Calderite-rich garnets from metamorphosed manganese silicate rocks of the Sausar Group, India, and their derivation

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Abstract

Manganiferous garnets occur in metamorphosed Mn silicate–oxide and silicate–carbonate–oxide rocks of the Sausar Group, India. The garnets of the carbonatic rock show maximum calderite content (~50 mole%). Increased Ca content in the bulk has been observed to be sympathetically related to the concentration of calderite, rather than the expected andradite component of the garnets. This is the consequence of the preferential partitioning of Ca in coexisting pyroxmangite and/or carbonate. Petrochemical characteristics of the diverse assemblages in response to the physical conditions of metamorphism show that the calderite solubility in manganiferous garnet is not only a function of pressure. Such characteristics only indicate that the mobile components in the bulk influenced the mineralogy of the assemblages at the ambient physical conditions of metamorphism, and this in turn controlled the calderite solubility in garnet.

KEYWORDS: calderite, garnet, manganese, Sausar Group, India.

Introduction

THE name calderite was originally suggested for a 'silico–iron–manganese rock' described from India by Piddington (1850; cited by Fermor, 1909) for which a chemical analysis was given. Fermor recalculated Piddington's analysis assuming that the mineral components were garnet and quartz, and suggested that this garnet might have a composition of $3MnO . Fe_2O_3 . 3SiO_2$ and could itself be called calderite. Thus the name calderite passed from that of a rock to that of a garnet species based on an analysis recalculated from that of a rock of assumed mineral assemblage (garnet and quartz; cf. Fermor, 1909, p. 184). Thus, Fermor's (1909) calderite itself could deviate from the end-member composition suggested for it and the validity of the name also remained questionable for a long time. De Villiers (1951) and Vermaas (1952) noted the second occurrence of calderite-bearing garnet from Otjosondu, Namibia, and the same sample was re-examined by Dunn (1979). Calderite-bearing garnets were also described by Sastri (1963; Tirodi, India), Klein (1966; Wabush iron-formation, Labrador, Canada), Schreyer and Abraham (1977; Brezovica, Yugoslavia) and Muir Wood (1982; Panoche Pass, California). In most of these cases, the calderite content of the garnets analysed varied from 15 to 36%. Only those of the Wabush iron-formation showed a high calderite-content (57% and 69%), but for these garnets the Al_2O_3 content was not reported. Thus, unequivocal occurrence of garnets with a calderite content approaching the ideal end-member composition ($Mn_3^{2+}Fe_2^{3+}Si_3O_{12}$) are as yet unknown from natural deposits. The ideal end-member calderite was synthesized by Lattard and Schreyer (1983) at pressures above 22 kbar under a hematite–magnetite buffer and it was shown to be stable only under the very high pressure and low temperature conditions appropriate for subduction zone metamorphism.

As a part of our ongoing project on geological and petro–mineralogical re-examination of the manganese-ore-bearing Precambrian Sausar Group, India, study of metamorphosed manga-

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nese silicate rocks revealed the occurrence of gar-
nets commonly containing variable but significant
calderite components. This study aims to establish
the compositional characteristics of such garnets
in metamorphosed manganese-rich sediments in
relation to the ambient conditions of metamor-
phism.

Geological Background

The Proterozoic (sed. age *c*. 2000 Ma; met. age
c. 900 Ma) Sausar Group of rocks in central India
host major manganese deposits (Roy, 1966;
1981). The manganese-rich formations occur as
the following three lithotypes: (*a*) Mn oxide rocks
(ore); (*b*) Mn silicate–oxide rocks; and (*c*) Mn
silicate-carbonate-oxide rocks. The Mn silicate-
carbonate-oxide rocks have only recently been
identified from the Sausar Group (Roy *et al.*,
1986). The two types of silicate rocks are closely
associated in space and time but are never found
in physical contact. The Mn silicate rocks are
often internally laminated showing contrasting
mineral assemblages.

The calderite-rich garnets have been studied at
Chikla (Bhandara District, Maharashtra) and Tir-
odi and Netra (Balaghat District, Madhya Pra-
desh). In all these areas, the manganese-rich
formations occur as stratified bodies conformably
enclosed in pelitic rocks that were co-metamor-
phosed to amphibolite facies (Roy, 1966). Gar-
net–biotite thermometry in metapelites estimates
temperature of metamorphism at $600 \pm 20\,°C$ for
Chikla and $650 \pm 25\,°C$ for Tirodi and Netra areas
at adopted pressure of 6 kbar. The estimates are
consistent with the development of kyanite and
sillimanite respectively. The metapelites record
a Barrovian type of metamorphism with the deve-
lopment of garnet, staurolite, kyanite and sillima-
nite (Dasgupta *et al.*, 1984).

Garnetiferous assemblages

Both types of Mn silicate rocks show diverse
mineral assemblages. The garnet-bearing assemb-
lages relevant to the present study are listed
below. Each assemblage is confined to a single
band/lamina in the respective rock and the phases
in physical contact with garnet are shown with
plus sign.

(i) Garnet + quartz ± hematite ± alkali felds-
par; (ii) Garnet + braunite ± hematite ± alkali
feldspar; (iii) Garnet + jacobsite + pyroxmangite
± quartz ± hematite ± tirodite ± alkali feldspar
(only at Tirodi and Netra); (iv) Garnet + braunite
+ pyroxmangite + rhodochrosite + jacobsite ±
quartz ± hematite; and (v) Garnet + braunite

+ rhodochrosite ± jacobsite ± hausmannite ±
hematite. Assemblages (i), (ii) and (iii) belong
to the Mn-silicate–oxide rocks while (iv) and (v)
are present in the Mn-silicate–carbonate–oxide
rocks.

In assemblage (iv) the garnet is dark yellow
in colour and is distinctly anisotropic in shades
of grey. In assemblage (v) the garnet is again
faintly anisotropic. However, the garnets in
assemblages (i), (ii) and (iii) are light yellow and
isotropic. The garnets are porphyroblastic and
show equilibrated grain boundary textures with
the associated oxides, silicates and carbonates.
The strongly anisotropic garnets, which also show
high calderite contents (see later), are invariably
associated with pyroxmangite with a sharp mutual
boundary. In assemblage (i) the isotropic garnets
occur as equant grains in a mosaic of polygonized
quartz. Optical and X-ray data reveal that the
Fe oxide in all the assemblages is present as hema-
tite.

Compositional characteristics

Powder X-ray diffraction profiles of concen-
trated fractions or separated garnets from assemb-
lages (i) and (iv) were determined. The calculated
cell constants of isotropic garnet [assemblage (i)]
is $a = 11.65(\pm 0.02)$Å, while that of anisotropic
garnet [assemblage (iv)] is $a = 11.721(\pm 0.03)$Å.
While the former is comparable with that of spes-
sartine as per the ASTM data file, the latter
deviates considerably from that. The available cell
constants of calderite ($11.81–11.82$Å; cf. Lattard
and Schreyer, 1983) are of nearly pure end-
member composition. The calculated cell con-
stant of anisotropic garnet from assembalage (iv)
thus represents an intermediate member of the
spessartine–calderite solid solution.

The chemical compositions of garnets and the
associated phases in physical contact were deter-
mined using a JEOL JXA–5A EPMA. The oper-
ating conditions are given in our earlier
publication (Bhattacharya *et al.*, 1984) and the
compositions are given in Tables 1–4. The total
iron in garnets has been calculated as Fe^{3+} since
hematite, in total exclusion of magnetite, is ubi-
quitous in the assemblages. A small part of the
total Mn, calculated as Mn^{2+}, was converted to
Mn^{3+} to fill the remaining vacancy in the octahed-
ral sites. Entry of Mn as Mn^{3+} in garnet was
debated on crystallochemical basis (Strens, 1965).
However, Nishiziwa and Koizumi (1975) success-
fully synthesized $Ca_3Mn_2Si_3O_{12}$ garnet and Fren-
trup and Langer (1981) described garnet$_{ss}$ with
up to 40 mole % of this component. This shows

Table 1. Electron probe microanalyses of garnets from manganese silicate rocks.

Sample No.	TG2	TG1	TG3	CG1	CG2	NG1	TG8	TG4(I)	CG(LI)
Assemblage	(i)	(ii)	(iii)	(iv)	(iv)	(iv)	(iv)	(v)	(v)
SiO_2	37.41	36.96	35.35	34.42	33.68	33.58	34.09	35.35	35.57
TiO_2	0.30	0.04	0.20	0.02	0.39	0.10	0.06	0.18	0.15
Al_2O_3	19.32	19.93	17.21	8.18	9.28	8.69	10.04	14.51	14.46
Fe_2O_3	2.14	0.42	4.38	17.07	15.57	16.19	15.14	8.29	8.45
Mn_2O_3	0.72	0.37	0.06	0.70	1.28	1.48	0.83	0.66	0.68
MnO	34.48	38.91	39.20	37.24	37.34	37.93	38.33	35.60	34.75
MgO	3.37	0.77	0.08	-	0.13	0.10	1.10	0.25	0.22
CaO	3.09	2.37	2.00	2.74	2.56	1.92	2.04	4.74	5.59
Total	100.83	99.77	98.48	100.37	100.23	99.99	100.63	99.58	99.87
On the basis of (O)=12									
Si	2.989	3.022	2.989	2.995	2.93	2.942	2.946	2.98	2.985
Ti	0.018	0.002	0.013	0.002	0.026	0.007	0.004	0.012	0.01
Al	1.819	1.921	1.715	0.839	0.951	0.897	1.022	1.442	1.43
Fe^3	0.129	0.026	0.278	1.118	1.019	1.067	0.984	0.526	0.533
Mn^3	0.044	0.023	0.004	0.047	0.085	0.098	0.055	0.043	0.043
Mn^2	2.333	2.695	2.808	2.745	2.751	2.814	2.805	2.542	2.47
Mg	0.401	0.094	0.01	-	0.017	0.013	0.013	0.031	0.028
Ca	0.264	0.208	0.181	0.256	0.238	0.180	0.189	0.428	0.503
Spessartine	78.0	89.9	85.8	42.0	44.3	41.4	47.7	70.5	70.2
Pyrope	13.3	3.1	-	-	0.5	0.4	0.6	1.1	0.8
Andradite	8.7	2.4	6.1	8.0	8.0	6.0	6.2	14.2	16.7
Grossular	-	4.6	-	-	-	-	-	-	-
Calderite	-	-	8.1	50.0	47.2	52.0	45.5	14.2	12.3

TG Tirodi   NG Netra   CG Chikla

that Mn^{3+} can enter the garnet structure. In distributing Ca, preference was given to the andradite component over grossular because of the presence of Fe^{3+}. Only in assemblage (ii) the Ca/Fe^{3+} ratio is high enough to allow calculation of grossular in addition to andradite. Following this scheme, the end-member components of garnet were calculated and are shown in Table 1.

Amongst the non-carbonatic assemblages, a minor calderite component appears only in assemblage (iii). Excepting in assemblage (i), all the garnets only contain a low to insignificant pyrope component. These are predominantly spessartine-rich garnets with 6–8 mole % of the andradite and grossular end member. In the carbonate-bearing assemblages (iv) and (v) garnets are notably enriched in Fe_2O_3 and depleted in Al_2O_3 as compared to their counterparts in non-carbonatic assemblages. In the pyroxmangite-free assemblage (v) garnets are significantly enriched in CaO and contain 12–14 mole % of the calderite

component. The calderite content of garnets of pyroxmangite–bearing assemblage (iv) ranges between 45 and 52%.

Pyroxmangite in assemblages (iii) and (iv) contains 10–12% of the $CaSiO_3$ component while rhodochrosite in the same assemblages takes up 7–11% of calcite (Tables 2 and 3). Braunite in assemblages (ii), (iv) and (v) and jacobsite in assemblage (iii) contain appreciable amount of Fe_2O_3 in solid solution (Table 4).

Bulk chemical analysis of representative samples of the different assemblages, determined by wet chemical method, are presented in Table 5, along with the modal abundance of the phases. Barring alkali felspar, generally a subordinate phase, garnet acted as the sink for Al_2O_3 in the different assemblages. The carbonatic rocks are notably poor in silica and enriched in CaO and Fe_2O_3 compared to the non-carbonatic ones. The CaO/MnO ratio is the highest for assemblage (iv) and decreases in the order (iii)–(v)–(i). In Fig.

Table 2. Electron probe microanalyses of pyroxmangite from manganese
          silicate rocks.

Sample No.	TG3	CG1	CG2	NG1	TG8
Assemblage	(iii)	(iv)	(iv)	(iv)	(iv)
SiO_2	47.45	45.79	46.08	47.19	47.80
TiO_2	-	0.03	0.01	0.02	0.04
Al_2O_3	0.04	0.04	0.05	0.04	-
FeO	0.92	0.22	0.10	0.02	0.15
MnO	46.48	46.55	46.05	43.88	44.43
MgO	2.19	1.81	2.09	2.78	3.18
CaO	5.06	4.95	5.24	5.48	4.40
Total	102.14	99.39	99.62	99.41	100.00

On the basis of (0)=3

Si	0.990	0.986	0.987	0.999	1.004
Ti	-	0.001	-	-	0.001
Al	0.001	0.001	0.001	0.001	-
Fe	0.016	0.004	0.002	-	0.003
Mn	0.821	0.849	0.835	0.787	0.790
Mg	0.068	0.058	0.067	0.088	0.100
Ca	0.113	0.114	0.120	0.124	0.099

Total Fe as FeO and total Mn as MnO

Table 3. Electron probe microanalyses of rhodochrosite from manganese
          silicate rocks.

Sample No.	CG1	CG2	NG1	TG8	TG4	CG2(1)
Assemblage	(iv)	(iv)	(iv)	(iv)	(v)	(v)
FeO	0.51	0.07	-	0.52	0.04	0.10
MnO	53.73	54.10	55.02	53.77	52.34	53.52
MgO	0.53	0.43	1.30	0.36	0.43	0.68
CaO	6.20	5.24	4.21	4.73	6.8	5.38
(CO_2)	39.09	38.19	38.86	37.78	38.30	38.23
Total	100.06	98.03	99.39	97.16	97.91	97.91

Fe	0.008	-	-	0.008	0.001	0.002
Mn	0.853	0.879	0.878	0.883	0.848	0.869
Mg	0.015	0.012	0.037	0.01	0.012	0.019
Ca	0.125	0.108	0.085	0.098	0.139	0.110

Total Fe as FeO and total Mn as MnO

Table 4. Electron probe microanalyses of braunite and Jacobsite from manganese silicate rocks.

	Braunite							Jacobsite
Sample No.	TG1	CG1	CG2	NG1	TG8	TG4	CG2(1)	TG3
Assemblage	(ii)	(iv)	(iv)	(iv)	(iv)	(v)	(v)	(iii)
SiO_2	9.61	10.05	10.16	10.23	9.94	9.74	9.42	0.05
TiO_2	0.11	0.10	0.12	-	0.04	0.18	0.23	0.22
Al_2O_3	0.15	0.21	0.10	0.44	0.24	0.12	0.15	0.42
Fe_2O_3	16.16	9.15	8.83	9.09	7.78	12.89	10.74	51.42
Mn_2O_3	62.22	68.53	69.33	69.77	69.46	64.35	67.49	17.18
MnO	11.31	11.83	12.07	6.28	11.72	11.44	11.23	30.60
MgO	0.02	0.03	-	0.95	0.03	0.02	-	0.39
CaO	0.08	0.07	0.03	0.43	-	0.15	0.08	0.07
Total	99.66	99.97	100.64	97.19	99.21	98.89	99.34	100.35

On the basis of (O)=12

Si	0.972	1.011	1.016	1.011	1.007	0.992	0.056	0.002
Ti	0.009	0.008	0.009	-	0.003	0.014	0.018	0.006
Al	0.018	0.025	0.012	0.051	0.029	0.015	0.018	0.019
Fe^3	1.23	0.692	0.664	0.676	0.593	0.987	0.82	1.468
Mn^3	4.791	5.246	5.275	5.25	5.358	4.987	5.214	0.496
Mn^2	0.969	1.007	1.022	0.826	1.006	0.986	0.965	0.983
Mg	0.003	0.004	-	0.035	0.004	0.003	-	0.022
Ca	0.009	0.007	0.003	0.046	-	0.017	0.009	0.003

Total Fe as Fe_2O_3. $Si+Ti = Mn^2+Mg+Ca$

1 the bulk compositions of the different assemblages and the composition of the garnets are plotted in terms of their $CaO/MnO-Fe_2O_3/Al_2O_3$ ratios. It shows that the Fe_2O_3/Al_2O_3 ratios in the garnets can be correlated with the bulk compositions for assemblages (i), (iv) and (v). This further indicates that the available Al_2O_3 was mainly accommodated in garnets, the Fe_2O_3 content of which was largely buffered by the closely associated phases during the metamorphic reactions. On the other hand, in assemblage (iii), the Fe_2O_3/Al_2O_3 ratio in the garnet is lower than that in the bulk. This is related to the higher modal abundances of jacobsite. There is no appreciable correlation between the CaO/MnO ratios of garnets and the respective bulks. As discussed later, this was influenced by stabilization of other Ca-bearing phases in the assemblages during metamorphism.

Discussion

The foregoing analysis demonstrates that garnets in these manganese-rich rocks are primarily spessartines with variable amounts of calderite along with minor andradite in solid solution. Stability of such garnets, even with high (50 mole %) calderite content contrasts with that of the end member calderite synthesized by Lattard and Schreyer (1983). In the Barrovian type of metamorphism in this area, the presence of calderite-bearing spessartine and that too with variable calderite component in closely associated occurrences indicate that calderite solubility in spessartine is not particularly influenced by total pressure alone. This is also reflected in the development of similar solid solution phases in rocks of different metamorphic facies (Table 1 in Lattard and Schreyer, 1983).

Lattard and Schreyer (1983) also stated that the presence of Ca in the bulk may be responsible for the formation of manganiferous garnets with high calderite content in amphibolite and granulite facies rocks. Our analyses of the cation contents of garnets and those in the corresponding whole rocks indicate that calderite-rich garnets are indeed restricted to Ca-rich bulks. But such garnets, when present with other Ca-bearing phases such as primarily pyroxmangite and/or

Table 5. Bulk chemical composition of different assemblages in manganese silicate rocks along with modal abundance of the phases.

Sample No.	TG2	TG3	NG1	TG4
Assemblage	(i)	(iii)	(iv)	(v)
SiO_2	56.4	47.2	35.1	34.9
Al_2O_3	15.5	12.3	11.12	24.37
TiO_2	0.2	0.5	0.36	0.4
Fe_2O_3	4.6	8.5	20.15	10.03
MnO	20.2	17.3	19.96	16.77
CaO	0.57	4.4	9.14	6.25
MgO	0.64	3.16	1.87	0.94
(CO_2)	-	-	2.06	3.24
Na_2O	0.63	2.0	0.94	n.d.
K_2O	0.41	2.8	0.16	n.d.
Total	99.15	98.16	100.86	96.90

	Modal analysis %			
Garnet	56.2	52.3	56.0	74.1
Quartz	40.4	4.4	1.6	-
Pyroxmangite	-	11.4	16.5	-
Tirodite	-	5.3	2.5	-
Rhodochrosite	-	-	7.5	11.5
Feldspar	0.2	5.4	0.3	-
Oxides	3.2	21.2	15.6	14.4
Calderite mole % in garnet	-	6-8	45-52	10-15
CaO/MnO (A)	0.028	0.254	0.458	0.372
Fe_2O_3/Al_2O_3 (B)	0.296	0.691	1.812	0.412
B/A	10.57	2.72	3.95	1.10

n.d. : Not determined

rhodochrosite, invariably show a low andradite content (Table 1). In these natural assemblages, therefore, even in the presence of high Ca in the bulk, spessartine–calderite solid solution formed preferentially to spessartine-andradite solid solutions. However, in the absence of pyroxmangite [such as in assemblage (v)], the garnet solid solution accommodated an appreciable andradite component. This shows that the calderite solubility in spessartine is not controlled only by the concentration of individual cations such as Ca in the bulk, but is related to the nature of the mineral reactions that produced the stable phases in diverse assemblages at the ambient *P–T–X* condition of metamorphism.

In the studied rocks, garnets with a high calderite content are largely restricted to carbonate-bearing assemblages. In carbonate-free and low

calcic rocks, such as assemblage (i), no other Ca bearing metasilicate appeared at the ambient physical condition of metamorphism (~600 °C at 6 kbar), and the available Ca was accommodated in garnet. But in relatively Ca-enriched non-carbonatic rocks, such as assemblage (iii), pyroxmangite appeared, although at a slightly higher thermal regime (~650 °C at 6 kbar). Again in carbonatic rocks, pyroxmangite appeared in assemblage (iv), but not in assemblage (v) at similar physical conditions of metamorphism. The experimental study of Peters (1971) and recalibration by Winter *et al.* (1981) indicated a strong dependence of pyroxmangite stability on the X_{CO_2} of the fluid during metamorphic reactions involving a carbonatic precursor. Huebner (1967), however, observed that from a non-carbonatic precursor the stability of pyroxenoid is reached only at
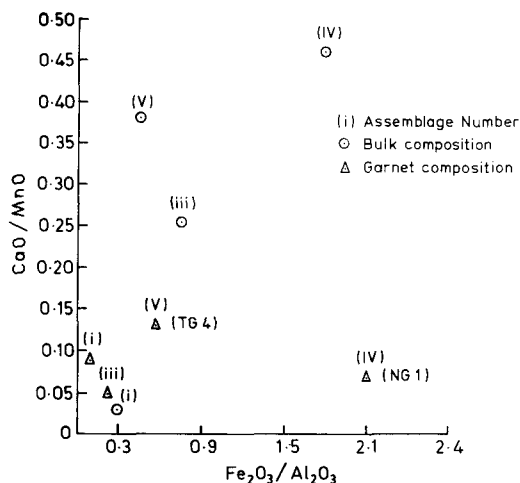
FIG. 1. Composition of garnets and respective bulk compositions in CaO/MnO–Fe_2O_3/Al_2O_3 space.

the highest grades of metamorphism. In this context, the nonappearance of pyroxmangite is evidently related to the physical condition of metamorphism at low $X_{CO_2}^{fl}$ in type (i) rocks even from favourable bulk with X_{Mn} [$=Mn(Mn + Mg + Ca)$] in the range of ~ 0.8 (cf. Huebner, 1986). But a slightly higher thermal regime stabilized pyroxmangite in the type (iii) assemblage. Preferential partitioning of Ca in pyroxmangite favoured the formation of garnets with low calderite component in such non-carbonatic rocks. On the other hand, the $X_{CO_2}^{fl}$ controlled the formation of pyroxmangite during metamorphism of a carbonatic precursor, and produced both pyroxmangite and rhodochrosite [type (iv)], or only rhodochrosite [type (v)] in different assemblages. So the mobile components in the bulk mainly controlled the exact nature of mineral reactions, produced the diverse assemblages, and in turn dictated calderite solubility in garnets. The ubiquitous presence of hematite and total absence of magnetite in these assemblages shows that f_{O_2} was restricted above the hematite–magnetite buffer during metamorphism.

We therefore conclude that (a) calderite solubility in manganiferous garnets is not only a function of pressure, and (b) the mobile components and the thermal regime of metamorphism, rather than only the Ca and Al contents of the bulk, influence

the evolution of variable manganiferous assemblages which in turn control calderite solubility in garnet.

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