

COMPOSITIONAL RANGE OF Mn-GARNET IN ZONED GRANITIC PEGMATITES

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ABSTRACT

A relationship has been established between garnet composition (determined by gravimetric analysis) and zones in large complex granitic pegmatites. Mn-rich garnet (MnO 41–27%) occurs in the inner and replacement zones, garnet with Mn \approx Fe in intermediate mineral assemblages and garnet with Fe > Mn in the wall and contact zones. Pure spessartine garnet is typical of replacement and quartz-rich core zones of pegmatites containing lithium minerals such as spodumene, lepidolite, petalite and amblygonite.

Keywords: spessartine–almandine garnet, classical analysis, zoned garnet, zoned lithium pegmatite, Africa.

SOMMAIRE

On établit la relation suivante entre la composition du grenat (analysé par voie humide) et la zonation de grandes pegmatites granitiques. Le grenat riche en manganèse (MnO entre 41 et 27%) se trouve près du centre et dans les zones de remplacement. Le grenat à Mn et Fe en teneurs à peu près égales se rencontre dans les assemblages intermédiaires et le grenat à Fe dominant, près des parois et dans les zones de contact. La présence de spessartine pure est typique des zones de remplacement et du centre quartzifère de pegmatites contenant des minéraux lithiques tels que spodumène, lépidolite, pétalite et amblygonite.

(Traduit par la Rédaction)

Mots-clés: grenat à spessartine et à almandin, analyse par voie humide, zonation du grenat, zonation d'une pegmatite lithique, Afrique.

INTRODUCTION

Structural units of contrasting mineralogy and texture within complex pegmatites have been referred to as border or contact zones, wall zones, intermediate zones and core zones (Cameron *et al.* 1949). The aim of this paper is to establish a relationship between the chemical composition of garnet found in pegmatites of granitic composition and the pegmatite zones. The pegmatites from which garnet has been studied occur in Africa, North America and

Finland, the most extensive sampling being from Africa (Fig. 1).

In addition to the generally recognized zones referred to above, there are several views on additional zones, usually containing replacement minerals. Of the pegmatite areas being studied, for example, Cameron *et al.* (1949) described a systematic distribution of replacement zones in the pegmatites of the Black Hills, South Dakota. The development of the replacement zones is generally controlled by fractures related to the main zonal patterns: these units are usually recognized by differences in mineralogical assemblage. For example, in the lithium-rich pegmatites, zones containing replacement minerals are developed adjacent to the quartz core. These zones are usually rich in cleavelandite and may contain discontinuous units of one or another of the lithium minerals. The cross-cutting nature of these zones suggests a late-stage crystallization, probably of replacement origin. Hornung & von Knorring (1962) preferred to consider a 'late stage' and a 'hydrothermal stage' for minerals crystallizing later than the core, because evidence of replacement is difficult to find in some pegmatites; the minerals cleavelandite and lepidolite, for example, appear in a strict zonal sequence in one part of the pegmatite but are discordant to the main structure in other parts. In the Alto Ligonha pegmatites of Mozambique, Hutchinson & Claus (1956) found minerals such as lepidolite restricted to the innermost intermediate zone, in contact with core quartz; they did not identify a replacement zone. Lowenstein (1969), on the other hand, recognized well-developed zones of primary crystallization in the pegmatites of southwestern Uganda and documented an extensive episode of late-stage replacement.

Garnet, a mineral commonly encountered in pegmatite bodies, shows great diversity in its distribution. It may crystallize throughout the pegmatite system, as a member of different mineral assemblages. Table 1 lists the minerals that are associated with the garnet specimens analyzed, and the zone of occurrence. Further details are provided by Baldwin (1979). Spessartine-rich garnet is found in complex pegmatites that contain large lithium-bearing zones rich in one or more of lepidolite, spodumene, petalite and amblygonite.

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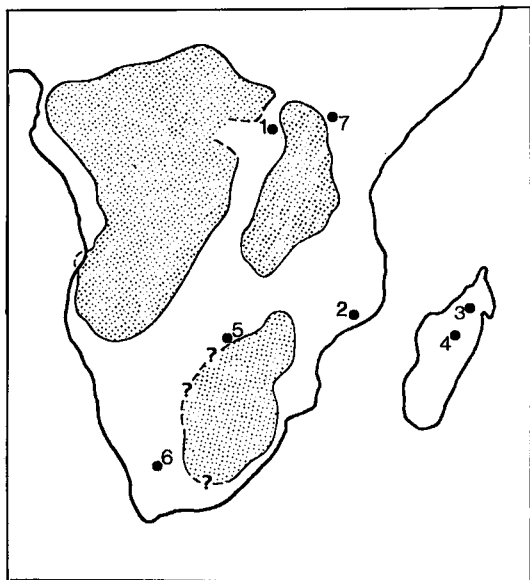


FIG. 1. Localities of African pegmatites: 1 southwest Uganda, 2 Alto Ligonha, Mozambique, 3 Berere, Tsaratanana region, Malagasy, 4 Sahatany region, Malagasy, 5 Zimbabwe, 6 Namaqualand, South Africa, 7 West Suk, Kenya. Stippled areas represent older cratons that have been stable at least since approximately 1500 Ma ago; unstippled areas represent younger orogens that have been deformed in the past 1200 Ma (from von Knorring 1970).

Cleavelandite is commonly present, as well as tantalite, microlite, tourmaline and Mn-apatite (1-14, Table 1). For example, Mn-rich garnet intergrown with microlite occurs as microscopic crystals enclosed in blue muscovite in the albite zone of the Mistress pegmatite, Salisbury, Zimbabwe (6, Table 1). In a non-lithium rare-earth pegmatite at Ntembeni, Makaha, Zimbabwe, garnet is intergrown with cyrtolite and cleavelandite in the replacement zone (16). Garnet may also be developed with core quartz; for example, garnet is intergrown with sphalerite and gahnite (20; von Knorring 1946) or with sillimanite and chrysoberyl (21) in the quartz core at Rosendal, Kimito, Finland. It is also associated with phosphate minerals (lithiophilite, strengite, metastrengite, libethenite, stewartite and apatite) at Noumas, Namaqualand (15), also with core quartz. In the Black Hills pegmatites, South Dakota, the main phosphate minerals are Fe-rich, and the main minerals intergrown with garnet in the intermediate zone are triphylite, triplite and graftonite (25, 26, 28). Rhodochrosite is also associated with the phosphate mineral assemblage at the Highland Lode pegmatite (22). Finally, garnet may also occur in the wall and contact zones, locally forming continuous bands with

black tourmaline in a sugary albite – muscovite matrix, as for example at Naipa, Mozambique (30).

GARNET IN GRANITIC PEGMATITES

Garnet has been selected from different zones of lithium-rich pegmatites, mainly, but garnet from some lithium-deficient pegmatites has also been included. The hand-picked garnet grains were analyzed for major elements by classical gravimetric techniques. The data for 31 specimens are presented in Table 1. Selected specimens were also studied with the electron microprobe to investigate compositional zonation; analytical details are given in the section on zoning. These data are not used in Table 1 or Figure 2.

The major variation in these almandine- and spessartine-rich garnet compositions involves iron and manganese. The $Mn/(Mn + Fe^{2+})$ molar ratio varies from 0.98 (Sebit) to 0.32 (Muiane). CaO is less than 1% (wt.) in all but five of the specimens, whereas MgO is less than 0.10% in all but those from Sahatany Valley, Malagasy (19), Naipa, Mozambique (30) and Nyakabingo, Uganda (18), which contain 0.77%, 0.48% and 0.47% MgO, respectively.

The specific occurrence of varieties of garnet in particular geological environments has been known since the 1930s; for example, garnet composed of spessartine and almandine end-members constitute 80-95% of all occurrences from pegmatite and granite. As a result of the present study, two distinct relationships can be recognized within the pegmatite field: (1) spessartine occurs in pegmatites that contain important lithium phases; (2) spessartine-almandine prevails where the lithium content is very low. In the garnet specimens from lithium pegmatites, the spessartine end-member ranges from 96.17 to 68.49% in the core and replacement zones; in the corresponding zones in the lithium-deficient pegmatites, the garnet contains between 72.35 and 65.35% spessartine (Table 1). These figures, therefore, suggest that in this particular geological environment, pure spessartine garnet is formed only in lithium pegmatites.

The composition of garnet varies according to its position within the pegmatite body. There is a distinct tendency for the late-formed garnet, in replacement zones and zones surrounding the core, to be richer in manganese and poorer in iron. This is clearly illustrated in Table 1 by the $Mn/(Mn + Fe^{2+})$ ratio. Among the group of lithium pegmatites, all specimens with a high $Mn/(Mn + Fe^{2+})$ ratio, between 0.69 at Noumas and 0.98 at Sebit, are either from the late replacement zone (R) or the outer core zone (C), with the exception of the garnet from Tsarafara, Malagasy (5), which crystallized in the wall zone of a small pegmatite vein. Conversely, there is a distinct tenden-

TABLE 1. GARNET IN PEGMATITES

No.	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	FeO	MnO	CaO	Fe ²⁺	Mn ²⁺	Atomic Mn Mn+Fe ²⁺	Spess %	Zone	Associated Minerals	Pegmatite
1	36.20	tr	20.40	0.15	0.80	41.21	0.93	0.44	23.08	0.98	96.17	R	cl-lep	West Suk, Kenya
2	36.35	0.03	20.60	0.10	2.01	40.28	0.61	1.11	22.47	0.95	93.59	R	ab-li/m	Muiane, Mozambique
3	36.63	0.06	20.95	-	1.63*	39.35	0.72	0.90	21.94	0.96	93.96	R	ab-mic-mus	Jordywitz, Zimbabwe
4	36.26	0.04	20.65	0.42	2.86	39.10	0.70	1.60	21.79	0.93	91.37	C	q-pt-sp-Mn/ap	Casa Ventura, Zimbabwe
5	36.33	0.08	20.63	0.00	3.18	38.97	0.99	1.75	21.70	0.92	90.61	W	li/m-q-tm	Tsarafara, Malagasy
6	38.11	tr	20.78	-	0.79*	38.15	1.32	0.43	21.05	0.98	93.97	R	ab-mus-mil-q	Mistress, Zimbabwe
7	36.85	0.07	20.41	0.00	3.38	37.56	0.53	1.87	21.04	0.92	90.34	R	cl-li/m-q	Lunya, Buganda, Uganda
8	36.63	0.07	21.08	0.27	2.00	37.25	2.36	1.09	20.65	0.94	88.29	R	cl-Mn/tan-mus	Mutaka, S.W. Uganda
9	36.48	tr	20.41	0.23	2.88	37.07	2.16	1.59	20.72	0.93	86.91	R	cl-mus	Rwemero, Zimbabwe
10	36.36	tr	20.36	0.50	5.60	36.65	0.56	3.09	20.46	0.86	85.43	R	ab	Rwemero, Zimbabwe
11	36.09	0.08	20.60	0.00	7.04	35.06	1.02	3.89	19.60	0.83	82.21	-	-	Sahatany V., Malagasy
12	36.34	0.10	20.25	0.26	7.89	34.51	0.44	4.36	19.32	0.81	80.74	C	q-cl-mus	Benson 4, Zimbabwe
13	36.25	0.08	20.64	0.78	7.78	33.88	0.54	4.28	18.90	0.81	80.22	R	cl-Mn/ap-mus	Maridge, Mozambique
14	36.02	0.07	20.49	0.93	8.52	33.77	0.45	4.70	18.85	0.79	78.61	R	ab	Nuaparra, Mozambique
15	36.14	0.12	20.05	0.76	9.70	32.50	0.36	5.38	18.24	0.79	76.42	C	ap-st-lib-lph	Noumas, Namaqualand
16	36.74	0.06	20.90	-	11.21*	30.07	0.34	6.17	16.77	0.73	72.35	R**	cl-cy-q-R.E.M.	Ntembeni, Zimbabwe
17	36.63	tr	20.79	-	13.19*	29.18	0.32	7.25	16.24	0.69	68.49	R	cl-q-sp	Noumas, Namaqualand
18	36.47	0.03	19.69	1.71	13.54	28.21	0.45	7.44	15.69	0.67	65.51	R**	cl-q-mus	Nyakabingo, S.W. Uganda
19	36.31	0.42	19.71	2.36	9.91	27.80	3.48	5.43	15.44	0.72	64.33	-	mag	Sahatany V., Malagasy
20	37.05	0.10	20.24	-	14.37*	27.21	0.24	7.93	15.22	0.66	65.27	C**	q-sph-gahn	Rosendal, Finland
21	36.26	0.03	20.42	0.58	14.23	27.12	0.26	7.91	15.26	0.65	65.35	C**	q-sill-chryb	Rosendal, Finland
22	36.23	-	20.63	-	19.70*	23.32	0.15	10.88	13.04	0.55	54.53	I	rh-tp1-Mn/ap	Highland Lode, S.D.
23	-	-	-	-	20.26*	22.45	-	-	-	0.53	-	W	ab	Noumas, Namaqualand
24	36.55	tr	20.33	0.37	20.52	21.61	0.48	11.32	12.08	0.51	50.88	CON	q-mus	Inrulle, Mozambique
25	36.41	0.07	20.50	0.82	20.63	20.95	0.34	11.39	11.71	0.50	50.17	I	gr-tp1-tp1	Highland Lode, S.D.
26	35.79	0.03	20.07	-	22.74*	20.85	0.15	12.68	11.78	0.48	47.94	I	tp1-q-viv	White Elephant, S.D.
27	36.74	0.07	20.06	0.75	21.19	20.63	0.33	11.70	11.53	0.49	49.15	I**	q-p	Berere, Malagasy
28	36.08	0.03	19.98	-	25.06*	18.12	0.15	13.97	10.23	0.42	42.08	I	tp1-tp1	Gap Lode, S.D.
29	36.85	0.04	20.77	0.87	25.83	15.24	0.35	14.17	8.47	0.38	36.34	CON	mus-ab-q	Muiane, Mozambique
30	36.69	0.05	20.85	0.37	25.79	15.06	0.31	14.20	8.37	0.37	36.01	CON	ab-mus-q-tm	Naipa, Mozambique
31	36.57	0.04	20.75	1.85	27.89	12.76	0.37	15.28	7.08	0.32	30.08	CON	mus-ab-q	Muiane, Mozambique

Abbreviations: S.D., South Dakota, U.S.A.; R, replacement; C, Core; I, intermediate; W, Wall; CON, contact; Spess %, spessartine end-member %; ab, albite; chryb, chrysoberyl; cl, cleavelandite; cy, cyrtolite; gahn, gehnite; gr, graffonite; lep, lepidolite; li/m, lithian mica; lib, libethenite; lph, lithiophilite; mag, magnetite; mic, microcline; mil, microcline; Mn/ap, Mn apatite; Mn/tan, Mn tanalite; mus, muscovite; q, quartz; R.E.M., rare-earth minerals; rh, rhodochrosite; p, perthite; pt, petalite; sill, sillimanite; sph, sphalerite; sp, spodumene; st, stengite; tm, tourmaline; tph, triphylite; tp1, triplite; viv, vivianite. Analyst: J.R. Baldwin; 12 & 20, O. von Knorring. Fe²⁺ & Mn²⁺, numbers of ions on the basis of 96 (O). MgO, trace with the exception of 19, 30 & 18 with 0.77%, 0.48% & 0.47%, respectively. * total iron as FeO; ** non-lithium pegmatite.

cy for the earliest-formed garnet, in contact and wall zones, to be poorer in Mn and richer in Fe. This is also illustrated in the Noumas pegmatite, Namaqualand, where the analysis of garnet from four different mineralogical zones yielded the following results: 36.85% MnO in one core, 32.50% in a separate core, 29.18% in the replacement zone and 22.45% in the wall. Similarly, in the Rosendal pegmatite, garnet contains 29.10% MnO in the replacement zone, 27.21% in the core and 23.08% in the intermediate zone. The general pattern of manganese concentration in garnet relative to the pegmatite zones is clearly illustrated in Figure 2, which shows a systematic increase in Mn in relation to the general sequence of mineral assemblages from contact through to core and replacement zones.

ZONING IN GARNET

Almandine-spessartine garnet from pegmatite and

aplite has been shown to be zoned to varying degrees (Foord 1976). Previous studies on compositional zoning within grains of garnet from aplite and granite have been made by Leake (1967) and Bizouard *et al.* (1970). Foord (1976) documented three major trends: an Fe-enrichment trend (core to rim), an Mn-enrichment trend and, more rarely, an Mn-enrichment trend followed by Fe enrichment. Two other possible trends (no zonation detectable and Fe enrichment followed by Mn enrichment) are also found (Foord 1976), but in terms of frequency of occurrence, they are relatively minor.

The electron microprobe was applied to investigate zoning in several samples of almandine-spessartine garnet in this study. The microanalyzer used was a Cambridge Instruments Microscan 5, with a gun potential of 20 kV and a beam current, measured using a Faraday cage, of 30 nA for crystal spectrometry and 60 nA for energy-dispersion spectrometry. A full ZAF-correction procedure, as

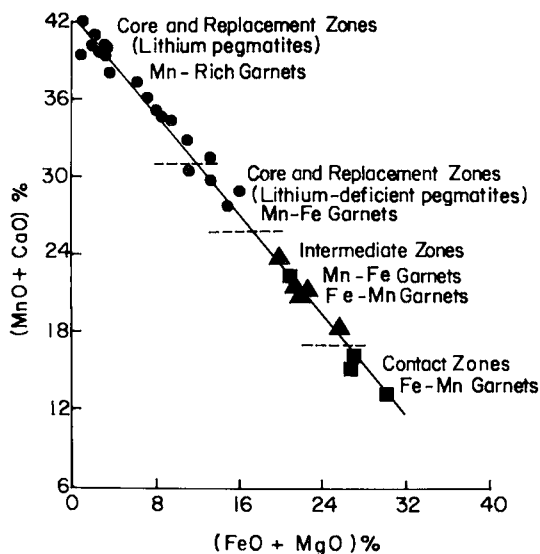


FIG. 2. Relationship between the chemical composition of garnet and pegmatite zones (see Table 1 for chemical data).

described by Sweatman & Long (1969), was used to calculate final concentrations.

Zoning in almandine-spessartine crystals from Muiane, Mozambique, was investigated by means of crystal-spectrometry techniques, in which the 95% confidence limit ($\pm 2\sigma$) for MnO and FeO, up to the 20 wt.% level, is ± 0.3 wt.%. In the contact zone of the pegmatite, trapezohedral garnet crystals occur in a feldspar-muscovite-quartz matrix. A microprobe traverse 60 mm across the largest and freshest crystal showed a simple symmetrical pattern of zoning from core maximum (19.1% MnO) to rim minimum (11.9% MnO) (Fig. 3), *i.e.*, a Mn-depletion trend with a corresponding enrichment in Fe; CaO is constant across the crystal. Microprobe traverses for two other garnet crystals (35 and 29 mm in diameter) from the same locality revealed the same pattern of zoning. This symmetrical pattern of zoning is a result of continuous rather than discontinuous growth. With regard to the variation from 30 mm, the slightly lower trend may be due to the fact that the traverse was taken across two sections that may not have been matched up exactly. However, in the two other analyzed crystals a similar variation in composition also exists around the core, and the complete traverse was taken across one section. It is therefore suggested that even within experimental error, this variation may be real, but perhaps the centre of the garnet grains should be examined more closely.

With respect to the other almandine-spessartine

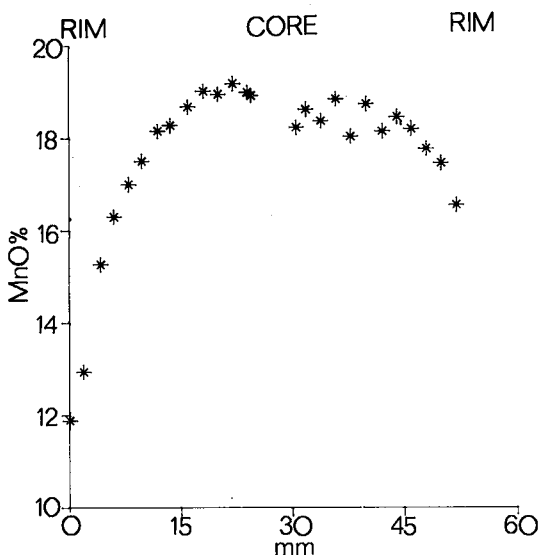


FIG. 3. The pattern of manganese concentration in a zoned garnet from the Muiane pegmatite, Mozambique. (Analyst P. Hill; analytical error, $\pm 2\sigma = \pm 0.3$ wt.% for each data point. The actual rim at 60 mm is not shown).

samples, a microprobe traverse across a 25-cm garnet from Inrulle (24, Table 1) did not reveal any compositional zoning; the MnO had a standard deviation of 0.14, which is within the experimental error for analysis by energy dispersion: $\pm 2\sigma = \pm 0.3$ wt.%. Two samples of garnet 2 mm in diameter from the wall zone at Noumas (23) reveal no compositional zoning. A third shows an increase in MnO from 23.4 to 25% from the rim to the core.

Among the Black Hills specimens, the garnet from White Elephant (26, Table 1) shows no compositional zoning. The Highland Lode garnet (22) shows a range in MnO content from 22.4 to 24.2%; whereas one very large crystal shows no variation in composition, another crystal 500 μm in diameter has a content of 23.3% MnO in the core and 22.4% in the rim, *i.e.*, an enrichment of Fe from core to rim, with a constant value for CaO. However, the South Dakota samples, on the whole, are intergrown with other phases, making it difficult to identify a core zone. Crystal-spectrometry techniques were used for these analyses.

In the spessartine-rich garnet compositions from the core and replacement zones, zoning was investigated in specimens from two localities. A 19-mm crystal from Mutaka (8, Table 1) contains a Ca-rich core. In the whole of the garnet, the MnO varies between 41.0 and 41.5%, which is within the experimental error of the energy-dispersion method of

analysis, with the exception of a 2-mm-wide area in the centre, which contains an average of 38.4% MnO. The garnet contains 2.4% CaO at the centre and 0.6% elsewhere, but the Fe content is uniform throughout the traverse across the crystal. This, therefore, signifies a depletion in Mn and an enrichment in Ca in the core. The spessartine garnet from Mistress (6) occurs as microscopic crystals, 0.25 mm to less than 50 μm in diameter, enclosed in blue muscovite. Microprobe analyses of three different crystals gave the same result, 43.1% MnO and 1.0% CaO. However, another crystal 50 μm in diameter contains 39.5% MnO and 4.1% CaO.

A comparison of electron-probe and gravimetric data indicates that the results from the latter fall within the range measured on the probe. There is, however, a discrepancy between the two methods in specimen 6, which is due to the slight impurity in these minute, 0.25-mm-to-50- μm crystals, hence the anomalously high value of 38.11% SiO_2 (Table 1) obtained by the classical technique.

DISCUSSION

Most granitic pegmatites are simple in their mineral parageneses. Garnet, where present, is predominantly almandine. The data presented here are largely concerned with a particular type of pegmatite characterized by the presence of large lithium-bearing zones (Baldwin 1979). It is in these lithium pegmatites, which are generally complex in their mineral parageneses, that spessartine garnet is prevalent. The association between Mn-rich garnet and lithium pegmatites is emphasized in Figure 2. Mn-rich garnet occurs in the core and replacement zones of lithium pegmatites, whereas Mn-Fe garnet is found in the core and replacement zones of lithium-deficient pegmatites: Mn-Fe and Fe-Mn garnet compositions occur in intermediate assemblages, and Fe-Mn garnet in contact zones.

The range of thermal stability of almandine and spessartine and the solidus and liquidus for the Harding pegmatite, New Mexico, are shown in Figure 4. Spessartine and almandine garnets are stable throughout the temperature range of granite-pegmatite consolidation. Divalent manganese is concentrated at the late stage of crystallization of pegmatite owing to its large ionic radius relative to that of the other divalent elements of the Mn-Fe group. Moreover, magmatic evolution is characterized by the increasing proportions of Mn^{2+} to Fe^{2+} , and this is exemplified in the sequential crystallization of various generations of pegmatite minerals. Almandine-spessartine garnet crystallizes early in the system in the wall and contact zones; in comparison, spessartine garnet crystallizes later in the system, either at the edge of the core or in the replacement zones. This is in contrast to a zoned garnet from the

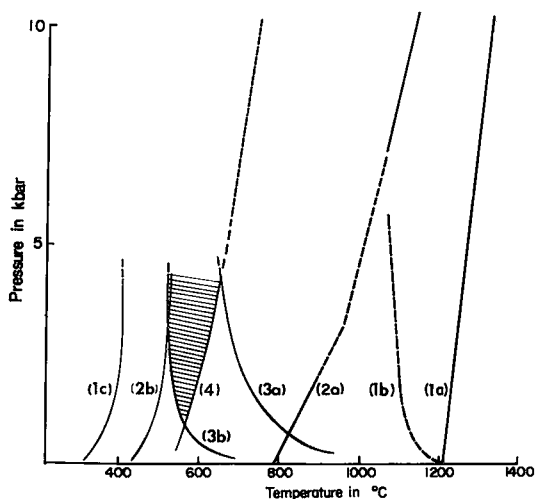


FIG. 4. Pressure-temperature conditions governing the formation of garnet in pegmatites. (1a) Anhydrous melting curve of spessartine (Mottana 1974). (1b) Hypothetical melting curve of spessartine, water saturated (Hsu 1968). (1c) Manganese chlorite + quartz = spessartine + fluid (Hsu 1968). (2a) Melting curve of almandine, water saturated (Keesmann *et al.* 1971). (2b) Iron chlorite + quartz = almandine + fluid (IQF buffer) (Hsu 1968). (3a) Pegmatite liquidus, water saturated (Jahns 1982). (3b) Pegmatite solidus, water saturated (Jahns 1982). (4) Breakdown of muscovite + quartz for $P(\text{H}_2\text{O}) = P_{\text{total}}$ (Kerrick 1972). Ruled area is the field of stability of a granitic pegmatite containing garnet + muscovite + quartz.

contact zone in the Muiane pegmatite, Mozambique, where the early compositions of garnet are Mn-rich and the late ones Mn-poor.

As all the pegmatites dealt with in this study contain muscovite in association with garnet, the P-T conditions showing the breakdown of muscovite (+ quartz) are presented in Figure 4 (curve 4). The pegmatite liquidus and solidus are represented by curves 3a and 3b, respectively. The hydrous conditions necessary for the crystallization of muscovite-quartz pegmatites from an evolved magma lie approximately at a temperature of 580°C at 1 kbar and between 550 and 650°C at 4 kbar (the shaded area between curves 3b and 4). The shaded area in Figure 4 also represents the field of formation of pegmatite containing garnet + muscovite + quartz. These temperatures are not only in agreement with Jahns (1982), who suggested that compositionally complex pegmatites with substantial amounts of B, F and Li could well have had crystallization temperatures extending down to 550°C, but are also consistent with a minimum temperature of 550°C deduced for the formation of garnet in the core zone of the Rosen-

dal pegmatite, Finland (21, Table 1) based on the presence of intergrown sillimanite. Mueller & Saxena (1977) gave a minimum temperature of 550°C for the formation of sillimanite (at the Al_2SiO_5 "triple" point).

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