15			{ 0005	1.432
		{ 2022	3.04	tr.	1.427	{ 6391	1.428
2	3.03	{ 2241	3.03			{ 1015	1.421
1	2.89	4040	2.892			{ 6173	1.419
1	2.77	2132	2.770	4	1.419	{ 8081	1.417
10	2.69	4041	2.682			{ 5054	1.416
tr.	2.61	3032	2.624			{ 1125	1.400
$\frac{1}{2}$	2.52	4150	2.525	tr.	1.397	{ 3364	1.395
		{ 3142	2.390			{ 5492	1.369
1	2.38	{ 0003	2.387	2	1.370	{ 4483	1.368
		{ 4151	2.381			{ 3035	1.342
		{ 4042	2.250	2	1.342	{ 8082	1.340
7	2.25	{ 1123	2.248			{ 7183	1.289
		{ 2023	2.206			{ 5274	1.287
tr.	2.20	{ 5051	2.202	tr.	1.287	{ 9090	1.286
		{ 4260	2.187			{ 4045	1.283
		{ 3252	2.132	3	1.282	{ 7.3.10.1	1.281
tr.	2.13	{ 3361	2.126			{ 8192	1.267
		{ 2133	2.095	$\frac{1}{2}$	1.268	{ 9091	1.265
$\frac{1}{2}$	2.09	{ 4261	2.091			{ 3255	1.260
		{ 5160	2.078	tr.	1.259	{ 5493	1.259
tr.	2.02	3033	2.030			{ 6174	1.257
tr.	1.930	6060	1.928			{ 4155	1.245
tr.	1.912	3143	1.915			{ 6393	1.244
tr.	1.892	3362	1.891	tr.	1.245	{ 6.4.10.2	1.244
		{ 4043	1.841			{ 8.2.10.1	1.243
5	1.840	{ 4371	1.838	3	1.238	{ 8083	1.236
		{ 5162	1.797	tr.	1.226	{ 7.3.10.2	1.223
tr.	1.797	{ 5271	1.794			{ 5055	1.218
		{ 1014	1.769			{ 7293	1.216
$\frac{1}{2}$	1.766	{ 6170	1.764	tr.	1.216	{ 5384	1.214
		{ 4153	1.734			{ 7074	1.214
tr.	1.734	{ 1124	1.729			{ 0006	1.193
5	1.672	4480	1.670	tr.	1.192	{ 8.2.10.2	1.191
<i>I.</i>	<i>d</i> (meas.).	<i>I.</i>	<i>d</i> (meas.).	<i>I.</i>	<i>d</i> (meas.).	<i>I.</i>	<i>d</i> (meas.).
$\frac{1}{2}$	1.178	tr.	1.094	$\frac{1}{2}$	1.0180		
tr.	1.152	2	1.0873	$\frac{1}{2}$	1.0100		
$\frac{1}{2}$	1.142	4	1.0820	tr.	1.0055		
4	1.126	tr.	1.060	5	0.9953		
tr.	1.118	4	1.0472				
1	1.102	$\frac{1}{2}$	1.0298				

two-thirds the intensity given by Frondel and Bauer (1953). As preferred orientation was avoided by using approximately equidimensional fragments and ball-shaped mounts, the patterns suggest that the intensities of basal diffractions given by Frondel and Bauer are enhanced by pre-

ferred orientation of platy cleavage fragments. The weakness or absence of some high-angle diffractions in the data of Frondel and Bauer as compared with table I may result from a greater amount of grinding (cf. McAndrew, 1952).

*Chemical composition.* An analysis of fragments of a few small crystals of pyrosmalite from the vugh at the 1370-foot level, Broken Hill South mine, carefully freed from the coating of quartz and sturtite, is given in table II, together with an analysis of pyrosmalite from the specimen of M. A. Mawby from the North Broken Hill mine.

The low total of both analyses is noticeable. The atomic ratio of Si to  $R^{2+}$  is slightly below the integral ratio of 12:16, suggesting either that the figures for silica may be slightly low, or that an element that substitutes for silicon has not been determined. The latter is probable, as As replaces Si in the related schallerite, and although not sought in the analysis, As was subsequently found to be present in the North mine specimen by a qualitative test.

TABLE II. Chemical analyses and unit-cell contents of Pyrosmalite, Broken Hill, New South Wales.

	(1)	(2)		(1a)	(2a)
FeO . . . . .	17.05	30.58	Fe . . . . .	5.02	8.99
MnO . . . . .	32.96	21.51	Mn . . . . .	9.83	6.40
ZnO . . . . .	1.92	1.08	Zn . . . . .	0.50	0.28
CaO . . . . .	1.29	1.21	Ca . . . . .	0.49	0.46
MgO . . . . .	tr.	nil			
SiO <sub>2</sub> . . . . .	33.42	34.18	Si . . . . .	11.76	12.01
Al <sub>2</sub> O <sub>3</sub> . . . . .	nil	nil			
Fe <sub>2</sub> O <sub>3</sub> . . . . .	nil	nil	O . . . . .	29.52	30.05
Cl . . . . .	3.78	3.79			
H <sub>2</sub> O + . . . . .	9.36	7.65	OH . . . . .	17.41	17.93
H <sub>2</sub> O - . . . . .	0.07	0.04	Cl . . . . .	2.26	2.26
	99.85	100.04	H <sub>2</sub> O . . . . .	2.28	0.00
Less O for Cl . . . . .	0.85	0.85			
			Σ (O, OH, Cl, H <sub>2</sub> O) .	51.47	50.24
Total . . . . .	99.00	99.19			
D (meas.) . . . . .	3.11	3.14			

(1) Pyrosmalite, crystals from vugh, 1370-foot level, section J2i, Broken Hill South mine, Broken Hill. Analyst, G. C. Carlos.

(2) Pyrosmalite, about 75 feet above 1700-foot level, no. 11 section North Broken Hill mine, Broken Hill. Analyst, G. C. Carlos.

(1a, 2a) Empirical unit-cell contents.

The unit-cell contents of pyrosmalite from the North Broken Hill mine correspond closely to  $2[(\text{Fe}, \text{Mn}, \text{Ca}, \text{Zn})_8 \text{Si}_6 \text{O}_{15} (\text{OH}, \text{Cl})_{10}]$ , putting determined H<sub>2</sub>O as OH, in agreement with the cell contents found by Gossner and Mussnug (1931) and by Frondel and Bauer (1953) as re-computed by Hey (1956). The crystals from the Broken Hill South

mine contain appreciably more water. To obtain their cell contents in a form suitable for comparison, the determined O and H<sub>2</sub>O is split into O, OH, and H<sub>2</sub>O on the assumption that silica is slightly low. The oxygen content is taken as 30.00 less the oxygen equivalent of the deficiency of Si below 12.00, thereby also fixing OH and H<sub>2</sub>O. Computed in this manner, the analysis of column 1, table II, gives a unit-cell content close to 2[(Mn,Fe,Zn,Ca)<sub>8</sub>Si<sub>6</sub>O<sub>15</sub>(OH,Cl)<sub>10</sub>·1.1H<sub>2</sub>O].

The proportion of Mn:Fe varies from 2:1 in analysis 1 to 2:3 in analysis 2, and it is unlikely that these ratios indicate the limits of variation in pyrosmalite at Broken Hill; the crystals are intermediate in ratio of Mn:Fe between the Swedish and Sterling Hill pyrosmalites. Their water content is appreciably higher than the highest value of 8.55 % recorded for pyrosmalite from Sweden (Gorgeu, 1884 *b*), but is comparable with the 9.60 % of water reported in the related mineral friedelite from the Hautes Pyrénées by Gorgeu (1884*a*). The water content of pyrosmalite is evidently variable. The anion content of the unit cell is close to 2[O<sub>15</sub>(OH,Cl)<sub>10</sub>] and 2[O<sub>15</sub>(OH,Cl)<sub>10</sub>·1.1H<sub>2</sub>O] in the two analysed specimens from Broken Hill. Although the Sterling Hill pyrosmalite has a water content intermediate between that of these two specimens, the anion unit-cell content is close to 2[(O,OH)<sub>15</sub>(OH,Cl)<sub>10</sub>] (Hey, 1956), requiring some substitution of OH for O but no excess of total anions above fifty, in contrast to the crystals from the Broken Hill South mine.

*Variation.* The variation in chemical composition of pyrosmalite is accompanied by a small although distinct variation in other properties. These are summarized in table III and compared with the Sterling Hill pyrosmalite, Frondel and Bauer (1953).

TABLE III. Variation in properties of Pyrosmalite.

Composition.	$\omega$ .	$\epsilon$ .	Spacing of high-angle lines in X-ray pattern, Å.			Locality.
			1-0835	0-9938		
Not analysed . . . . .	1-682	1-650	1-0835	0-9938		Broken Hill
(Fe <sub>0.55</sub> Mn <sub>0.46</sub> Zn <sub>0.02</sub> Ca <sub>0.03</sub> ) <sub>8</sub> Si <sub>6</sub> O <sub>15</sub> (OH,Cl) <sub>10</sub>	1-670	1-639	1-0873	1-0820	0-9953	Broken Hill
(Mn <sub>0.62</sub> Fe <sub>0.32</sub> Zn <sub>0.03</sub> Ca <sub>0.03</sub> ) <sub>8</sub> Si <sub>6</sub> O <sub>15</sub> (OH,Cl) <sub>10</sub> ·1 H <sub>2</sub> O	1-664	1-634	1-0890	1-0862	0-9983	Broken Hill
(Mn <sub>0.72</sub> Fe <sub>0.23</sub> Mg <sub>0.02</sub> Zn <sub>0.03</sub> ) <sub>8</sub> Si <sub>6</sub> O <sub>15</sub> (OH,Cl) <sub>10</sub>	1-669	1-631	—	—	—	Sterling Hill

It can be observed, table III, that the refractive index decreases slightly with increasing amount of manganese and of water, with a simultaneous increase in the cell dimensions. The highest refractive index and lowest cell dimensions occur in pyrosmalite that is associated with dannemorite and pyroxmangite, and presumably rich in iron.

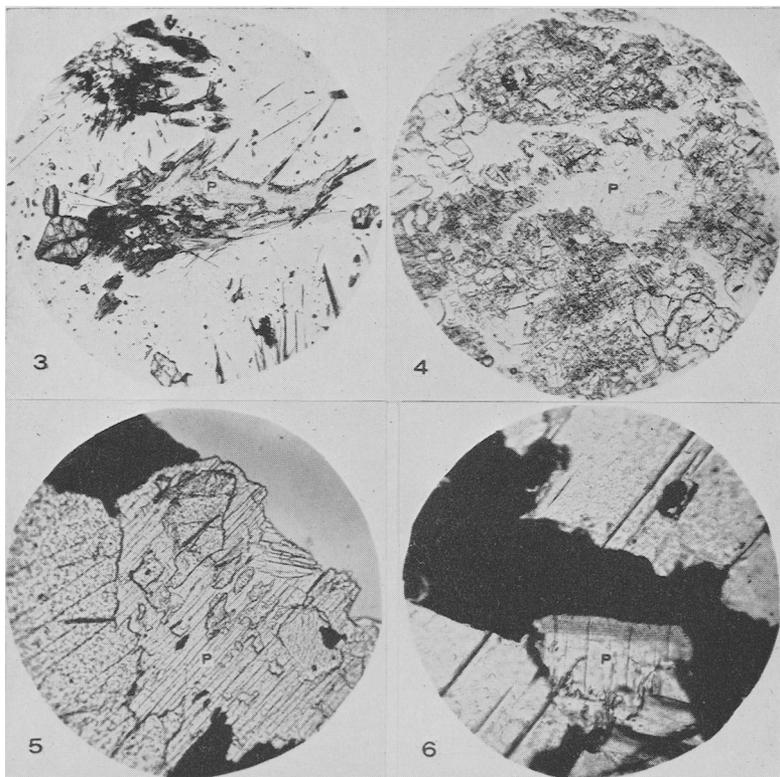
The increase in cell dimensions is best revealed by the position of the

strong line in the X-ray powder pattern at a diffraction angle of about  $150^\circ$ , which varies in spacing from 0.9938 to 0.9983 Å. in patterns obtained from narrow, weakly absorbing specimens in which systematic displacement of line position is a minimum (Straumanis, 1949). The variation in separation of the doublet with spacings of approximately 1.087 and 1.082 Å. reveals a variation in the axial ratio,  $c/a$ .

The ratio of Mn:Fe is 2:1 in the crystals from the Broken Hill South mine, intermediate between the highest value of 1.05:1 recorded for Swedish material (Zamboni, 1901) and the ratio 3:2:1 in pyrosmalite from Sterling Hill (Fronzel and Bauer, 1953). Fronzel and Bauer describe the manganese-rich mineral from Sterling Hill as mangan-pyrosmalite, and are followed by Hutton (1956) for a Broken Hill specimen. In doing so, they relegate the name pyrosmalite from that for a species as found with approximately equal amounts of Fe and Mn to that of a variety with  $Fe > Mn$ ; this departure from the earlier usage is undesirable. The name pyrosmalite is here retained for all members of the isomorphous series. The small differences in optical properties with variation in the Mn:Fe ratio render varietal distinctions inapplicable to petrological studies in thin section, where the mineral is identified without the possibility or necessity of chemical analysis.

It may be noted that the name manganpyrosmalite was first used by M. Bauer (1885) in a review of Gorgeu's paper (1884a). Bauer suggested that friedelite might be 'ein FeOfreier Manganpyrosmalith', using the name for a hypothetical end-member.

*Occurrence.* Specimens of pyrosmalite from vughs and fractures, such as have been analysed, are rare in Broken Hill. Nevertheless, pyrosmalite is a widespread, though very minor, mineral in the large bodies of the manganese silicates occurring in the Broken Hill lode. It is commonly found with galena or in the neighbourhood of galena on the margins or grain boundaries of rhodonite, knebelite, bustamite, and manganhedenbergite; it appears sometimes in the interstices between garnets or in veinlets in a garnet quartzite. In many instances it is clearly derived by replacement of the anhydrous manganese silicates; it occurs rather abundantly in the massive rhodonite and garnet that is attached to the specimen of pyrosmalite from the North Broken Hill mine and ragged unreplaced residuals of rhodonite are embedded in it (fig. 4). Unreplaced residuals of knebelite within a plate of pyrosmalite are shown in fig. 5 and the similar orientation of both large and small residuals indicates that they have been derived from a single crystal. The varying proportion of manganese to iron in pyrosmalite can be



FIGS. 3-6.

FIG. 3. Ragged plate of pyrosmalite (*P*) in quartz, with associated dannemorite (lower left), galena, garnet, and needles of dannemorite or actinolite. D.D. 588 at 89 feet, No. 11 level, Junction mine, North Broken Hill Ltd.  $\times 13$ .

FIG. 4. Pyrosmalite (*P*) replacing rhodonite, with some garnet at lower right. Section mounted in medium with a refractive index of 1.66 to reduce relief. About 75 feet above 1700-foot level, No. 11 section, North Broken Hill mine.  $\times 20$ .

FIG. 5. Pyrosmalite (*P*) enclosing unreplaced residuals of roepperite (zinc-bearing knebelite). Black areas are galena. Junction North mine, North Broken Hill Ltd.  $\times 60$ .

FIG. 6. Pyrosmalite (*P*) showing alteration along the contact with galena (black) within a crystal of rhodonite. The cleavage of the pyrosmalite extends into the altered margin and the cleavage of the rhodonite extends as a crack into pyrosmalite. King shaft ore body, D.D. 602, between 24 feet and 30 feet, No. 10 level, Junction mine, North Broken Hill Ltd.  $\times 60$ .

expected to be a reflection of the varying proportions of manganese to iron in the replaced silicate. The small percentage of lime may likewise be derived from the replaced silicate and it is possible that the small percentage of zinc may come from the same source. The widespread occurrence of apatite within the bodies of manganese silicates reveals the presence of chlorine in the early stages of lode formation.

The common association of pyrosmalite with galena within the bodies of manganese silicates indicates that this type of hydration and chlorination of the manganese silicates occurs with the introduction of the sulphides. Pyrosmalite is thus a late hydrous manganese silicate in contrast to the early or high-temperature anhydrous silicates—rhodonite, pyroxmangite, knebelite, bustamite, and manganhedenbergite. This type of hydration of the manganese silicates is limited in degree and is probably much less than the hydration that produces bementite, sturtite, or their equivalents.

Pyrosmalite is not found associated with blende in the same way as with galena, and one can surmise that in the presence of blende the available manganese and iron is absorbed in the composition of the latter. Blende occurring with pyrosmalite from the vugh from the 1370-foot level, Broken Hill South mine, contains 1.56 % Mn and 11.66 % Fe.

When pyrosmalite is observed in small particles in thin sections, it is apt to be confused with a colourless mica from which it is distinguished by a higher refractive index and a strictly uniaxial interference figure. Moreover, pyrosmalite undergoes alteration to a greenish mineral with a faint but distinct pleochroism. This alteration is shown in fig. 6 along a contact with galena in a crystal of rhodonite. The altered zone is finely fibrous between crossed nicols with the fibres at right angles to the contact and parallel to the cleavage of pyrosmalite. This greenish mineral is often observed within galena in rhodonitic ore, where it has been found to have a negative uniaxial figure, high birefringence, and refractive indices straddling 1.54; its identity is uncertain but it appears comparable, except in colour, with parsettensite, another hydrous manganese silicate. Finely fibrous margins from this mineral have been observed to extend into the surrounding galena and may indicate the origin of the clusters of very fine fibres that are commonly observed in galena on a polished surface of rhodonitic ore but are too thin to be clearly observed in thin section. These fibres have not been observed in blende, which, in contrast to galena, contains some manganese in its composition.

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