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Margarite in the Central Alps*

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Abstract

A distribution map for margarite of the Central Alps based on some 270 specimens from 47 localities is presented. Margarite-bearing rocks formed during the Tertiary Alpine metamorphism and range from lower greenschist to upper amphibolite facies. Margarite coexists with various combinations of the following minerals (listed in decreasing order of abundance): quartz, muscovite, graphite, chlorite, plagioclase, calcite, epidote group minerals, biotite, garnet, dolomite, paragonite, chloritoid, kyanite, staurolite, hornblende and corundum. Microprobe analyses of ten margarites and six coexisting muscovites are presented. A review of the crystal chemistry of margarites (based on 61 analyses) reveals that the dominant substitution is $NaSiCa_{-1}Al_{-1}$ in analogy with the plagioclase series. Appendix I is a compilation of margarite occurrences described in the literature.

Introduction

Optical, X-ray diffraction and electron microprobe work during the last fifteen years (SAGON, 1967, 1970, 1978; FREY & NIGGLI, 1972; HÖCK, 1974a, b; FREY & ORVILLE, 1974; FREY, 1978; FRANK, in prep. and others) demonstrates that margarite, $CaAl_2[Al_2Si_2O_{10}](OH)_2$, is an important rock-forming mineral. A summary of occurrences of margarite as a prograde metamorphic mineral described in the literature is presented in Appendix I. According to these data margarite is found in metamorphosed pelites, marls, bauxites, basites and anorthosites and at metamorphic grade ranging from lower greenschist to upper amphibolite facies. In addition, margarite often forms pseudomorphs after other Al-rich minerals such as andalusite, kyanite, sillimanite or corundum, and, more rarely, after chloritoid, staurolite or even muscovite (e.g. HIETANEN, 1963; URONO & KANISAWA, 1965; RAMBERG, 1967, p. 115; VELDE, 1970; JAN et al., 1971; CHINNER, 1974; LANPHERE & ALBEE, 1974, p. 547; GUIDOTTI & CHENEY, 1976; MILLER, 1977, p. 227; GUIDOTTI et al., 1979; TEALE, 1979; YARDLEY et al., 1979; COOPER, 1980; BALTATZIS & KATAGAS, 1981).

* *Dedicated to Professor Ernst Niggli on the occasion of his 65th birthday.*

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The aim of this communication is to present a distribution map for rock-forming margarite of the Central Alps and to provide further chemical data for margarite as well as some coexisting muscovite. In addition, the crystal chemistry of margarite will be discussed in some detail. The phase relations of margarite-bearing rocks of the Central Alps in the system CaO-Al_2O_3-SiO_2-(C-O-H) will be presented elsewhere (BUCHER et al., 1983).

Early descriptions of margarite from the Central Alps

During a literature search for margarite in the Swiss Alps NIGGLI (1955) was able to find some eight references. According to this compilation GRUBENMANN (1888) was the first to identify margarite as a rock-forming mineral. He used a wet chemical analysis of minerals in a mesometamorphic schist from Lake Ritom (locality 25 in Fig. 1 of this paper). This occurrence was later confirmed by HARDER (1956, p. 245 and Table 8) who mentions the assemblage margarite-muscovite- paragonite-zoisite-garnet with accessory quartz, biotite and plagioclase. The occurrence of «clintonite» described by SCHMIDT (1891) from Lukmanier pass (locality 30 in Fig. 1 of this paper) was re-examined by NIGGLI (op. cit.) using X-ray analysis and found to be margarite. The remaining six margarite occurrences mentioned by Niggli are either doubtful due to the lack of precise optical, chemical and X-ray data or are regarded as late-stage products; those in open fissures or joints will not be discussed further here.

NIGGLI (op. cit.) also suggested that margarite might be an important mineral in relatively Ca-rich mesometamorphic schists on the southern border of the Gotthard «massif» and the lower to middle Pennine nappes. This suggestion was confirmed in part by FREY & NIGGLI (1972) who reported in a short note that margarite is an important rock-forming mineral in a Liassic black shale formation of epimetamorphic grade. More detailed data were later published by FREY (1978). Other recent references to margarite occurrences of the Central Alps can be found in Appendix II.

Distribution of margarite in the Central Alps

More than 270 margarite-bearing specimens from 47 localities are represented in Fig. 1. Margarite in all these samples was verified either by microprobe analyses, X-ray diffraction studies or optical methods by the present authors. Different symbols are used for the localities with quartz-free (No. 1–5) and quartz-bearing (No. 6–47) assemblages. Table 1 is a list of minerals which were encontered in margarite-bearing rocks. These rocks belong to different tectonic

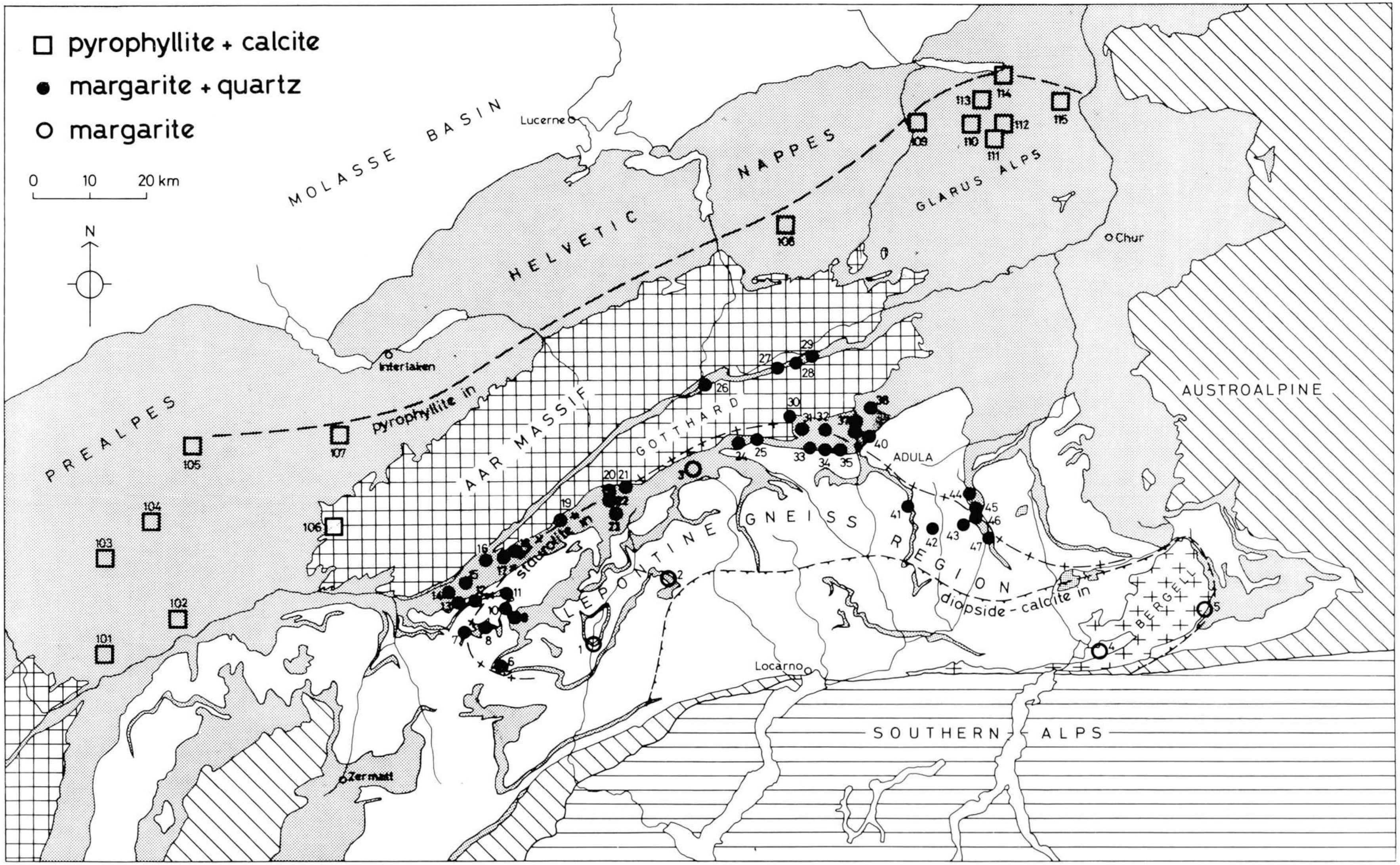

Fig. 1 Margarite distribution map of the Central Alps. Localities with mineral assemblages and sources of data are listed in Appendix II. Mineral zone boundaries are after NIGGLI (1970) for staurolite-in, TROMMSDORFF (1972) for diopside-calcite-in and FREY & WIELAND (1975) as well as unpublished data for pyrophyllite-in.

Table 1 Minerals coexisting with margarite in 271 specimens from the Central Alps.

mineral name	no of specimens	%
quartz	267	98.5
muscovite	265	97.8
graphite	235	86.7
chlorite	219	80.8
plagioclase	174	64.2
calcite	162	59.8
epidote-group	145	53.5
biotite	123	45.4
garnet	84	31.0
dolomite	84	31.0
paragonite	68	25.1
chloritoid	50	18.5
kyanite	24	8.9
staurolite	17	6.3
hornblende	11	4.1
corundum	3	1.1

units[2]) and occur in a variety of lithologies. The greatest concentration of margarite occurrences is in the Mesozoic cover of the Gotthard «massif» (No. 15–22, 24–38), particularly in metamorphosed graphitic pelites and marls of Liassic age as described by FREY & NIGGLI (1972), FREY & ORVILLE (1974), FOX (1974, 1975), FREY (1978) and FRANK (1979a). Some margarite occurs in graphite-free metamarls of the Upper Triassic Quartenschiefer formation where this mineral was overlooked by FREY (1969) but subsequently reported by FOX et al. (1974).

Margarite is a major mineral in graphitic Bündnerschiefer/schistes lustrés, that is, in metamorphosed Mesozoic marls with minor pelites of the Pennine domain between the Simplon area in the west (No. 6–9, 11, 13) and the Misox zone in the east (No. 44–47). However, a systematic search for this mineral has so far been undertaken only in the Simplon area (FRANK, 1979a, in prep.) and the Misox zone (TEUTSCH, in prep.).

Margarite seems to be a rare mineral in pre-Mesozoic pelitic schists of the Pennine nappes. KLEIN (1976), for example, reported this Ca-mica from only three out of about 400 samples studied from the Adula and Simano nappes (No. 41–43) and a peculiar occurrence of margarite was described by FRANK (1979b) from a zoisite-plagioclase gneiss of the Berisal nappe (No. 10). In addition, margarite was described from two marbles of possible Mesozoic age from the Bergell area (No. 4: MOTICSKA, 1970; No. 5: H. R. WENK, pers. comm.)

Margarite shows a simple pattern of regional distribution as do other index minerals of the Central Alps (e.g. NIGGLI & NIGGLI, 1965). About half of the

[2]) Tectonic units are designated according to the tectonic map of Switzerland (Spicher, 1980).

margarite localities shown in Fig. 1 are located in Niggli's chloritoid zone, while the other half occurs in the staurolite zone. This distribution pattern may change somewhat as more petrographic data become available. It is expected that margarite may also be found in the Zermatt area, in the Urseren zone between the western end of the Gotthard «massif» and Andermatt (that is between localities 16 and 26), and in Bündnerschiefer rocks in an area between the eastern end of the Gotthard «massif», Chur and the Bergell area. The present low-grade margarite mineral zone boundary does not correspond to a reaction-isograd (WINKLER, 1979, p. 66). According to FREY (1978) the first margarite was formed from pyrophyllite + calcite or from pyrophyllite + dolomite. These assemblages are found in the Helvetic nappe zone and the Prealps (Fig. 1, No. 101–115). However, due to unfavourable bulk compositions in the southern Glarus Alps and the Aar massif and lack of field data in the sedimentary cover at the SW end of the Aar massif a reaction-isograd has not yet been mapped.

The majority of margarite-bearing samples occur in Mesozoic rocks and were therefore formed during the Alpine regional metamorphism. From textural evidence this is believed also to be true for margarites found in some pre-Mesozoic rocks. More specifically, the areal distribution and textural evidence suggest that all margarites were formed during mid-Tertiary time.

Textural observations

In thin section margarite occurs mainly in the matrix or as porphyroblasts but pseudomorphic margarite was also observed.

Synkinematic margarite is found in the matrix of many phyllites and mica schists of the lower and middle greenschist facies. If intergrown on a small scale with other sheet silicates, especially muscovite and paragonite, it is difficult to detect; and optical and X-ray methods (FREY, 1978, p. 114) are needed. However, almost monomineralic layers of margarite are found in some metamarls of the greenschist facies and are then easily identified with the microscope. In some thin sections it could be observed that these margarite-rich layers pass laterally into plagioclase-rich layers (Fig. 2a). From this observation and the regional distribution pattern of margarite and plagioclase it is believed that much plagioclase in Liassic black shales and the Bündnerschiefer formed by margarite-consuming reactions (see also FREY & ORVILLE, 1974; BUCHER et. al., 1983, Fig. 3).

Postkinematic porphyroblasts of margarite were found in phyllites and mica schists of the upper greenschist and lower amphibolite facies. At localities 21 and 30 these porphyroblasts reach a length of several millimetres. In thin section (Fig. 2b; see also FREY, 1978, Fig. 8; FRANK, in prep.) they can be easily dis-

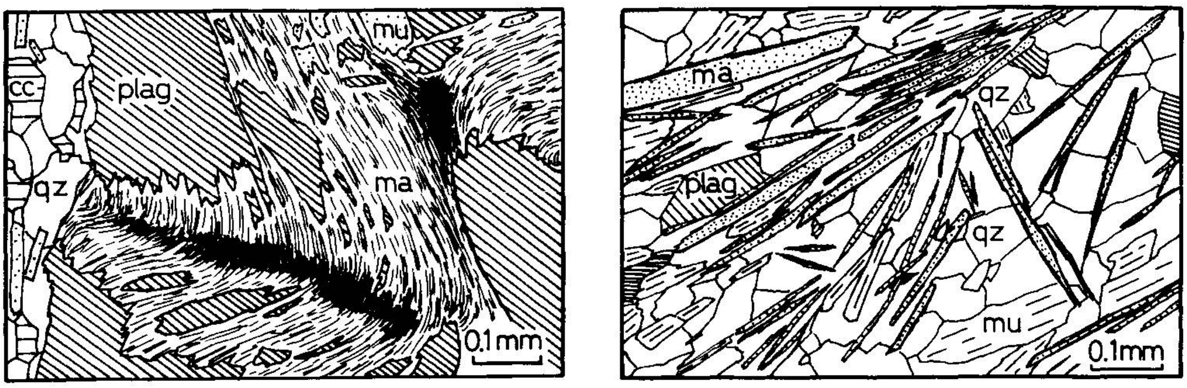

Fig. 2 Margarite textures
a) Syn- and pre-kinematically grown margarite layer partially replaced by plagioclase. Sample EF 41, locality 11.
b) Porphyroblastic margarite. Sample MF 1934, locality 21.
c) Margarite + chlorite pseudomorphic after chloritoid. Sample MF 1608, locality 24.

tinguished from muscovite or paragonite by higher relief, lower birefringence, and angle of extinction.

Magarite and chlorite pseudomorphous after chloritoid porphyroblasts were found at locality 24 close to the staurolite «isograd» (Fig. 2c). Due to the lack of chemical data no detailed reaction can be proposed at present. Another example of chloritoid replaced by margarite (and muscovite) was mentioned by TEALE (1979).

Mineral chemistry

Mineral analyses were performed with an ARL-SEMQ microprobe operating in a combined energy and wavelength dispersive mode (TN 2000, PDP 11/04), that is, all elements were determined simultaneously at the same spot (SCHWANDER & GLOOR, 1980). In each thin section «coexisting» minerals in three to four areas several millimetres apart were analysed. If possible, minerals in direct contact were used, but sometimes the distance was as large as 1-2 mm.

Ten *margarite* analyses are presented in Table 2. The first nine analyses are from graphite-bearing metapelites or metamarls while analysis Sci 1519 is from a marble. These analyses come from upper greenschist and amphibolite facies

Table 2 Microprobe analyses of some margarites from the Central Alps. Samples are arranged with increasing metamorphic grade. Analysts: H. SCHWANDER and E. FRANK.

Sample No.	MF 521	MF 1934	EF 1058	Blen 25	Mis 70	Kl 327	G 147	EF 870	EF 1070	Sci 1519
locality (Fig. 1)	30	21	10	37	47	42	3	1	2	5
No. of analyses	5	15	6	4	7	3	18	2	4	5
SiO_2	29.87	30.30	32.71	32.20	32.41	32.38	30.81	30.70	30.86	29.61
TiO_2	0.10	0.05	n.d.	0.19	0.09	0.08	0.3	n.d.	n.d.	0.00
Al_2O_3	50.29	50.03	49.79	48.14	48.89	47.96	48.19	49.74	49.92	50.84
FeO*	0.27	0.39	0.13	0.72	0.55	0.86	0.51	0.25	0.41	0.17
MnO	0.04	0.02	n.d.	0.03	0.05	0.00	n.d.	n.d.	n.d.	0.00
MgO	0.26	0.15	0.41	0.00	0.54	0.38	0.2	0.10	0.97	0.40
CaO	11.39	11.96	11.09	8.65	10.86	9.14	10.48	12.58	10.76	12.10
Na_2O	1.42	0.97	1.44	1.96	1.78	1.80	1.95	0.77	1.77	0.92
K_2O	0.11	0.11	(<0.10)	0.14	0.23	0.32	0.29	0.10	(<0.10)	0.00
Total	93.75	93.98	95.57	92.03	95.40	92.92	92.73	94.24	94.69	94.04
Atomic proportions on the basis of 22 oxygen atoms										
Si	4.029	4.076	4.298	4.381	4.290	4.378	4.202	4.119	4.114	3.979
Al^{iv}	3.971	3.924	3.702	3.619	3.710	3.622	3.798	3.881	3.886	4.021
ΣZ	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al^{vi}	4.024	4.010	4.009	4.102	3.918	4.022	3.949	3.984	3.959	4.032
Ti	0.010	0.005	–	0.019	0.009	0.008	0.031	–	–	0.000
Fe^{2+}	0.030	0.044	0.014	0.082	0.061	0.097	0.058	0.028	0.046	0.019
Mn	0.005	0.002	–	0.003	0.006	0.000	–	–	–	0.000
Mg	0.052	0.030	0.080	0.000	0.107	0.077	0.041	0.020	0.193	0.080
ΣY	4.121	4.091	4.103	4.206	4.101	4.204	4.079	4.032	4.198	4.131
Ca	1.646	1.724	1.561	1.261	1.540	1.324	1.532	1.808	1.537	1.742
Na	0.371	0.253	0.367	0.517	0.457	0.472	0.516	0.200	0.457	0.240
K	0.019	0.019	–	0.024	0.039	0.055	0.050	0.017	–	0.000
ΣX	2.036	1.996	1.928	1.802	2.036	1.851	2.098	2.025	1.994	1.982
End-member molecules										
margarite	80.9	86.4	81.0	70.0	75.6	71.5	73.0	89.3	77.1	87.9
paragonite	19.2	12.7	19.0	28.7	22.5	25.5	24.6	9.9	22.9	12.1
muscovite	0.9	0.9	(<1.0)	1.3	1.9	3.0	2.4	0.8	(<1.0)	0.0

* total Fe as FeO

n.d. = not determined

grade. It was not possible to analyse margarites from the lower greenschist facies due to fine grain size. Some of the analyses show low anhydrous totals of only 92–94 wt% which are presumably caused by impurities, like water, on the surface of the thin sections. However, it is believed that calculated structural formulae are not affected by the low oxide totals since the atomic proportions are in agreement with the majority of other published margarite analyses as discussed later.

These ten margarite analyses show a relatively large variation in their Si/Al ratios (0.494–0.573), which is, for the most part, higher than in the end-member margarite (0.5). The octahedral positions are mainly occupied by Al

(3.918–4.102) with minor amounts of Mg (0.000–0.193) and Fe (0.014–0.097) but very small amount of Ti (0.000–0.031) and Mn (0.000–0.006). In the interlayer position an appreciable amount of Ca is replaced by Na (10–29% paragonite end-member), but K is always low (up to 3% muscovite end-member molecule). The substitution of Na for Ca shows a rather large range in some samples (e. g. sample Mis 70: $ma_{59.7}pa_{35.4}mu_{4.8}$ to $ma_{85.4}pa_{14.0}mu_{0.6}$) as depicted in Fig. 3. Similar large Ca-Na substitutions ranges within a single thin section were also reported by CHINNER (1974), HOINKES (1978, Fig. 7a) and SCHREYER et al. (1981, Fig. 5) and could be caused by chemical zonation in margarite or small-scale intergrowth of margarite with paragonite.

A chemical zonation was detected in samples MF 1934 and Mis 70 with relatively Na-rich rims and Ca-rich centers. A typical analysis yielded

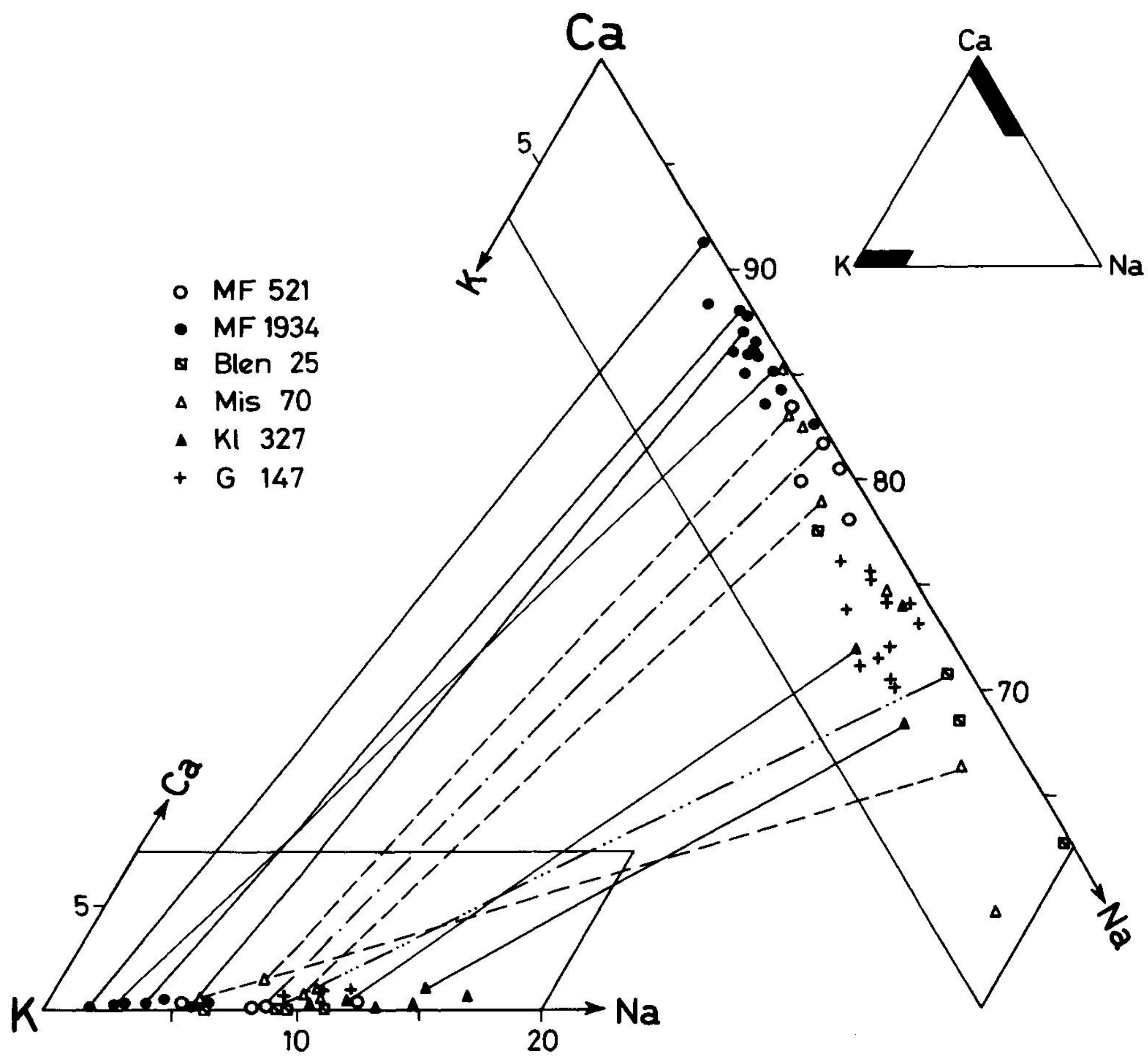

Fig. 3 Ca-Na-K plot of analysed margarites and muscovites. Tie-lines connect coexisting grains.

Table 3 Microprobe analyses of some muscovites coexisting with margarites from the Central Alps. Analyst: H. SCHWANDER.

Sample No.	MF 521	MF 1934	Blen 25	Mis 70	Kl 327	G 147
No. of analyses	4	9	4	6	6	8
SiO_2	45.16	46.23	46.83	47.18	46.59	46.40
TiO_2	0.33	0.30	0.30	0.42	0.55	0.5
Al_2O_3	34.05	34.67	33.92	32.67	33.75	33.30
FeO*	1.48	1.35	1.48	1.50	1.59	1.55
MnO	0.00	0.00	0.03	0.00	0.02	n.d.
MgO	1.38	1.03	0.42	1.82	1.07	1.6
CaO	0.05	0.05	0.02	0.10	0.05	(<0.1)
Na_2O	0.68	0.32	0.65	0.65	1.04	0.78
K_2O	10.85	10.75	9.82	10.42	9.99	10.19
Total	93.98	94.70	93.47	94.76	94.65	94.32
Atomic proportions on the basis of 22 oxygen atoms						
Si	6.194	6.198	6.325	6.325	6.248	6.249
Al^{iv}	1.806	1.802	1.675	1.675	1.752	1.751
ΣZ	8.000	8.000	8.000	8.000	8.000	8.000
Al^{vi}	3.580	3.677	3.725	3.487	3.583	3.535
Ti	0.033	0.030	0.030	0.042	0.055	0.051
Fe^{2+}	0.166	0.151	0.167	0.168	0.178	0.175
Mn	0.000	0.000	0.003	0.000	0.002	--
Mg	0.244	0.206	0.085	0.364	0.214	0.321
ΣY	4.023	4.064	4.010	4.061	4.032	4.082
Ca	0.007	0.007	0.003	0.014	0.007	--
Na	0.177	0.083	0.170	0.169	0.270	0.204
K	1.857	1.839	1.692	1.782	1.709	1.751
ΣX	2.041	1.929	1.865	1.965	1.986	1.955
End-member molecules						
muscovite	91.0	95.3	90.7	90.7	86.0	89.6
paragonite	8.7	4.3	9.1	8.6	13.6	10.4
margarite	0.3	0.4	0.2	0.7	0.4	(<0.7)

$ma_{80.5}pa_{19.1}mu_{0.4}$ (rim) $-ma_{86.2}pa_{13.1}mu_{0.7}$ (center) for MF 1934 and $ma_{66.6}pa_{30.9}$-$mu_{2.5}$ (rim) – $ma_{74.8}pa_{23.8}mu_{1.3}$ (center) for Mis 70. In samples MF 521, G 147 and Sci 1519 no chemical zoning was found while the small grain size in the remaining samples did not allow any conclusion. Zoned margarite was also reported by JONES (1971), but in that case the margins were poorer in Na than the centers.

Margarite and paragonite intergrown on a scale smaller than the electron beam presumably occurs in nature (D.J. MILTON, written comm. 1981), but no paragonite could be detected in the ten samples of Table 2 by X-ray diffraction methods.

Six analyses of *muscovite* coexisting with margarites are presented in Table 3. As noted before (e. g. HÖCK, 1974a; HOINKES, 1978) these muscovites contain more Fe + Mg + Ti than the coexisting margarites, although the «phengite» content of these muscovites is still rather small which may be the result of the aluminous bulk rock composition. In addition, the margarite component is negligible (0.2–0.7%) while the paragonite content is variable (4.3–13.6%). Note that these muscovites are more homogeneous with respect to K-Na than the margarites with respect to Ca-Na substitution (Fig. 3). Margarite-muscovite tie-lines shown in Fig. 3 for contacting grains or grains within a distance of less than 1–2 mm are subparallel within a single thin-section although some crossing tie-lines do exist in MF 1934 and Mis 70. These data suggest, that at least on the scale of several millimetres, chemical exchange-equilibrium was approached.

A few *plagioclases* were analysed for this study, but additional data can be found in FREY & ORVILLE (1974) and BUCHER et al. (1983). In general, the Ca/(Ca + Na) ratio is lower in plagioclase than in the coexisting margarite. From Fig. 4 it can be seen that plagioclases are in some cases even more inhomogeneous within a single thin section than coexisting margarites. These observations are consistent with those of ACKERMAND & MORTEANI (1973), HOINKES (1978) and GIBSON (1979).

Crystal chemistry of margarite

The following discussion is based on 10 analyses from Table 2 and 51 microprobe analyses taken from the literature (JONES, 1971 [2]*; ACKERMAND & MOR-

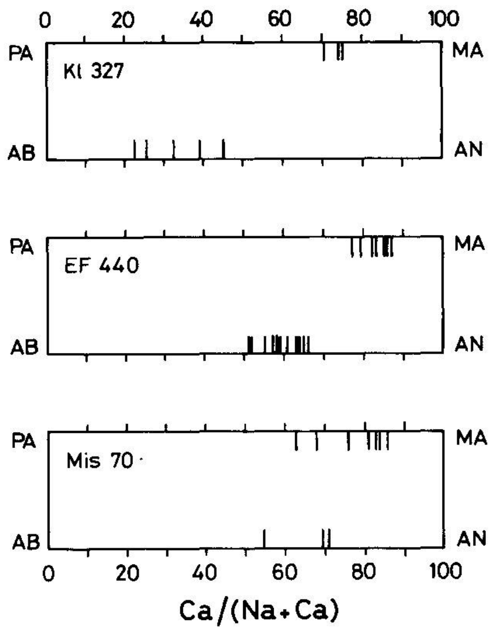

Fig. 4 Ca/(Na+Ca) plot of coexisting margarites and plagioclases. Samples Kl 327 (locality 42) and EF 440 (locality 9) come from slightly above the staurolite "isograd" while sample Mis 70 (locality 47) is located just below the staurolite "isograd". Mineral assemblages are listed in Appendix II.

* number of analyses.

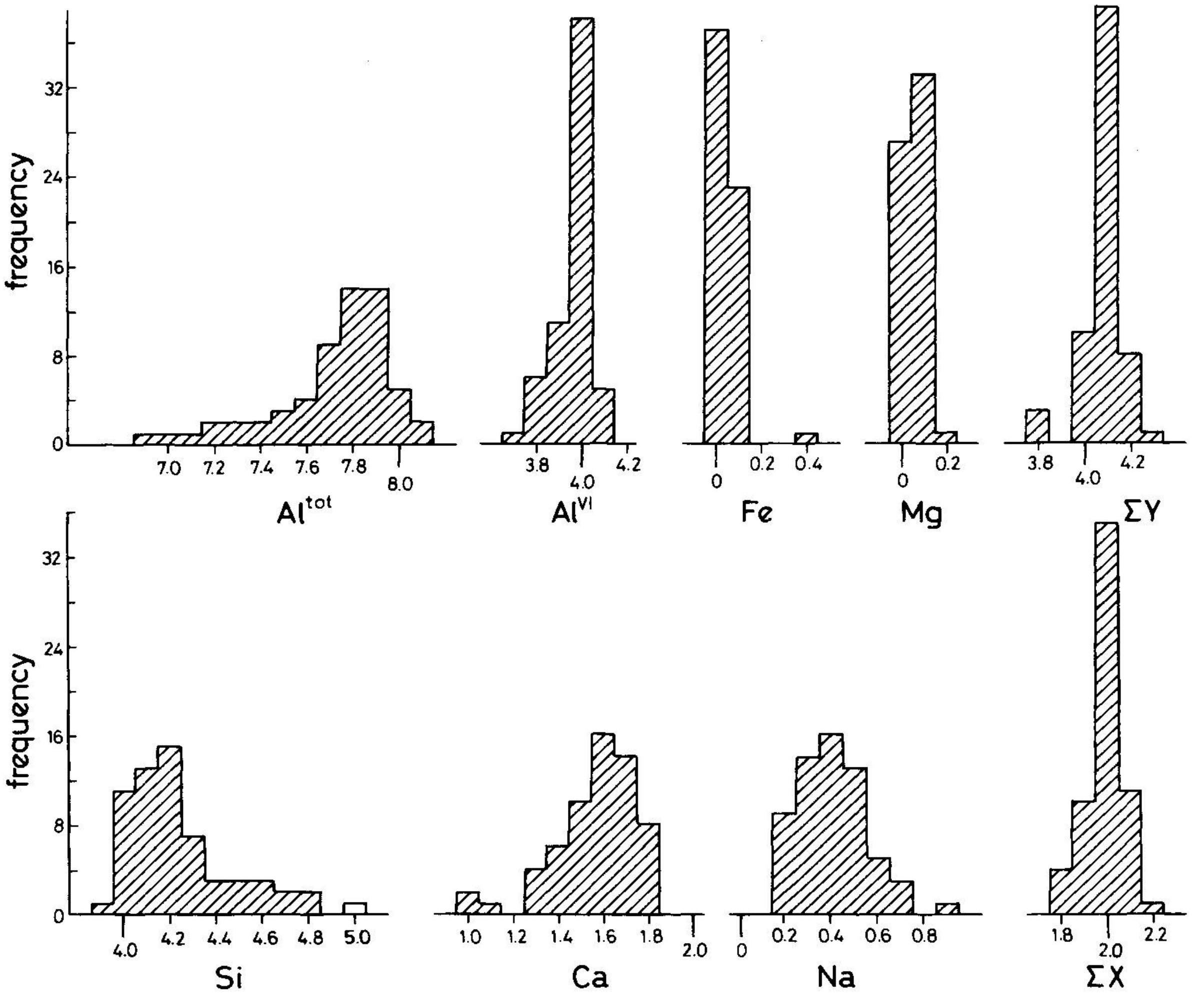

Fig. 5 Histograms showing the formula proportions of 60 margarites.

TEANI, 1973 [6]; FOX, 1974 [1]; HÖCK, 1974a [2]; GUGGENHEIM & BAILEY, 1975 [1]; GUIDOTTI & CHENEY, 1976 [1]; CHOPIN, 1977 [5]; MILLER, 1977 [1] and written comm., 1981 [2]; HOINKES, 1978 [2] and written comm., 1981 [2]; FREY, 1978 [1]; GIBSON, 1979 [6]; GUIDOTTI et al., 1979 [2]; TEALE, 1979 [4]; CRAWFORD et al., 1979 [1]; COOPER, 1980 [5]; LABOTKA, 1980 [3]; BALTATZIS & KATAGAS, 1981 [1]; SCHREYER et al., 1981 [3]).

Atomic proportions

Fig. 5 shows the distribution of some elements expressed as atoms per formula unit in the form of histograms. The following points are worth mentioning:

(1) Si ranges from 3.93 to 5.03 and the majority of natural margarites have a higher Si-value than end-member margarite (Si = 4.00). Tetrahedrally coordinated Al varies accordingly from 2.97 to 4.07.

(2) Octahedrally coordinated Al shows a narrow range of values from 3.75 to 4.10, that is with only small deviation from the 4.0 value of end-member margarite.

(3) Other octahedrally coordinated atoms are, with a few exceptions noted below, only present in small amounts. Fe (total Fe calculated as Fe^{2+}) ranges in all but one sample from 0.01 to 0.13. One margarite shows an unusually high Fe-content of 0.39 (MILLER, 1977) which might be due to an inclusion, since two other analyses from the same specimen yielded low Fe-contents of 0.05 atoms per formula unit (MILLER, written comm. 1981). Mg ranges from nil to 0.19 while Ti (0.00–0.03) and Mn (<0.01) show very small amounts. Other elements which are only rarely reported include Cr (0.00–0.20 in 16 margarites) and Li (0.002 wt-% Li_2O [FREY, 1978] and <40 ppm Li [CHOPIN, 1977]).

(4) The sum of the cations in octahedral position ranges from 3.83 to 4.28 and is in most samples slightly higher than in end-member margarite ($\Sigma Y = 4.0$). This effect is believed to be real and not only caused by analytical error, that is most natural margarites show a slight tri-octahedral character.

(5) Ca ranges from 1.00 to 1.82 with a maximum around 1.6. These values are distinctly less than that for end-member margarite (Ca = 2.0). This is mainly caused by the replacement by Na, which ranges from 0.17 to 0.90 with most values around 0.4. The K-content is always small ($\leqq 0.10$) except for two of the analyses reported by SCHREYER et al. (1981) which show unusually high K-contents of 0.15 and 0.35 atoms per formula unit. These two analyses were done on «apparently homogeneous» material, but the presence of an «intricate intergrowth of margarite with muscovite or fuchsite» (SCHREYER et al., op. cit., p. 200 and Fig. 4) suggests the possibility that a mixture of margarite and muscovite may have been analysed. Trace amounts of Ba (0.002 atoms per formula unit) are reported only from two samples by GUIDOTTI et al. (1979).

Substitutions in margarite

The most important substitution in natural margarites seems to be the coupled substitution $Na^{+}Si^{4+} = Ca^{2+}Al^{3+}$ in analogy with the plagioclase series (Fig. 6). The reason for the fact that most data points lie above the join margarite-paragonite is not clearly understood at the present time. Since Li can not be analysed with the microprobe this element is rarely reported, but if so the results do not suggest that the substitution NaLi = Ca □ (SCHALLER, 1967) towards the tri-octahedral end-member ephesite, $Na(LiAl_2)[Al_2Si_2O_{10}](OH)_2$, is important in these margarites. On the other hand a correction for a possible Tschermak-type substitution (as discussed below) would move the data points to the left, that is even farther away from the join margarite-paragonite.

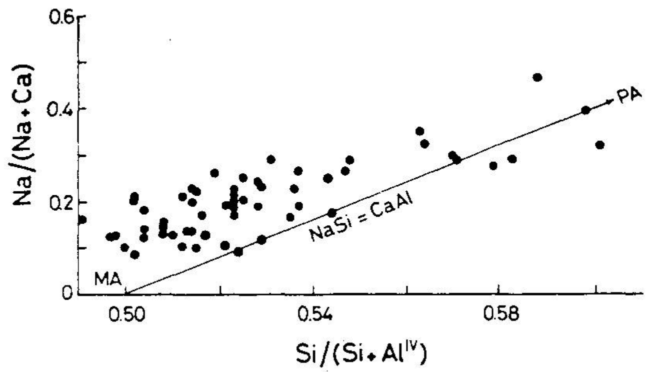

Fig. 6 Na/(Na+Ca) vs Si/(Si+Al^{vi}) diagram for 60 microprobe analyses of margarite.

It is difficult to decide how Fe and Mg are substituted in the octahedral layer since the ferric/ferrous iron ratios are not known from microprobe data (although Fe^{3+} should be minimal since most specimens with margarite also have graphite present) and because margarite show low Fe- and Mg-contents (Fig. 5). Three possible substitutions would be: $Al^{iv}Al^{vi}$ = Si(Fe^{2+}+Mg), $2Al^{3+}$ = $3(Fe+Mg)^{2+}$, and Al^{3+} = Fe^{3+}. The first mentioned substitution is well documented for the series muscovite-celadonite. However, using a triangular Si-Al-(Fe+Mg) plot (not shown here), the margarite data points showed a relatively large scatter along the Al-Si side which was somewhat reduced if a correction for the Al/Si ratio was applied caused by the above mentioned substitution NaSi = CaAl. With the low (Fe+Mg)-contents, which amount (with one exception) only up to 2% in such a Al-Si-(Fe+Mg) plot, no correlation towards a hypothetical end-member as e.g. $Ca(Mg,Fe)Al[AlSi_3O_{10}](OH)_2$ could be detected. Similarly, a $2Al^{vi}$ vs 3(Fe+Mg) plot (not shown here) displayed a large scatter with only a very weak correlation.

In Fig. 7 margarite analyses are plotted in a Ca-Na-K diagram together with some microprobe analyses of paragonites (ACKERMAND & MORTEANI, 1973 [9]*; ERNST & DAL PIAZ, 1978 [8]; FOX, 1974 [7]; GIUDOTTI et al., 1979 [2]; HÖCK, 1974a [4]; HOFFER, 1978 [3]; HOLLAND, 1979 [1]; KROGH, 1980 [3]; MARESCH & ABRAHAM, 1981 [1]; MILLER, 1974 [1]; MILLER, 1977 [3]; THOMPSON et al., 1977 [2]). The composition range of muscovites is also schematically indicated. As noted before (e.g. ACKERMAND & MORTEANI, 1973; GIBSON, 1979) margarite shows mainly a solid solution towards paragonite as is true also for muscovite. The more Na-rich margarites generally seem to contain also a slightly higher K-content (see also Fig. 3). Although the solid solution in paragonite is not as extensive as in the other two di-octahedral white micas the Ca/K ratio is quite variable and

* number of analyses.

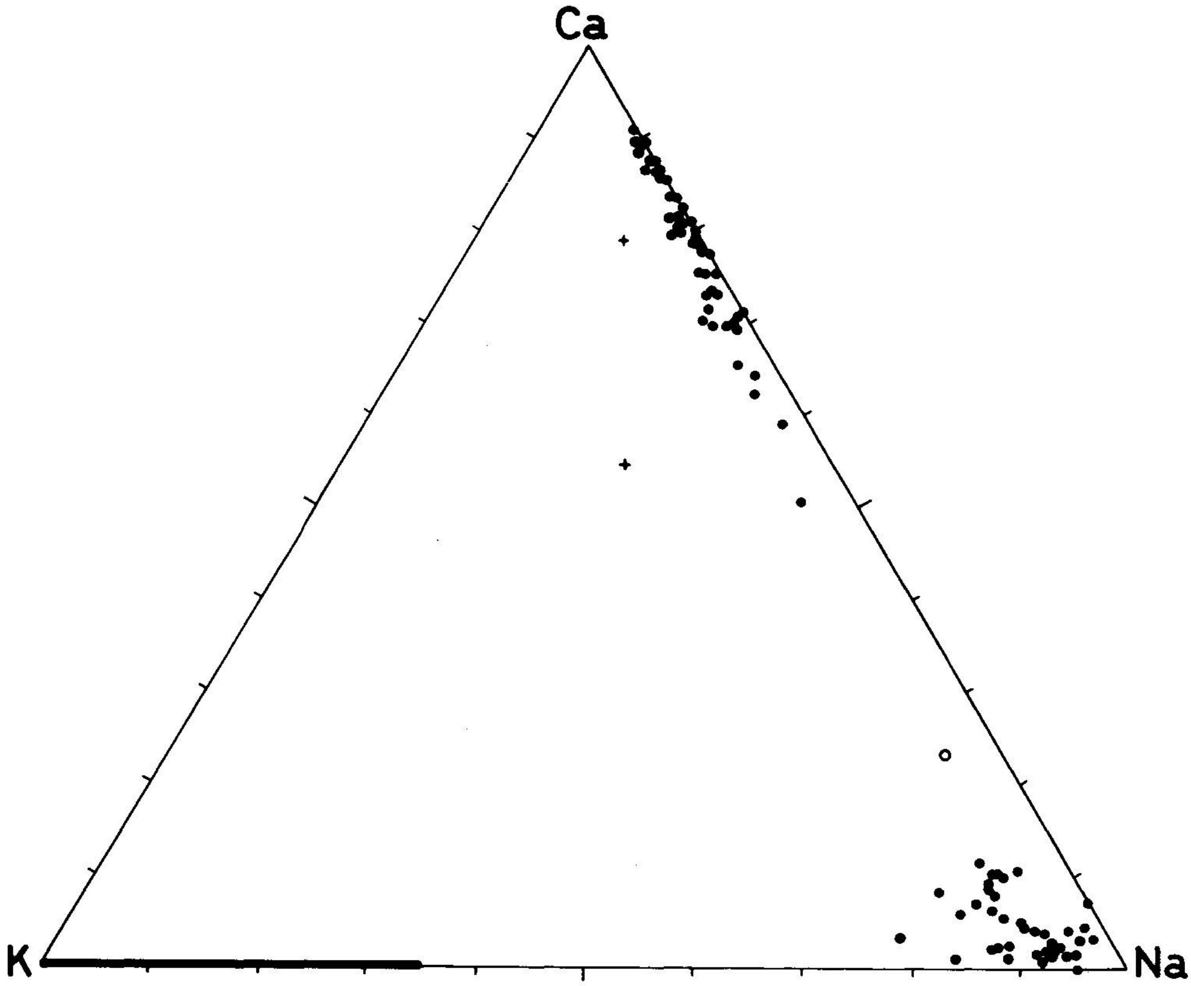

Fig. 7 Ca-Na-K end-member white mica diagram. The open circle refers to a "sodium-margarite" analysed by wet chemical methods (AFANASEV & AIDINYAN, 1952). The two crosses refer to margarite analyses possibly contaminated by muscovite (SCHREYER et al., 1981). Other data sources are given in the text.

many paragonites show similar amounts of Ca and K. The microprobe data for margarite and paragonite are consistent with the experimentally determined asymmetric solvus with a maximum at about $ma_{60}pa_{40}$ (FRANZ et al., 1977). The most Na-rich margarite analysed by microprobe technique is reported by ACKERMAND & MORTEANI (1973) and has the composition $ma_{50.8}pa_{44.2}mu_{5.0}$. Note, however, that a «sodium-margarite» formed by intrusion of a pegmatite into amphibolite has the composition $ma_{23.0}pa_{71.4}mu_{5.6}$ as determined by wet chemical technique (AFANASEV & AIDINYAN, 1952; cited in DEER et al., 1962, p. 97).

Although some differences exist, the solid solution behaviour between the three di-octahedral white micas margarite, paragonite and muscovite is similar to that in the series alkali feldspars-plagioclases.

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SMPM = Schweiz. Mineral. Petrogr. Mitt.

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Appendix I Some occurrences of margarite described in the literature. Pseudomorphic margarite and examples from the Central Alps are excluded.

No	assemblage*	metamorphic grade	area	reference
	metapelites			
1	MaQzEp - MuMtTo	upper greenschist facies	Phurni island, Greece	Köhne (1937)
2	MaQz(tr)Ky(tr)Pl - MuPaBiStIlTo	amphibolite facies	Armoricain massif, Brittany, France	Harder (1956, p. 259)
3	Ma - MuPaTo	amphibolite facies(?)	Ural, USSR	Harder (1956, p. 259)
4	MaQz - MuChRuGr MaQzPy - MuCtGr	lower greenschist facies	Armoricain massif, France	Sagon (1967, 1970, 1978)
5	MaQz - MuPaChIlToGr	lower greenschist facies	Thuringian massif, NE-Bavaria, Germany	Ludwig (1972)
6	MaQz - MuChBiCtGaOpTo MaQz - MuChCtHoOpTo MaQz - MuBiCtStOp	upper greenschist to lower amphibolite facies	Flinton-Madoc area, Grenville Province, Canada	Thompson (1972, pers. comm. 1981)
7	MaQzCzPl - ChBiGaGr	upper greenschist facies	Dalradian, Scotland	Chinner (1974, written comm. 1981)
8	MaQz - MuChGaOp	greenschist facies	Ardennes, Belgium	Bêthune de (1977)
9	MaQz - MuChBiGaOx	upper greenschist facies	Whetstone Lake area, Grenville Province	Carmichael et al. (1978)
10	MaZoKy - MuChBiGaSt	upper amphibolite facies	Coast Ranges, British Columbia, Canada	Crawford et al. (1979)
11	MaQz - Mu St MaQz - MuPa(?)Gr MaQzKy - Gr	upper greenschist facies	W Scandinavian Caledonides, Norway	Andreasson & Lagerblad (1980)
12	MaQzPl - MuChBiCt MaQzEpPl - MuChBiGaHoOp	upper greenschist facies	Whetstone Lake area, Grenville Province	Leclair (1982)

No assemblage*	metamorphic grade	area	reference
metamarls			
13 MaQzEpCc - Mu	upper greenschist facies	Cordilleran belt, SE British Columbia	Jones (1971)
14 MaQzZoCc - MuPaChCtDoPtPoGr	greenschist facies	Hohe Tauern, Eastern Alps, Austria	Höck (1974a,b)
15 MaQzCc - MuPaChGr	lower greenschist facies	Corsica, France	Delcey (1974)
16 MaQzPlCc - MuCh MaEpCoPlCc - MuChBiTr	lower greenschist facies lower amphibolite facies	Naxos island, Greece	Jansen & Schuiling (1976)
17 MaQzCzPlCc - MuChBiGa	upper greenschist facies	Schneebergerzug, Eastern Alps, Austria	Hoinkes (1978)
18 MaQz - MuChCtRu	upper greenschist facies	Funeral Mountains California, USA	Labotka (1980)
metabauxites			
19 MaDi - MuCtHeRu MaCo - MuMt MaCo - CtMtRu	greenschist facies	SW Anatolia, Turkey	Önay (1949)
20 MaCo - HcMt	high grade contact metamorphic	Shin-kiura Mine, Japan	Aoki & Shimada (1965)
21 MaCoCc - MuMt MaCoKy - BiCtMt MaCoCc - MuBiMtHe MaEpCoKy - MuBiMt MaEpKy - MuChCtMtHe MaCo - MuChCtSt MaCoPlCc - MuTo MaCo - Mt	lower greenschist facies upper greenschist facies lower amphibolite facies higher amphibolite facies	Naxos island, Greece	Jansen & Schuiling (1976), Jansen (1977)

No assemblage*	metamorphic grade	area	reference
metabasites (including some possible metamarls)			
22 MaEpKyPl - PaBiGaHoAk	lower amphibolite facies	Tauern window, Eastern Alps, Austria	Ackermand & Morteani (1973)
23 MaZo - MuChCtGa	blueschist facies	Gran Paradiso, Western Alps, France	Chopin (1977, written comm. 1980)
24 MaKy(or Co)Pl - ChPhStHoSu MaCzKyPl - MuChHoSu	amphibolite facies	central Fiordland, New Zealand	Gibson (1979)
metanorthosites			
25 MaEpKyPl - Bi	upper amphibolite facies	outer Nordfjord, W Norway	Bryhni (1966, written comm. 1980)
26 MaCoPl - Ch	amphibolite facies	SW Norway	Brueckner (1977)
27 MaCzKyPl - ChHoSu	amphibolite facies	central Fiordland, New Zealand	Gibson (1979)

* The most complete assemblages are given only, neglecting existing sub-assemblages. First quoted phases belong to the model system $CaO-Al_2O_3-SiO_2-H_2O-CO_2$

Mineral abbrevations:

Ak	ankerite	Di	diaspore	Ho	hornblende	Ox	oxide	Qz	quartz	(tr) = trace amounts
Bi	biotite	Do	dolomite	Il	ilmenite	Pa	paragonite	Ru	rutile	
Cc	calcite	Ep	epidote	Ky	kyanite	Ph	phlogopite	St	staurolite	
Ch	chlorite	Ga	garnet	Ma	margarite	Pl	plagioclase	Su	sulphide	
Co	corundum	Gr	graphite	Mt	magnetite	Po	pyrrhotite	To	tourmaline	
Ct	chloritoid	Hc	hercynite	Mu	muscovite	Pt	pyrite	Tr	tremolite	
Cz	clinozoisite	He	hematite	Op	opaque	Py	pyrophyllite	Zo	zoisite	

Appendix II Ocurrences of margarite, margarite + quartz and pyrophyllite + calcite of the Central Alps.

No.	#	locality	coordinates	tectonic/stratigraphic position	original specimen	assemblage	reference
1	1	Verampio	668.9/119.6	Bündnerschiefer	EF 870	MaKyPl	Frank(in prep.)
2	1	Bosco	679.1/130.5	Bosco series/kyanite segregation	EF 1070	MaCoKyPl-Ch	Frank(in prep.)
3	1	Misura	686.1/151.2	Tremorgio-S.Giacomo zone/ Bündnerschiefer	G 147	MaCzKyPl-MuBiGaSt	Günthert(pers. comm.)
4	1	Val Priasca	757.2/117.4	Bellinzona zone/"Triassic"?	PM 296a	MaCo-MuHc	Moticska(1970)
5	1	Preda Rossa glacier	777.0/124.6	moraine boulder/"Triassic"?	Sci 1519	MaCzCo-ChPh	Wenk(pers.comm.)
6	2	Alte Kaserne	650.0/115.3	Bündnerschiefer	Str 5497	MaQzCzKyPl-MuChBiGaGr	Frank(1979a)
7	2	Simplonpass	645.7/121.1	Bündnerschiefer	EF 835	MaQzCzPlCc-MuBi	Frank(1979a)
8	12	Kaltwasserpass	648.8/122.7	Bündnerschiefer	Str 5820b	MaQzCzPlCc-MuChBiGaGr	Frank(1979a)
			649.6/123.1	Bündnerschiefer	Str 4339	MaQzCzPl-MuBiGaHoGr	Frank(1979a)
			649.3/123.4	Bündnerschiefer	Str 7011	MaQzCzKyPl-MuBiGa	Frank(1979a)
			649.1/123.4	Bündnerschiefer	EF 270	MaQzCzPl-MuPaChGaStGr	Frank(1979a)
9	9	Alpe Veglia	653.3/126.2	Bündnerschiefer	EF 440	MaQzCzPl-MuBiGaGr	Frank(1979a)
10	1	Bortelhorn	652.4/127.0	Berisal nappe/amphibolite	EF 81	MaQzEpPl-MuPaChHo	
	1	Isenwegg	649.6/125.3	Berisal nappe/leucocratic gneiss	EF 1058	MaQzZoPl-Mu	Frank(1979b)
11	21	Steinental	650.7/127.9	Quartenschiefer	Str 4103	MaQzPl-MuPaChBiGaHo	Frank(1979a)
			650.5/127.8	Bündnerschiefer	EF 40	MaQzCzPlCc-MuChBiGr	Frank(1979a)
			652.1/128.9	Bündnerschiefer	EF 170	MaQzCzPl-MuChBiGaGr	Frank(1979a)
12	2	Eisten	646.7/127.5	Quartenschiefer	EF 981	MaQzCzPlCc-MuPa	Frank(1979a)
13	8	Schallberg	644.7/127.6	Bündnerschiefer	EF 623	MaQzPl-MuPaChCtGr	
			644.7/127.6	Bündnerschiefer	EF 627	MaQzPlCc-MuPaChGr	
			645.1/127.2	Bündnerschiefer	Str 4407	MaQzCzPlCc-MuChBiGr	
14	3	Saltinaschlucht	643.0/128.4	Quartenschiefer	EF 1021	MaQzPlCc-MuPaCt	

No.	#	locality	coordinates	tectonic/stratigraphic position	original specimen	assemblage	reference
15	9	Termen	643.8/129.4	Termen zone/"Liassic"	EF 927	MaQzPlCc-MuPaChGr	
		Simplontunnel	644.1/130.3	Termen zone/"Liassic"	Si-7	MaQzCzCc-MuPaChGr	
16	14	Bettligraben	650.0/133.7	Termen zone/"Liassic"	Lis 22.9.62.3	MaQzPlCc-MuPaChDoGr	Frey&Orville(1974)
17	3	Untergraben	652.7/135.4	Termen zone/"Liassic"	Lis 28.9.62.2	MaQz-MuChCtGr	
					Lis 15.9.62.1	MaQzPlCc-MuPaDoGr	Frey&Orville(1974)
18	5	Ausserbinn	654.8/136.0	Termen zone/"Liassic"	Lis 24.9.62.4	MaQzPl-MuPaChCtGr	Frey&Orville(1974)
			655.4/137.2	Termen zone/"Liassic"	WN 914	MaQzCz-MuChCtGaGr	
				Termen zone/"Liassic"	Str 4117	MaQzPlCc-MuChDoGr	
19	10	Blinnental	665.4/142.7	Termen zone/"Liassic"	Lis 29.8.62.15	MaQzCzPlCc-MuChBiGaGr	Frey&Orville(1974)
					Lis 29.8.62.9	MaQz-MuChBiCtGr	
20	14	Altstafel	671.5/147.0	Nufenen zone/"Liassic"	MF 1663	MaQzCzPlCc-MuChBiGaDoGr	
21	1	Nufenenstock	673.2/147.0	Nufenen zone/"Liassic"	MF 1934	MaQzCz-MuChBiGaGr	
22	1	Griessee	671.8/146.2	Nufenen zone/"Liassic"	HA 224	MaQzCzPlCc-MuBiGaDoGr	Frey&Orville(1974)
23	1	Valle di Morasco	672.4/143.1	Bündnerschiefer	EF 519	MaQzPl-MuBiGaStGr	
24	6	Camoghe	694.7/155.4	Piora zone/Quartenschiefer	MF 1608	MaQzCzPlCc-MuChBiHoDo	
				Piora zone/"Liassic"	MF 1614	MaQzCzPlCc-MuChBiGaGr	
25	6	Lago Ritom	697.0/155.7	Piora zone/"Liassic"	MF 1628	MaQzCzPlCc-MuBiGaGr	
26	1	Gotthard road tunnel	-	Urseren zone/"Liassic"	N 4250	MaQzCc-MuChDoGr	
27	8	Alp Tgom	700.8/167.7	Urseren zone/Cardinia beds	MF 857	MaQz-MuPaChCtGr	Frey(1978)
28	6	Val Gierm	704.5/169.1	Urseren zone/"Liassic"	MF 900	MaQzCc-MuChDoGr	Frey(1978)
29	1	hydroelectric tunnel	707.3/169.5	Urseren zone/"Liassic"	CS KVR 1253	MaQzCc-MuDoGr	
30	2	Val Rondadura	703.9/159.8	Scopi zone/Stgir formation	KAW 643	MaQzCz-MuChBiDoGr	Frey(1978)
31	25	Lukmanierpass	704.5/156.8	Scopi zone/Stgir formation	MF 948	MaQzCzPlCc-MuChBiGaDoGr	Frey(1978)
	1	Scopi	706.6/158.7	Scopi zone/Coroi formation	MF 1256	MaQz-MuPaChCtGr	
32	4	Stabbio Nuovo	710.7/157.2	Scopi zone/Stgir formation	MF 213	MaQzKy-MuPaChCtGr	
	1	Stabbio Nuovo	710.9/157.6	Scopi zone/Stgir formation	F 46	MaQzCzPlCc-MuBiGaDoGr	Fox(pers.comm.)
33	3	Frodalera	706.8/154.2	Piora zone/Stgir formation	MF 1576	MaQzCzKyPl-MuPaChBiGaStGr	
	4	Brönich	707.5/154.0	Piora zone/Stgir formation	MF 1584	MaQzCzKyPlCc-MuChBiGaStDoGr	Frey(1978)
34	8	Campra	709.0/153.6	Piora zone/Stgir formation	MF 1601	MaQzCzKyPlCc-MuChBiGaStGr	Frey(1978)

No.	#	locality	coordinates	tectonic/stratigraphic position	original specimen	assemblage	reference
35	1	Camperio	712.5/153.7	Piora zone/Quartenschiefer	Kl 492	MaQzCzPlCc-MuChBiHo	Klein(1976)*
36	3	Campo Blenio	715.1/156.9	Peidener Schuppenzone/ "Liassic"	Blen 1	MaQzCzPlCc-MuChGr	
37	1	Val Camadra	715.1/160.0	Scopi zone/"Liassic"	Blen 21	MaQzCc-MuPaDoGr	
38	8	Valle Cavalasca	717.5/160.1	Peidener Schuppenzone/ "Liassic"	MF 1316	MaQzCzPlCc-MuPaChDoGr	Frey(1978)
40	1	Val di Carassino	717.2/156.2	Piz Terri-Lunschania zone/ Bündnerschiefer	Blen 36	MaQzCzPlCc-MuChDoGr	
41	1	Madra	724.4/143.9	Simano nappe/pre-Triassic metapelite	Kl 484	MaQzCzPl-MuChBiGa	Klein(1976)*
42	1	P. del Ramulazz	728.2/140.5	Adula nappe/pre-Triassic metapelite	Kl 327	MaQzCzPl-MuChBiGa	Klein(1976)*
43	1	Alp de Trescolmen	733.6/139.9	Adula nappe/pre-Triassic metapelite	Kl 269	MaQzCzPl-MuChBiGa	Klein(1976)*
44	1	Portela	734.7/146.0	Misox zone/Bündnerschiefer	MF 1893	MaQzZoCc-MuPaChGaGr	
45	1	Spina	736.3/143.8	Misox zone/Bündnerschiefer	MF 1872	MaQzCc-MuPaChGr	
46	1	Giumela	735.2/141.3	Misox zone/Bündnerschiefer?	MF 1969	MaQzKy-MuPaChCtGaGr	
47	1	Mesocco	738.0/138.4	Misox zone/Bündnerschiefer	Mis 70	MaQzCzPl-MuBiGaGr	Klein(1976)*
101		Chamoson		Morcles nappe/"Aalénian"		PyCcQz...Om	Massaad(1973)**
102		Arbaz		Ultrahelvetic/"Aalénian"		PyCcQz...Om	Massaad(1973)**
103		Col du Pillon		Ultrahelvetic/"Aalénian"		PyCcQz...Om	Massaad(1973)**
104		Lauenen		Ultrahelvetic/"Aalénian"		PyCcQz...Om	Massaad(1973)**
105		Spillgerten		Prealps/paleokarst	Genge 169	PyCcDi-IlChHe	Baud et al.(1979, p.447;Baud(pers. comm.)
106	1	Ferdenpass	619.8/138.3	Parautochthonous/"Hettangian"	MF 1685	PyCcQz-IlPa/MuOmCh	
107	1	Bürgli/Kiental	626.3/154.4	Wildhorn nappe/"lower Liassic"	BK 72/253	PyCcQz-IlChDoOm	

No.	#	locality	coordinates	tectonic/stratigraphic position	original specimen	assemblage	reference
108	6	Seelitalgraben	703.3/192.7	Axen nappe/"Infralias" and Cardinia beds	MF 539	PyCcQz-IlPaPa/MuChDoOm	
109	1	1km E Ennenda	725.8/210.6	Glarus nappe/Bommerstein fm	MF 43	PyKaCcQz-IlOm	
110	1	Wissmil-Magerrain	735.8/210.3	Axen nappe/Cardinia beds	MF 637	PyCcQz-IlPaPa/MuChOm	
111	2	Schnüerligrat	739.8/207.4	Axen nappe/"Infralias", Prodkamm fm	MF 719	PyCcQz-IlPa/MuChOm	
112	2	Guggenegg	740.8/210.5	Axen nappe/Cardinia beds	MF 648	PyCcQz-IlPaPa/MuChDoOm	Frey(1970)
	1	Lauifurggla	740.3/208.6	Axen nappe/"Infralias"	MF 371	PyCcQz-IlPa/MuChOm	Frey(1969)
113	1	Ziger	737.6/214.8	Axen nappe/Prodkamm formation	MF 570	PyCcQz-IlPaPa/MuChOm	
114	4	1 km E of Mols		Mürtschen nappe/Mols formation	KB 70/1-1	PyKaCcQz-IlRePa/MuChDoOm	
115	3	Pflastertobel	750.8/214.3	Axen nappe/Mols formation	MF 1017	PyCcQz-IlPa/MuChOm	

column 1: No. 1-5 margarite-bearing assemblages
No. 6-47 margarite+quartz-bearing assemblages
No. 101-115 pyrophyllite+calcite-bearing assemblages

column 2: number of samples per locality

column 6: collection of: BK = Künzi (1975), HA = Hansen (1972), Lis = Liszkay (1965), Str = Prof. Streckeisen, WN = Prof. Nabholz, Blen,Mis,Sci = Blenio, Misox, Sciora collection of the Mineralogical Institute at Basel

column 7:*

Bi	biotite	Di	diaspore	He	hematite	Mu	muscovite	Pl	plagioclase
Cc	calcite	Do	dolomite	Ho	hornblende	Om	organic material	Py	pyrophyllite
Ch	chlorite	Ep	epidote	Il	illite	Pa	paragonite	Qz	quartz
Co	corundum	Ga	garnet	Ka	kaolinite	Pa/Mu	mixed-layer paragonite/muscovite	Re	rectorite
Ct	chloritoid	Gr	graphite	Ky	kyanite			St	staurolite
Cz	clinozoisite	Hc	hercynite	Ma	margarite	Ph	phlogopite	Zo	zoisite

column 8: * Klein (1976) reports incomplete assemblages
** Massaad (1973) gives no information regarding coordinates and complete assemblages

* If more than one sample per locality is available, then the most complete assemblages are given only, neglecting existing sub-assemblages. First quoted phases belong to the model system $CaO-Al_2O_3-SiO_2-H_2O-CO_2$.