MINERALOGICAL MAGAZINE, SEPTEMBER 1979, VOL. 43, PP. 433–5

Margarite from the Olary Province of South Australia

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SYNOPSIS

IN Lower Proterozoic rocks of the Olary Province of South Australia margarite occurs in conjunction with sodic muscovite, pseudomorphing chiastolitic andalusite in graphite-rich schist, and as a pseudomorphic phase in rocks consisting of coarse muscovite together with lesser amounts of chloritoid, sillimanite, staurolite, and corundum. In the latter occurrence the assemblage (muscovite–sillimanite – chloritoid $\pm$ staurolite $\pm$ corundum) replaces massive andalusite. The margarite and sodic muscovite within the pseudomorphs lack a preferred orientation which suggests that their development was associated with the waning stages of the Palaeozoic Delamerian Orogeny (cf. Glen *et al.*, 1977) which was the last major tectonothermal event in the region.

Within the chiastolitic andalusite pseudomorphs, equal proportions of margarite and sodic muscovite are separated from the rock matrix by a rim of coarser-grained muscovite, which has developed at the pseudomorph margins. A suggested reaction for the development of margarite is

$$10Al_2SiO_5 + 1.7Ca^{2+} + 1.2K^+ + Na^+ + 10H_2O \rightleftarrows (Ca_{1.6}Na_{0.4})Al_4(Si_4Al_4O_{20})(OH)_{3.6} + (K_{1.2}Na_{0.6}Ca_{0.1})Al_4(Si_6Al_{20}O_{20})(OH)_4 + 6Al^{3+} + 12.4OH^-.$$

The excess aluminium and hydroxyl ions from the above reaction leave the pseudomorph system but probably react with free quartz and more K^+ ions to produce the muscovite fringe.

$$6SiO_2 + 2K^+ + 6Al^{3+} + 12OH^- \rightleftarrows K_2Al_4(Si_6Al_2O_{20})(OH)_4 + 8H^+.$$

The above reactions are pertinent only for margarite and sodic muscovite produced by the pseudomorphism of chiastolitic andalusite.

Margarite and sodic muscovite in the muscovite–chloritoid – sillimanite $\pm$ staurolite $\pm$ corundum rocks replace coarser-grained (up to 1 mm) muscovite, sillimanite, corundum, and chloritoid; staurolite is unaffected. The coarse muscovite of the earlier pseudomorphic assemblage contains 7–16 mole% paragonite in solid solution whereas the sodic muscovite assocated with margarite contains 22–33 mole% paragonite in solid solution. Margarite contains negligible muscovite but substantial (17–26 mole%) paragonite in solid solution. The phases plagioclase and paragonite were not detected in any of the investigated samples. Staurolite has an Mg/(Mg+Fe) value of 0.21 and associated chloritoid a value of 0.20.

It is possible that much of the previously described sericitic alteration of andalusite (cf. D'arcy, 1977) within schists of the north-eastern Willyama Complex (north of Broken Hill) involves the production of margarite.

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[*Manuscript received 26 February 1979*]

MARGARITE FROM THE OLARY PROVINCE OF SOUTH AUSTRALIA

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The occurrence of margarite, once considered a rare mineral (Velde, 1971), has been recently reported from many locations (Jan et al., 1971; Frey and Niggli, 1972; Hock, 1974; Chinner, 1974; Lanphere and Albee, 1974; Guidotti and Cheney, 1976) but to the author's knowledge it has not previously been reported from Australia.

In South Australia margarite has been found in Lower Proterozoic aluminous rocks in the Olary Province, which is situated approximately 160km west of Broken Hill and approximately 330km NNE of Adelaide. The metamorphics of the Olary Province, which are a westward extension of the well-known Willyama Complex at Broken Hill (Binns, 1964; Vernon, 1969) have been multiply deformed and metamorphosed (Glen et al., 1977; Berry et al., 1978) and are overlain unconformably by Upper Proterozoic metasediments (Adelaidean series).

Within the Lower Proterozoic metamorphics of the Olary Province, chiastolitic andalusites have been partially or totally pseudomorphed by margarite and/or sodic muscovite but still preserve their graphite cross. The occurrence described by Guidotti and Cheney (1976), from the Rangeley Area, Maine, is similar. Massive andalusite is also developed in the Olary Province and in this form it has undergone a more complex pseudomorphism (Oliver, in prep.). The lack of any preferred orientation of margarite and muscovite within these pseudomorphs suggests that their development was associated with the waning stages of the Palaeozoic Delamerian Orogeny (c.f. Glen et al., 1977) which was the last major tectonothermal event in the area. This paper describes three margarite-bearing specimens from this area.

Chemistry of the margarite-bearing pseudomorphs. The chemical analyses presented in Table I reflect to a certain extent the chemistry of the precursor material which is assumed to have been massive andalusite in samples 454-1301 and 454-382 and a chiastolite porphyroblast in sample Bimb/And; these have been totally pseudomorphed during subsequent metamorphic events. The analyses (Table I), compared with the composition of normal andalusite, indicate that during pseudomorphism there has been a substantial loss of Al_2O_3 (in the order of 15-20 wt.%) from the system whereas SiO_2 has remained almost constant and CaO, K_2O and Na_2O have been added in varying amounts. The amount of CaO present in each sample determines the modal % margarite as no other phase contains appreciable amounts of CaO.

Petrology of the margarite-bearing pseudomorphs. Sample Bimb/And is a totally pseudomorphed chiastolite from a graphite-rich schist which contains chiastolitic andalusites up to 14 cm in length and 2-3 cm in width set in a matrix of quartz, muscovite, graphite (up to 8 modal %), minor chlorite, and accessory tourmaline and rutile. The analysis presented in Table I (Bimb/And) represents the chemical composition of a pseudomorphed chiastolite with no matrix included. A relatively coarse-grained muscovite with graphite inclusions developed at the margins of the pseudomorph and separates the margarite and sodic muscovite within the pseudomorph from the rock matrix. Equal quantities of margarite and muscovite occur within the pseudomorph; the grains are randomly oriented. Margarite can be distinguished optically from muscovite by its lower birefringence and higher relief.

Sample 454-382 contains large blue corundum crystals (~1 cm), coarse muscovite, and finer-grained (~0.4 mm) chloritoid, all of which are partially replaced by a fine-grained aggregate of margarite and sodic muscovite. The margarite, which constitutes approximately 8 modal % of the sample, is well twinned with a much higher relief than co-existing muscovite. Small trails of rounded tourmaline grains have developed along with margarite and sodic muscovite. Tourmaline is sometimes associated with chloritoid breakdown. The large corundum grains have inclusions of earlier-formed coarse muscovite which is similar to that being replaced by the finer-grained micas in the matrix. The muscovite inclusions in corundum are unaffected by margarite and sodic muscovite replacement. Situated at the margins of the large corundum porphyroblasts are small remnant corundum grains which have escaped replacement by the fine micas. These smaller corundum grains (originally part of the larger porphyroblast) are partially altered to diaspore.

Table I. Major element analyses of margarite-bearing pseudomorphs

	454/1301	454/382	Bimb/And
SiO_2	37.42	37.31	39.75
Al_2O_3	47.76	47.37	43.60
Fe_2O_3*	2.31	1.15	0.52
MnO	0.05	<0.01	<0.01
MgO	0.37	0.24	0.50
CaO	0.04	1.19	4.75
Na_2O	1.18	1.02	N.D.
K_2O	6.85	7.36	4.60
TiO_2	0.06	0.09	0.03
P_2O_5	0.01	0.05	0.03
L.O.I.	3.67	4.33	N.D.
Total	99.72	100.09	93.78

: 454/1301 - Muscovite - Staurolite - Chloritoid - Margarite - Corundum - Sillimanite

: 454/382 - Muscovite - Margarite - Chloritoid - Corundum - Tourmaline

: Bimb/And - Margarite - Muscovite

*Total iron analysed as Fe_2O_3.

N.D. - Not determined.

(Analyst for Bimb/And, Dr. R.H. Flood, Macquarie University).

Sample 454-1301 exhibits textures and mineralogy indicative of a complex metamorphic history. The sample contains a coarse (up to 1 mm) and a fine-grained muscovite, together with corundum, staurolite, chloritoid, fibrolitic sillimanite, and rare margarite. Associated rocks still have andalusite preserved, and although the sample under discussion does not contain andalusite there are small trails of corundum grains within the coarser muscovite which appear to outline pre-existing andalusite cleavage traces. Fibrolitic sillimanite is present as folded trails within chloritoid, staurolite, and coarse muscovite. Fine-grained sodic muscovite and rare margarite replace the coarse muscovite and its included fibrolitic sillimanite and have marginally replaced the chloritoid, whereas staurolite is unaffected.

Quartz is absent from samples 454-1301 and 454-382 and in sample Bimb/And it is present in the matrix only. Margarite within the chiastolite pseudomorph appears to be armoured from quartz by the rim of coarser muscovite. Paragonite and feldspar were not detected in the samples investigated.

Mineral chemistry. Table II provides chemical data for margarite and other phases present in the rocks under discussion. Phases from samples 454-1301 and 454-382 were analysed using a TPD microprobe following the method of Reed and Ware (1975), whilst phases from the Bimb/And sample were analysed using an Etec Autoprobe following the method of Bence and Albee (1968). The margarites investigated contain negligible muscovite but substantial paragonite in solid solution (17-26 mole %). Co-existing fine-grained muscovites (numbered 1 in Table II) are Al-saturated and quite sodic, with a range of from 22 to 33 mole % paragonite in solid solution. Fig. 1 is a Na-Ca-K plot of co-existing margarite-muscovite pairs from the three samples analysed.

The coarse muscovites of samples 454-1301 and 454-382 that are partially replaced by the above micas are less sodic, with 17 and 7 mole % paragonite respectively. These coarse muscovites together with minor corundum are thought to represent an earlier pseudomorphic replacement of andalusite.

Table II. Analyses of margarite and associated minerals.

	Sample No. 454/1301				
	Marg.	Musc.(1)	Musc.(2)	Staurolite	Chloritoid
SiO_2	31.31	45.95	44.51	26.63	24.88
Al_2O_3	50.12	38.35	37.70	56.20	42.94
FeO^a	0.25	0.54	0.54	9.61	23.39
MnO	0.00	0.00	0.00	0.30	0.85
MgO	0.47	0.35	0.40	1.46	3.29
Na_2O	2.09	2.64	1.27	0.00	0.59
CaO	10.75	0.00	0.00	0.00	0.00
K_2O	0.14	8.14	9.91	0.00	0.00
ZnO	-	-	-	4.50	-
Total	95.13	95.97	94.33	98.70	95.94
	22(0)	22(0)	22(0)	46(0)	12(0)
Si	4.155	6.002	5.961	7.362	1.972
Al^{iv}	3.845	1.998	2.039	(18.310)	4.010
Al^{vi}	3.995	3.906	3.912		
Fe^{2+}	0.028	0.059	0.060	2.222	1.550
Mn	0.000	0.000	0.000	0.070	0.057
Mg	0.093	0.068	0.080	0.602	0.389
Zn	0.000	0.000	0.000	0.918	0.000
Ca	1.529	0.000	0.000	0.000	0.000
Na	0.538	0.669	0.330	0.000	0.091
K	0.024	1.356	1.693	0.000	0.000
Total Y	4.116	4.033	4.052		
Total X	2.091	2.025	2.023		

	Sample Bimb/And			Sample 454/382		
	Marg.	Marg.	Musc.	Marg.	Musc.(1)	Musc.(2)	Chloritoid
SiO_2	30.94	31.60	44.60	30.86	45.20	44.83	24.04
Al_2O_3	50.04	50.37	38.64	51.04	38.40	38.06	41.92
FeO^a	0.28	0.29	0.41	0.36	0.44	0.35	24.25
MnO	0.00	0.00	0.00	0.00	0.00	0.00	0.72
MgO	0.14	0.15	0.22	0.29	0.22	0.16	2.75
Na_2O	1.35	1.76	2.17	1.59	1.68	0.49	0.31
CaO	11.77	10.99	0.99	10.73	0.00	0.18	0.00
K_2O	0.00	0.00	7.17	0.11	9.28	10.65	0.06
ZnO	-	-	-	-	-	-	-
Total	94.52	95.16	94.20	94.98	95.22	94.72	94.05
	22(0)	22(0)	22(0)	22(0)	22(0)	22(0)	12(0)
Si	4.130	4.183	5.914	4.095	5.971	5.980	1.956
Al^{iv}	3.867	3.817	2.086	3.905	2.029	2.020	(4.019
Al^{vi}	4.010	4.041	3.952	4.077	3.950	3.963	(
Fe^{2+}	0.031	0.032	0.046	0.040	0.049	0.039	1.650
Mn	0.000	0.000	0.000	0.000	0.000	0.000	0.050
Mg	0.028	0.030	0.044	0.057	0.043	0.032	0.334
Zn	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ca	1.684	1.559	0.141	1.526	0.000	0.026	0.000
Na	0.350	0.452	0.558	0.409	0.430	0.127	0.049
K	0.000	0.000	1.213	0.019	1.564	1.812	0.006
Total Y	4.069	4.103	4.042	4.174	4.042	4.034	
Total X	2.034	2.011	1.912	1.954	1.994	1.965	

a Total Fe as FeO.

(Analyst for Bimb/And, Dr. R.H. Flood, Macquarie University.

The staurolite of sample 454-1301 has a Si content below the range (7.64-7.94 atoms of Si per formula unit) quoted by Griffen and Ribbe (1973) from a compilation of twenty analyses. It has negligible or undetectable TiO_2, MnO, CaO, Na_2O, and K_2O but has high Al_2O_3 and ZnO and consequent low FeO values (Table II). MnO is partitioned strongly into chloritoid, with staurolite the only other phase containing manganese.

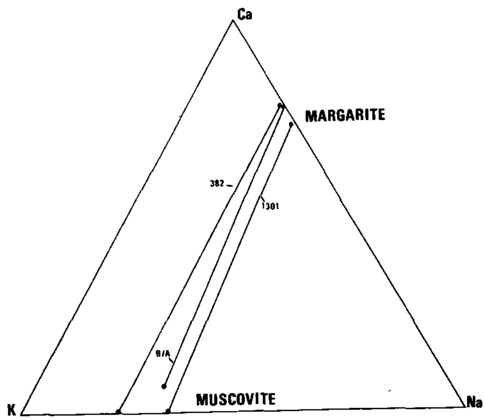

Fig. 1. Co-existing margarite-muscovite pairs plotted on Na-Ca-K diagram.

Discussion. This paper presents data on unusual quartz-free margarite-bearing pseudomorphs which are low in SiO_2 but rich in Al_2O_3, with Al_2O_3 + SiO_2 comprising approximately 85 wt % of the bulk sample. Some pseudomorphs are complex mineralogically (e.g. 454-1301) as a result of the metastability of earlier formed phases, with only sodic muscovite, diaspore and tourmaline co-existing stably with margarite.

It is possible to postulate a reaction that could have led to the formation of margarite in the pseudomorphed chiastolite sample (Bimb/And). Any reaction proposed must take account of (1) the fact that equal proportions of margarite and muscovite are present within the pseudomorph; (2) the muscovite fringe about the perimeter of the pseudomorph; and (3) Al_2O_3 mobility. Using the chemical formulae for muscovite and margarite (Table II) and taking into account the above factors the following reaction is suggested:

$$10\ Al_2SiO_5 + 1.7\ Ca^{2+} + 1.2\ K^+ + Na^+ + 10\ H_2O$$
$$\rightleftharpoons (Ca_{1.6}Na_{0.4})\ Al_4\ (Si_4\ Al_4\ O_{20})\ (OH)_{3.6} +$$
$$(K_{1.2}\ Na_{0.6}\ Ca_{0.1})\ Al_4\ (Si_6\ Al_2\ O_{20})\ (OH)_4 + 6\ Al^{3+} + 12.4\ OH^-$$

The excess aluminium and hydroxl ions from the above reaction leave the pseudomorph system but probably react with free quartz and more K^+ ions to produce the muscovite fringe.

$$6\ SiO_2 + 2K^+ + 6\ Al^{3+} + 12\ OH^- \rightleftharpoons K_2\ Al_4\ (Si_6\ Al_2\ O_{20})\ (OH)_4 + 8H^+$$

The above reactions satisfy chemical, modal and textural observations in sample Bimb/And only. In the other samples investigated the assemblage margarite-muscovite replaced earlier pseudomorphic coarse muscovite-fibrolitic sillimanite-corundum and the story is more complex.

Margarite has been noticed in thin sections of a number of other chiastolitic schists from within the Olary Province. It is possible that much of the previously described sericitic alteration of chiastolitic andalusite (c.f. D'arcy, 1977) within schists of the north-eastern Willyama Complex (north of Broken Hill) involves the production of margarite.

Acknowledgments. The author would like to thank Drs. R.L. Oliver, R.H. Flood, and I.R. Plimer for commenting on an earlier draft of this paper. Dr. R.H. Flood also supplied the sample and analyses of Bimb/And. The author would also like to thank Mr. N. Ware of the Research School of Earth Sciences, Australian National University for instruction in and access to electron microprobe facilities.

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