

A CHEMICAL STUDY OF GARNETS FROM THE STONY RAPIDS AREA SASKATCHEWAN

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ABSTRACT

Garnet gneisses from the Stony Rapids area consist of quartz, K feldspar, plagioclase (An_{32}), magnetite, biotite, and garnet; in addition sillimanite or hornblende may be present. If the first four minerals are defined as excess components of constant composition, the remaining minerals can be represented in a chemical system of three determining components A , F , and M , where $A = Al_2O_3 - (K_2O + Na_2O + CaO)$, $F = FeO - Fe_2O_3$, and $M = MgO$. Rocks containing biotite-garnet-sillimanite or biotite-garnet-hornblende constitute two assemblages, in which the number of phases equals the number of inert components, not counting TiO_2 and MnO . The effect of isomorphous components TiO_2 and MnO on the composition of minerals probably can be neglected at low concentrations. In such a case, the composition of minerals in each of the two assemblages is determined entirely by external conditions. If temperature, pressure, and chemical potential of water are considered to be interrelated physical variables of a geological environment, such assemblages are univariant. And if temperature is chosen as the only independent variable, the variation in the composition of garnet from a particular univariant assemblage may provide a continuous thermometer scale, within the stability range of the assemblage. This composition variation differs greatly in the two univariant assemblages. The composition of garnet in the bivariate assemblage biotite-garnet (without sillimanite or hornblende) depends not only on temperature, but also on the bulk composition of the rock.

INTRODUCTION

The compositions of minerals forming solid solutions may be used as measures of geological temperatures, provided certain requirements of phase equilibria are met. If a mineral assemblage is treated as a closed thermodynamic system, it should obey the phase rule

$$F = C - P + 2$$

where F is the variance, C is the number of components, and P is the number of phases. For a mineral assemblage to be stable at some arbitrarily fixed temperature and pressure, the variance must at least be two. Thus $F \geq 2$ and the equation reduces to

$$P \leq C$$

which is Goldschmidt's mineralogical phase rule. Commonly it is necessary to treat a mineral assemblage as a system open to certain mobile components, water being the most important one. Therefore, inert and mobile components (C_i and C_m) must be distinguished. The amounts of inert

components, and their chemical potentials at fixed temperature and pressure, are determined by the original composition of the system. On the other hand, mobile components can be exchanged between the system and the surroundings; their chemical potentials are controlled by conditions outside the system under consideration. Thus the mineral assemblage is affected by the amounts of inert components and the chemical potentials of mobile components. Each mobile component, due to an externally controlled chemical potential, increases the number of independent intensive variables by one, and the phase rule for open systems becomes

$$F = C_1 - P + (2 + C_m)$$

For a mineral assemblage to be stable at arbitrarily fixed values of temperature, pressure, and chemical potentials of mobile components, the variance must be at least $(2 + C_m)$, and hence

$$P \leq C_1$$

which is Korzhinsky's mineralogical phase rule (Korzhinsky, 1959, p. 18).

If the number of phases in a system is equal to the number of inert components, phases belonging to solid solution series may show continuous compositional variation in response to changes in temperature, pressure and chemical potentials of mobile components, within the stability range of the particular phase assemblage; this variation is independent of the bulk composition of the system. If the number of phases is less than the maximum possible, the compositions of solid solutions will depend also on the proportions of the inert components.

So far, temperature, pressure, and chemical potentials of mobile components have been considered as independent intensive variables defining the external conditions of the system. However, the physical variables of a geological environment constituting the external conditions for the rocks probably are not entirely independent (Turner & Verhoogen, 1960, p. 506). Particularly under conditions of high-grade metamorphism, an approximation of treating only one variable (e.g. temperature) as independent may be justified. Granting this assumption, a phase assemblage in which the number of phases equals the number of inert components is univariant, and the composition of any phase is potentially a continuous thermometer scale over the stability range of the phase assemblage. It must be emphasized, however, that the composition of a single phase is a significant indicator of temperature only if this phase coexists with all the phases of a specified univariant assemblage.

In metamorphic rocks, garnet is common in a variety of mineral assemblages. If any of these were univariant, the composition of garnet could provide a very useful geological thermometer. Kretz (1959) has

made a chemical study of garnet gneisses, which he considers to have recrystallized at approximately the same temperature. He found that the composition of garnet, even from a specific mineral assemblage, was not constant. He drew attention to a correlation between an increase in the iron-magnesium ratio and an increase in the manganese content, and possibly also calcium content, of garnet. Apparently, either he was not dealing with univariant assemblages, or the rocks did not recrystallize at the same temperature. The purpose of the present study is to compare garnets from different mineral assemblages in Precambrian gneisses of the Stony Rapids area, Saskatchewan, and to examine the possibility of univariance in specific mineral assemblages, in which case the composition of garnet might be used as a relative temperature indicator.

GEOLOGICAL SETTING

The area under consideration is about 9 by 17 miles in size and constitutes the west half of the Clut Lakes topographic map-sheet, between latitudes $59^{\circ}15'$ and $59^{\circ}30'$ North and longitudes $105^{\circ}45'$ and $106^{\circ}00'$ West. The settlement of Stony Rapids, on the Fond-du-Lac River, is situated at its southeast corner. The area is included in a reconnaissance map by Furnival (1941), and has been mapped more recently on a scale of one inch equals one mile by Colborne (1960). The geology of the area and petrography of the rock types are described in Colborne's report.

The rocks of the area are almost entirely Precambrian gneisses and intrusive rocks. Metamorphism has obliterated all primary structures, but excellent compositional layering and the composition of the rocks suggest a sedimentary origin for most of the gneisses. The grade of metamorphism inferred on the basis of mineral assemblages is uniform throughout the area and corresponds to the sillimanite-almandine-orthoclase subfacies of the almandine-amphibolite facies of regional metamorphism (Turner & Verhoogen, 1960, p. 549).

GRAPHICAL REPRESENTATION OF THE PHASE ASSEMBLAGE

The mineral assemblages of rocks from which the garnets have been analyzed are given in Table 1. Analyses of the garnets are listed in Table 2.

In order to treat polyphase assemblages graphically, certain simplifications are necessary. It is assumed that the mineral assemblage of a rock has reached equilibrium at least over a distance of a few inches, because samples for analysis are separated from a volume of hand specimen size. If equilibrium is indeed attained in this chosen thermodynamic system, the mineral assemblage, under constant external conditions, is entirely

TABLE 1. MINERAL ASSEMBLAGES OF GARNET GNEISSES FROM THE STONY RAPIDS AREA

	Quartz	K feldspar	Plagioclase	Biotite	Garnet	Hornblende	Hypersthene	Sillimanite	Magnetite	Spinel	Apatite	Zircon	Pyrite	Anorthite content of plagioclase
M59- 3	X	X	X	X	X	X	—	—	X	—	X	X	—	35
M59- 6	X	X	X	X	X	X	—	—	X	—	X	X	—	35
F59- 30	X	X	X	X	X	—	—	—	X	—	—	X	—	33
F59- 41	X	X	X	X	X	—	—	X	X	—	—	X	—	33
F59- 52	X	X	X	X	X	—	—	—	X	—	—	X	—	30
F59- 55	X	—	X	X	X	—	—	—	X	—	X	X	—	32
F59- 56	X	X	X	X	X	X	—	—	X	—	X	X	X	30
F59- 89	X	X	X	X	X	X	—	—	X	—	X	X	—	33
F59- 92	X	X	X	X	X	—	—	—	X	—	X	X	—	32
F59-103	X	X	X	X	X	X	—	—	X	—	X	X	X	31
F59-106	X	X	X	X	X	X	—	—	X	—	X	X	—	35
F59-113	X	X	X	X	X	X	—	—	X	—	X	X	—	34
F59-125	X	X	X	X	X	—	—	X	X	—	—	X	—	30
F59-130	X	X	X	X	X	X	X	—	X	—	X	X	—	30
F59-133	X	X	X	X	X	X	—	—	X	—	X	X	X	32
F59-138	X	X	X	X	X	—	—	—	X	—	—	X	—	28
F59-139	X	X	X	X	X	—	—	—	X	—	X	X	—	33
F59-140	X	X	X	X	X	—	—	—	X	X	X	X	X	33
F59-147	X	X	X	X	X	—	—	—	X	—	—	X	—	30
F59-162	X	X	X	X	X	X	—	—	X	—	X	X	—	32

determined by the proportions of inert components. If a number of rocks differ only in the proportions of inert components, they are part of the same chemical system defined by these components, even though each rock constitutes a separate thermodynamic system of fixed composition in terms of inert components. Each rock represents a special case of composition possible in the chemical system, and for given values of external conditions each composition will be expressed by a characteristic mineral assemblage. For rocks belonging to the same chemical system, the relation between composition in terms of inert components and resulting mineral assemblage can be shown on a phase diagram, if the number of components is small. Variations of external conditions will be expressed by changes in the pattern of the phase diagram.

The problem of defining a common chemical system for a series of rocks under consideration can be simplified, if the rocks contain common minerals of constant composition, by grouping components in an advantageous manner (Korzhinsky, 1959, p. 66-69). Neglecting trace amounts of apatite, zircon, and pyrite, the mineral assemblages listed in Table 1 suggest the following components: SiO_2 , Al_2O_3 , K_2O , Na_2O , CaO , Fe_2O_3 , TiO_2 , FeO , MgO , and MnO . TiO_2 and MnO normally occur

TABLE 2. GARNET ANALYSES FROM THE STONY RAPIDS AREA

	M59- 3	M59- 6	F59- 30	F59- 41	F59- 52	F59- 55	F59- 56	F59- 89	F59- 92	F59- 103
SiO ₂	—	38.6	—	39.6	—	—	39.7	—	—	—
Al ₂ O ₃	—	22.2	—	23.6	—	—	23.3	—	—	—
Fe ₂ O ₃	—	1.9	—	0.6	—	—	1.2	—	—	—
FeO	27.1	27.5	24.6	24.2	26.0	27.4	27.0	27.9	26.3	30.1
MnO*	1.2	1.1	0.33	0.38	1.4	1.5	1.1	1.1	0.45	2.2
MgO*	2.2	2.2	7.6	10.5	4.7	4.9	6.3	2.5	7.0	1.5
CaO*	6.2	6.7	3.2	1.6	4.3	2.5	1.8	7.5	3.8	8.2
TiO ₂ *	—	0.46	—	0.07	—	—	0.10	—	—	—
P ₂ O ₅	—	0.17	—	0.04	—	—	0.05	—	—	—
		100.8		100.6			100.5			
Unit cell length	—	11.61	—	11.51	—	—	11.52	—	—	—
Refractive index	—	1.803	—	1.783	—	—	1.794	—	—	—
Specific gravity	—	4.10	—	4.02	—	—	4.07	—	—	—
	F59- 106	F59- 113	F59- 125	F59- 130	F59- 133	F59- 138	F59- 139	F59- 140	F59- 147	F59- 162
SiO ₂	—	—	—	—	—	—	—	38.6	40.5	—
Al ₂ O ₃	—	—	—	—	—	—	—	22.9	22.9	—
Fe ₂ O ₃	—	—	—	—	—	—	—	1.0	0.4	—
FeO	29.0	26.8	28.6	26.5	26.7	25.1	27.7	27.9	24.4	29.5
MnO*	1.1	0.57	0.81	1.2	1.2	0.54	0.64	0.54	0.57	1.1
MgO*	2.4	3.3	6.8	1.8	1.8	9.8	3.9	5.4	9.9	1.9
CaO*	7.1	6.8	1.5	6.4	7.1	1.1	5.5	4.2	1.3	8.0
TiO ₂ *	—	—	—	—	—	—	—	0.08	0.17	—
P ₂ O ₅	—	—	—	—	—	—	—	0.03	0.05	—
								100.6	100.2	
Unit cell length	—	—	—	—	—	—	—	11.53	11.51	—
Refractive index	—	—	—	—	—	—	—	1.798	1.784	—
Specific gravity	—	—	—	—	—	—	—	4.09	3.99	—

ANALYST: E. Froese.

*Spectrographic determinations.

in small amounts in lattice sites mainly occupied by other elements. Such isomorphous components do not cause the appearance of a phase. Because of their small amount they are neglected, but they may under some circumstances affect the composition of coexisting minerals. As a next step, use is made of the fact that quartz, K feldspar, plagioclase of a restricted composition, and magnetite are present in all rocks. Components are now defined in such a way that the composition of these minerals become excess components. The excess components can be excluded from the chemical system, resulting in a simpler system. This

procedure is analogous to Thompson's (1957) method of projection through a common phase of constant composition. Actually plagioclase varies somewhat in composition (An_{28-35}), but it is assumed that such small variation will not appreciably affect the composition of other minerals. In the simpler system, the remaining minerals occur as products of the proportions of the so-called determining components. In this manner, the following classification of inert components, using molecular amounts, is arrived at.

Excess components: SiO_2 ; $K_2O \cdot Al_2O_3$; $Fe_2O_3 \cdot FeO$; ($Na_2O \cdot Al_2O_3$ and $CaO \cdot Al_2O_3$ in the proportion of plagioclase of composition An_{32}).

Determining components: $Al_2O_3 - (K_2O + Na_2O + CaO)$; $FeO - Fe_2O_3$; MgO .

Isomorphous components: TiO_2 ; MnO .

If the determining components are designated as A , F , and M , it is seen that A is obtained by subtracting from the total Al_2O_3 those amounts used in the formation of the feldspars; F is equal to FeO less the amount necessary to combine with Fe_2O_3 to form magnetite, and M is equal to MgO . Actually the determining components should also be corrected for any possible amounts combined with the isomorphous components, if the isomorphous components are thought to form end-members of certain minerals. Thus, in plotting the composition of garnet, the amount of Al_2O_3 which combines with MnO to form spessartine can also be subtracted. However, it is not always clear what end member the isomorphous component forms and this correction cannot be applied. Fortunately, because of the small amount of isomorphous components, this correction is not very important.

By defining the components as indicated, the various mineral assemblages of rocks under consideration, excluding those minerals common to all assemblages, can be adequately represented on a phase diagram in terms of three components A , F , and M . It should be noted that with this particular choice of components, A and F may have negative values. Figure 1 is such a phase diagram, with the composition of minerals shown schematically. Together with the four common phases, it is thus possible to account for a maximum of seven phases at arbitrarily fixed external conditions. In one rock (F59-130), due to the occurrence of hypersthene, eight phases are present. This could be a disequilibrium assemblage, even though there is no textural evidence. On the other hand, the rock might constitute an invariant assemblage, assuming only one independent variable of external conditions. Another assemblage (F59-140) contains spinel as inclusions in garnet; this probably is a disequilibrium assemblage. These two rocks will not be considered further in this study.

On the phase diagram shown in Fig. 1, the composition of rocks and

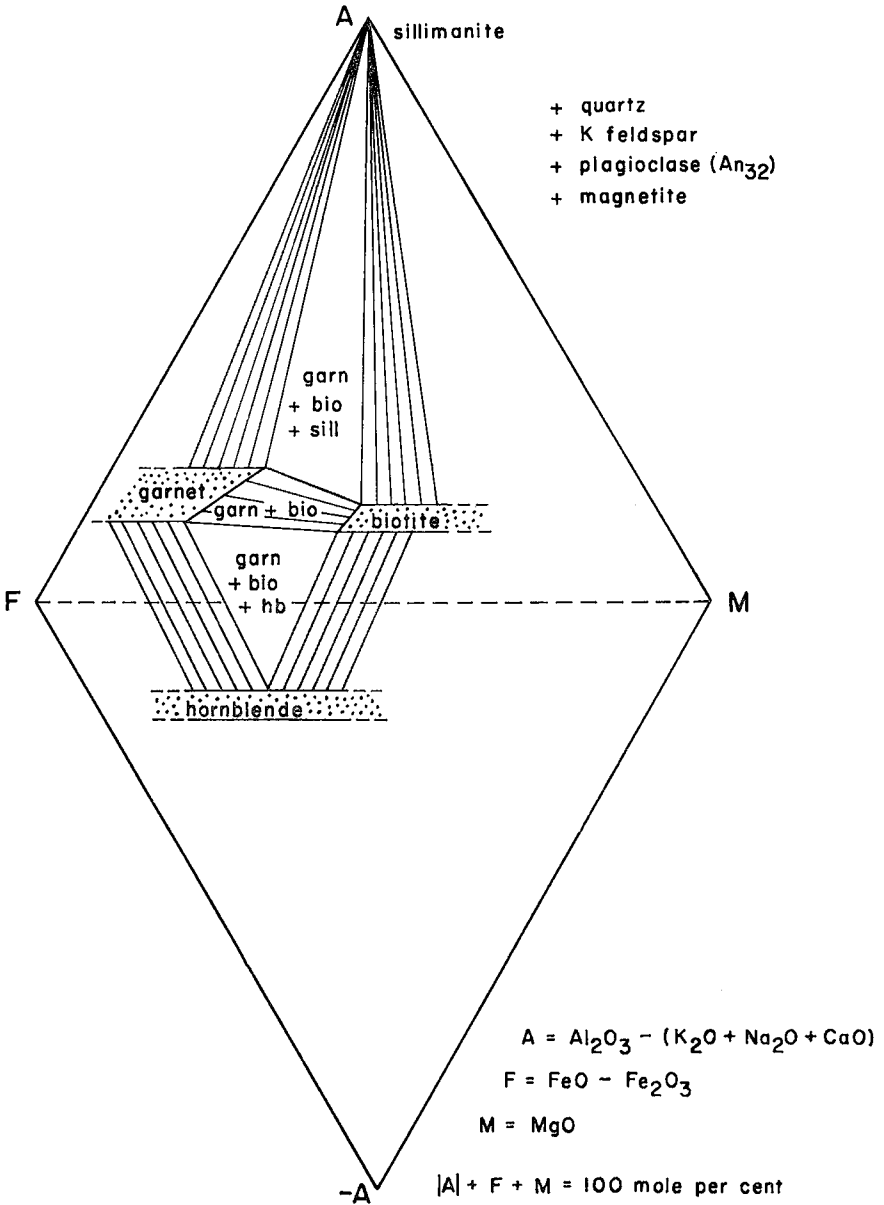


FIG. 1. Phase diagram of mineral assemblages in garnet gneisses from the Stony Rapids area. The composition of minerals is shown diagrammatically.

minerals in the chemical system can easily be plotted. The analyzed garnets (except F59-130 and F59-140) are plotted in Fig. 2, which is an enlarged portion of Fig. 1. Fe_2O_3 was not determined for all garnets and the magnetite correction could not be applied to FeO. Because the amount of Fe_2O_3 is small, particularly in comparison to the amount of FeO, this is not very serious. In order to obtain an Al_2O_3 value for plotting, the garnet is assumed to consist entirely of almandine, pyrope, and grossularite; i.e. these end members are recalculated to 100 per cent. This is analogous to Thompson's (1957) procedure.

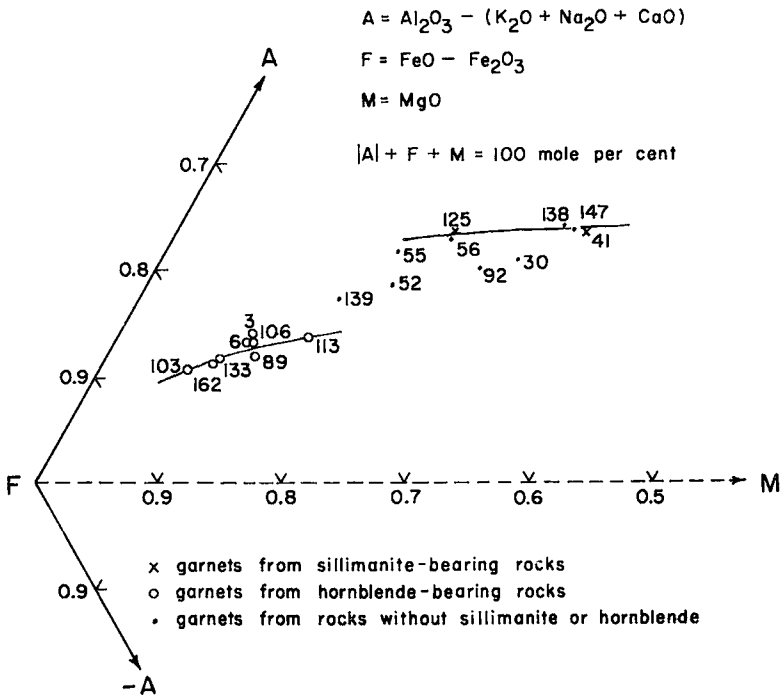


FIG. 2. The composition of garnets from the Stony Rapids area plotted on the type of phase diagram shown in Figure 1.

The phase diagram developed for the system is particularly useful to show phase relationships in rocks of different composition and to plot the composition of coexisting minerals. If the compositional variation of only one mineral is studied, a more conventional representation may be used, presenting the data in a more readily visualized form. In the case of garnet the Al_2O_3 content is constant (disregarding any andradite end member) and K_2O and Na_2O are absent. Therefore, the component

$A = \text{Al}_2\text{O}_3 - (\text{K}_2\text{O} + \text{Na}_2\text{O} + \text{CaO})$ is entirely determined by the amount of CaO, and garnet can be plotted in the three-component system FeO, MgO, and CaO, as shown in Fig. 3. On this diagram the mole per cent spessartine is given as well, in brackets after the specimen number.

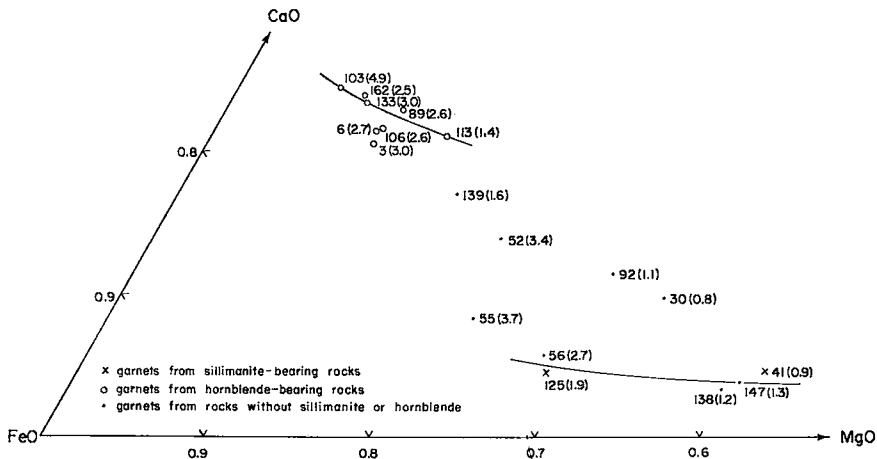


FIG. 3. The composition of garnets from the Stony Rapids area plotted on a ternary diagram of mole ratios FeO, MgO, and CaO. The number in brackets after the specimen number gives the mole per cent spessartine.

GARNET AS A TEMPERATURE INDICATOR

In the following discussion temperature, pressure, and chemical potential of water are assumed to be interrelated physical variables of geological environment. Temperature probably exerts the most important control over the phase assemblage, and will be considered as the only independent variable. If the isomorphous components do not appreciably affect the composition of minerals, seven phases constitute an univariant assemblage in the seven-component system defined in the previous section. Taking into account the four common phases, two such assemblages are indicated by the three-phase fields in the phase diagram of Fig. 1, namely garnet-biotite-sillimanite and garnet-biotite-hornblende, separated by a two-phase field of garnet-biotite. In univariant assemblages the mineral compositions represented by the corners of the three-phase fields may trace out continuous curves in response to variations in temperature, within the stability range of the particular assemblage. The composition of garnet from the two seven-phase assemblages is in fact not constant; the analyses plot along a line for each of the assemblages (Figs. 2 and 3). In this connection it should be pointed out that sillimanite occurs in very

small amounts in two rocks only. Garnets from the two-sillimanite-bearing rocks are characterized by a very low content of CaO (F59-41 and F59-125). Garnets from some other rocks (F59-56, F59-138, and F59-147) have a similarly low CaO content. Probably the composition of these rocks very nearly permitted the appearance of sillimanite. Accordingly, garnet compositions from these rocks were used as well to draw the curve for garnets from sillimanite-bearing rocks.

If the seven-phase assemblages are indeed univariant, the composition of garnet from a particular assemblage is a potential geothermometer, like the composition of sphalerite coexisting with pyrrhotite, and the observed variation in composition could well reflect temperature differences from rock to rock. The prerequisite condition of univariance hinges on the crucial question of the effect of isomorphous components. Strictly speaking every isomorphous component, because it does not cause the appearance of a phase, increases the number of degrees of freedom by one. Thus an additional composition variation is made possible. If the isomorphous component occurs as a trace element, this possible variation can safely be neglected. As the amount of the isomorphous components is increased, their effect may become appreciable. In the present study TiO_2 and MnO could be troublesome. Lack of information prevents an evaluation of the effect of TiO_2 . The possible influence of MnO on the composition of coexisting minerals will be discussed by considering MnO temporarily as an effective component, making the two seven-phase assemblages bivariant. There is a good correlation between MnO content and composition of garnet as plotted on Fig. 3. The compositional variation of garnet from either of the two seven-phase assemblages is mainly in the FeO/MgO ratio; the CaO content is considerably less variable. By neglecting the CaO content, garnet can be plotted on a ternary diagram of FeO, MgO, and MnO (Fig. 4), which shows the correlation between MnO content and FeO/MgO ratio of the garnet. The writer was at first inclined to interpret the two curves in Fig. 4 as isothermal lines; the temperature within the area was thought to have been uniform and the variation in garnet composition was attributed to differences in manganese content of the rocks. However, in the case of the sillimanite-bearing assemblage MnO is almost a trace element and its amount varies very slightly; this should hardly cause such a large variation in FeO/MgO ratio. Furthermore, the shape of the curves do not support this interpretation. Isothermal lines would be expected to intersect the FeO-MgO line of the diagram, like the experimentally determined isotherms of sphalerite intersecting the ZnS-FeS line (Fig. 5); instead the curves on Fig. 4 become tangential. Also the general stability of garnet suggests isotherms steeply inclined to the FeO-MgO line, if facies

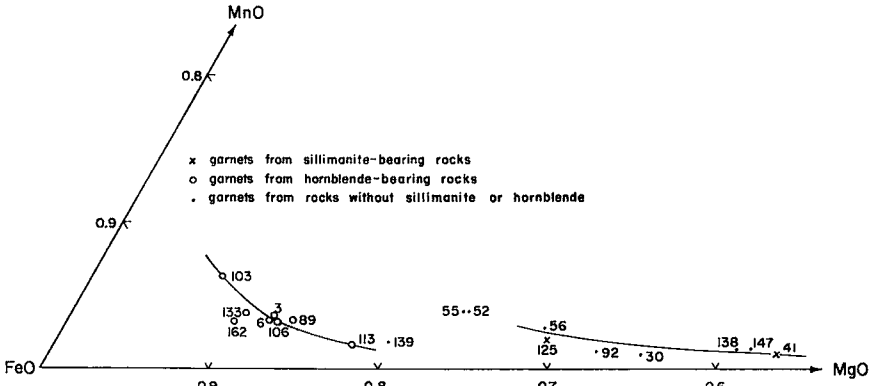


FIG. 4. The composition of garnets from the Stony Rapids area plotted on a ternary diagram of mole ratios FeO, MgO, and MnO.

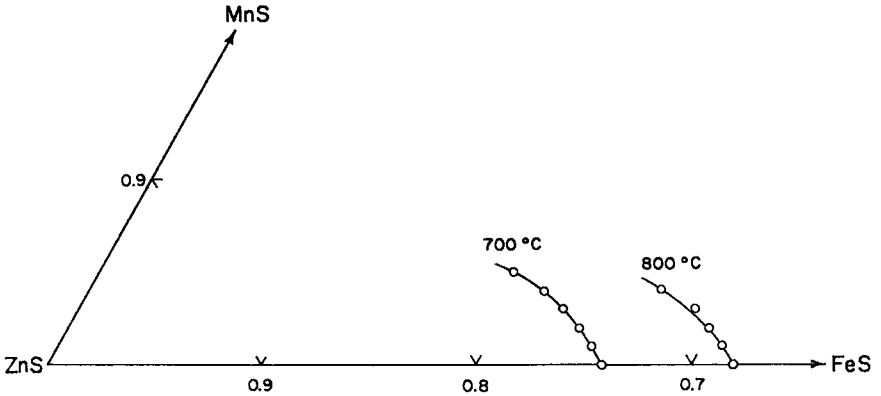


FIG. 5. The composition of sphalerite coexisting with pyrrhotite and the vapour of the system, plotted on a ternary diagram of mole ratios ZnS, FeS, and MnS. (Data from Skinner, 1959.)

boundaries can be taken as a rough indication of the position of isotherms (Fig. 6).

It still is necessary to explain the correlation between MnO content and FeO/MgO ratio of garnet. Dr. T. N. Irvine drew to the attention of the writer that the variation in MnO content and FeO/MgO ratio may both be temperature effects; i.e. the curves of Fig. 4 are path lines of compositions traced out in response to temperature changes. This interpretation is illustrated in Fig. 7. Garnets from the hornblende-bearing assemblage are taken as a possible example and hypothetical isotherms, steeply inclined to the FeO-MgO line, are superimposed on the composi-

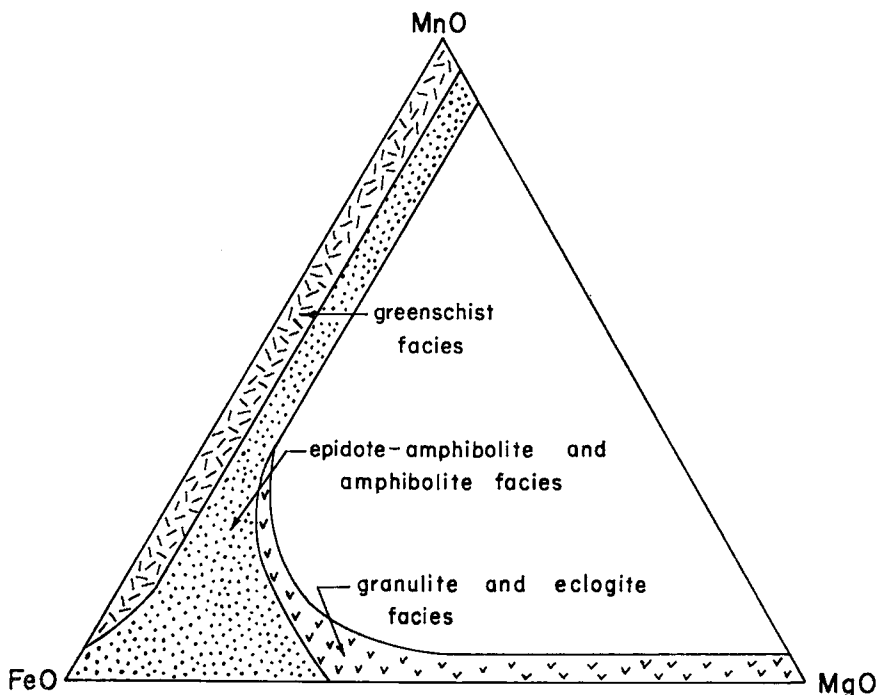


FIG. 6. The composition of calcium-poor garnet from rocks of different metamorphic facies, plotted on a ternary diagram of mole ratios FeO, MgO, and MnO (from Miyashiro, 1953).

tion curve. If a rock contains no MnO, this is a univariant assemblage, which is first formed at temperature T_1 . The composition of garnet changes along the FeO-MgO line (path A) in response to increasing temperature, until the assemblage becomes unstable at T_4 . If MnO is a component, the assemblage becomes bivariant. The composition of garnet, when the assemblage first is formed, may lie anywhere along the curve T_1 , such as point B, C, or D. In response to increasing temperature the garnet composition will then trace out different paths as shown. Obviously the composition at any stage is not only controlled by the temperature, but also by the MnO content of the system, which decided what composition the garnet would have at temperature T_1 . If the composition of rocks is such that garnets, when the assemblage is stabilized at T_1 , have about the same composition, garnets from rocks subjected to different temperatures will plot along a line. This apparently is the case in rocks from the Stony Rapids area. If the isotherms are steeply inclined to the FeO-MgO line, the MnO content will have little effect on the

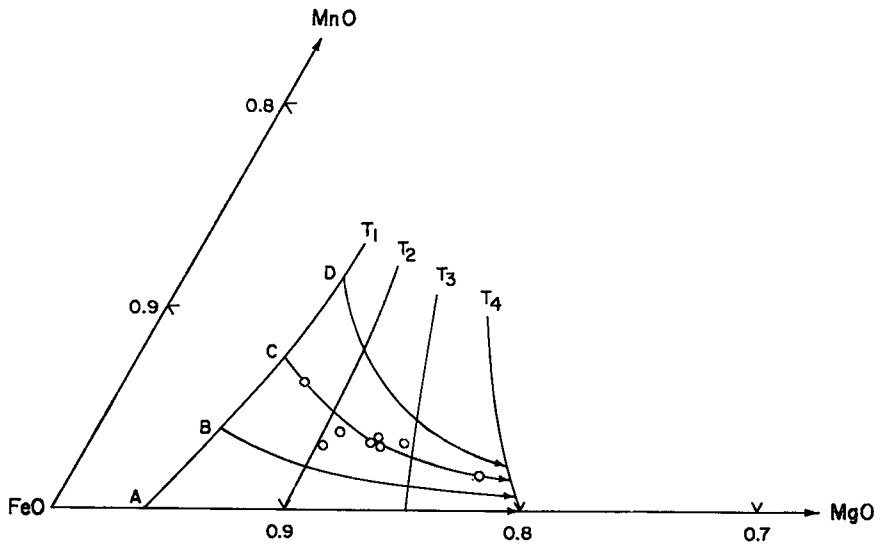


FIG. 7. Hypothetical diagram showing the change in composition of garnet from a certain bivalent assemblage (considering MnO as a component) in response to rising temperature (T_1 to T_4). Different paths (for example A, B, C, or D) will be followed depending on the composition of garnet when the assemblage first formed at temperature T_1 . Garnets from hornblende-bearing rocks from the Stony Rapids area are plotted as a possible example (path C).

FeO/MgO ratio. For example, along curve T_2 this ratio remains about 0.9. It is probably reasonable to assume that at low concentrations (below about 3 per cent spessartine) MnO does not appreciably influence the FeO/MgO ratio. Neglecting MnO, seven-phase assemblages can, in effect, be considered univariant. The composition of garnet is determined by temperature, the FeO/MgO ratio being particularly sensitive to temperature changes.

The composition of garnet from the two univariant assemblages is very different. Garnets from hornblende-bearing rocks have a higher CaO content and a greater FeO/MgO ratio than those from sillimanite-bearing rocks. This fact emphasizes that only garnets from a particular univariant assemblage can be used as a continuous thermometer scale. Garnets from multivariant assemblages cannot be used at all. For example, the composition of garnets from the bivalent assemblage garnet-biotite (Fig. 1), besides varying with temperature is considerably influenced by the bulk composition of the rock.

CONCLUSIONS

Considering temperature as the only independent physical variable of a geological environment, the composition of garnet (or any other mineral of variable composition) from a specified univariant assemblage may be used as a measure of temperature. It is erroneous to attribute a temperature significance to differences in composition of garnet from different univariant assemblages or from multivariant assemblages.

In garnet gneisses from the Stony Rapids area two mineral assemblages are thought to be univariant: quartz, K feldspar, plagioclase (An_{32}), magnetite, biotite, garnet, sillimanite or hornblende. Garnets from the two assemblages show considerable compositional differences; those from the hornblende-bearing assemblage have a higher CaO content and a higher FeO/MgO ratio than the ones from the sillimanite-bearing assemblage. But also garnets from a particular univariant assemblage do not have a constant composition. This variation, predominantly in the FeO/MgO ratio, is interpreted as a reflection of temperature differences. Because the two mentioned assemblages are very common in metamorphic rocks, the FeO/MgO ratio of garnet may provide a continuous thermometer scale, at least in a relative sense, within the stability range of a particular univariant assemblage. If both sillimanite and hornblende are absent the mineral assemblage is bivariant, and the composition of garnet, besides varying with temperature is considerably influenced by the bulk composition of the rock.

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The chemical determinations were carried out under the supervision of Mr. A. W. Hounslow, using his adaptations of methods of rapid silicate analysis. Spectrographic determinations were made by methods developed in the Miller Research Laboratory for silicate analysis (Hawley & MacDonald, 1956). The work was done under the supervision of Mr. J. G. MacDonald. The writer gladly records that both Mr. Hounslow and Mr. MacDonald took personal interest in these results, beyond their capacity as supervisors.

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