

GARNETS FROM UPPER AMPHIBOLITE LITHOLOGIES OF THE BAMBLE SERIES, KRAGERÖ, SOUTH NORWAY

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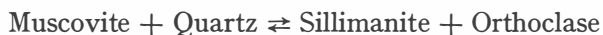
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Abstract. Chemical analyses and certain physical properties of garnets are presented from various lithologies within the Bamble series, near Kragerö, South Norway. Detailed mapping has shown these lithologies to be of the K-felspar-sillimanite sub-facies. The results are compared with similar published data.

Introduction

Recent surveys (e.g. BURRELL 1964, MORTON 1964, PETTERSSON 1964) of the area around Kragerö, South Norway—a portion within the Kongsberg-Bamble district—have revealed suites of metamorphosed lithologies whose mineralogical assemblages demonstrate high stability modified by only slight, and sporadic, retrograde reaction. The author believes that the metamorphic physical environment of the

Levang peninsular sub-area may be placed with some confidence near the univariant reaction:



The reaction temperature is not known, but it is probably between 650 and 700° C. This reaction curve is virtually pressure insensitive. TURNER and VERHOOGEN (1960) suggest 8,000 bars as the upper limit of the amphibolite facies, and a value somewhere between 6,000 and 8,000 bars may be assumed for this area on the basis of the limited experimental evidence available. All data from the complete area have not as yet been fully integrated, but evidence to date suggests that these conditions obtained over a wide area. Thus, the garnet data presented here may be related to an approximately known formation environment. Seven garnet specimens from varying lithologies have been studied (Table 1) but it is not claimed that they are representative of any one particular rock type.

Table 1. *Parent rock of analysed garnets*

Specimen number	Type and mineralogical assemblage
Ø3/25	Garnetiferous amphibolite Plagioclase-Hornblende-Garnet
61/19	Garnetiferous amphibolite. Quartz-Plagioclase-Hornblende-Garnet-Biotite
61/G	Garnetiferous quartz-mica schist Quartz-Plagioclase-Garnet-Biotite
RB	Garnetiferous quartz-mica schist Quartz-Plagioclase-Garnet-Biotite
61/16	Garnetiferous amphibolite Plagioclase-Hornblende-Garnet
MJP	Garnetiferous amphibolite Plagioclase-Hornblende-Garnet
62/34	Foliated granite. Quartz-K-felspar-Plagioclase-Hornblende-Garnet-Biotite

Garnets are common constituents of metamorphic rocks and in these are particularly characteristic of pelitic assemblages. In high grade rocks, the garnet is equivalent in part to greenschist chlorite. Within the area studied garnet is, in fact, only sporadically developed in the

pelitic rocks, but it is an increasingly important constituent of basic rocks and commonly occurs in the foliated granites. Both ENGEL and ENGEL (1962) and LEAKE (1963) have stressed the importance of bulk chemical composition as a critical factor in deciding whether garnet will be formed, but the author finds difficulty in explaining this distribution solely in these terms. Since the presence of sillimanite confirms upper amphibolite conditions for the pelitic assemblages, development of garnet at the expense of biotite might be expected, excess potassium giving K-felspar. SHIDO (1958) has suggested that the development of garnet in amphibolite depends to some extent upon the pressures developed. High water vapour pressure might tend to favour a high biotite to garnet ratio in the pelitic schists, but this completely fails to explain the presence of garnet in granite.

Chemistry

INTRODUCTION

The compositions of individual garnets are generally related to six end-members. WINCHELL (1958) has divided these into two groups; pyrospites (pyrope, almandine, and spessartine) and ugrandites (uvarovite, grossular, and andradite). DEER *et al.* (1962) have noted that there is no continuous variation between these two groups.

Silicon occupies its usual position within the silica tetrahedra of the garnet structure. Both the divalent and trivalent ions are present in octahedral sites. The general formula may be represented as $R_3^{2+}R_2^{3+}Si_3O_{12}$, where R^{2+} is commonly Fe^{2+} , Ca, Mg or Mn and R^{3+} may be Al, Fe^{3+} or Cr. Within this group there appears to be little substitution of aluminium for silicon in the tetrahedral sites, although most published garnet analyses indicate insufficient Si to satisfy the above formula. DEER *et al.* (1962) have suggested that Ti may also replace some Si, but, although tetravalent, the ionic radius suggests affinity with the R^{3+} group.

ANALYTICAL METHODS

Techniques employed for the determination of major components were basically those of SHAPIRO and BRANNOCK (1956) modified somewhat to incorporate more recently improved procedures. Thus the methods

of BLOXAM (1961) and HILL (1956), for silica and alumina respectively, have been used throughout. Ferrous iron determinations utilized the microcolorimetric scheme of WILSON (1960). Most magnesia data were obtained using a simple atomic absorption spectrophotometric procedure that owed much to the work of ALLEN (1958). An excess of strontium was added to each sample solution to eliminate interference caused by aluminium. Atomic absorption spectrophotometry was used also to obtain the zinc and copper data. The method for zinc has been previously fully described (BURRELL 1965) and a similar procedure was employed for copper. Both determinations required ideally a greater quantity of starting material (1g) than was available for mineral analysis. These data are therefore possibly less accurate than values obtained for rock samples.

MAJOR COMPONENTS

The analysis results are given in Table 2. All major components have been determined except for the alkalis and halogens and no attempt was made to determine the possible chromium content. Table 3 gives the calculated ions on the basis of 24 oxygens per formula unit and also the recalculated end-member molecules.

Table 2. *Chemical analyses, refractive indices, and cell-edge length*

Spec. number	Ø3/29	61/19	61/G	RB	61/16	MJP	62/34
SiO ₂	39.36	37.37	38.90	38.98	38.64	39.75	38.51
TiO ₂ . . .	0.00	0.00	0.00	0.12	0.00	0.43	0.00
Al ₂ O ₃ . . .	21.03	18.18	20.26	19.18	21.07	19.22	15.86
Fe ₂ O ₃ . .	1.83	4.98	3.67	4.01	2.67	3.46	6.88
FeO	24.96	23.58	23.37	24.82	25.86	24.93	26.43
MnO . . .	1.12	1.95	0.57	0.96	1.03	1.12	2.15
MgO . . .	7.19	6.43	11.20	7.91	6.02	6.99	3.41
CaO	4.66	5.43	2.57	4.02	4.82	4.53	6.44
	100.15	97.92	100.54	100.00	100.11	100.43	99.68
Zn (ppm)	1,480	4,355	560	230	2,155	5,275	—
Cu (ppm)	1,250	9,490	2,240	2,080	670	1,990	—
N	1.791	1.791	1.782	1.791	1.793	1.792	1.806
a Å	11.55	11.57	11.53	11.56	11.56	11.57	—

Table 3. *Ionic content (24 O) and percentage end-member molecules*

	Ø3/25	61/19	61/G	RB	61/16	MJP	62/34
Si.....	6.081	6.012	5.937	6.073	6.018	6.156	6.226
Al ⁴	—	—	0.063	—	—	—	—
Al ⁶	3.825	3.441	3.577	3.518	3.866	3.496	2.992
Ti.....	—	—	—	0.009	—	0.046	—
Fe ²⁺	0.204	0.599	0.422	0.468	0.309	0.409	0.835
Fe ³⁺	3.221	3.170	2.983	3.200	3.369	3.227	3.575
Mn.....	0.148	0.216	0.074	0.122	0.131	0.149	0.292
Mg.....	1.652	1.546	2.547	1.834	1.395	1.609	0.826
Ca.....	0.770	0.938	0.421	0.674	0.805	0.753	1.117
Almandine .	55.63	53.59	49.50	56.10	59.10	56.24	61.53
Spessartine	2.56	4.41	1.23	2.08	2.30	2.59	5.02
Pyrope	28.53	26.14	42.27	31.31	24.47	28.04	14.22
Andradite .	5.06	14.83	7.00	10.51	7.40	10.35	19.23
Grossularite	8.22	1.03	—	—	6.73	2.78	—

The chemical variations are shown most clearly in graphical form. Mg, Ca, and Fe + Mn are plotted as percentage divalent ions in Fig. 1, and Fig. 2 gives the coexisting Mn and Fe ion contents. It may be

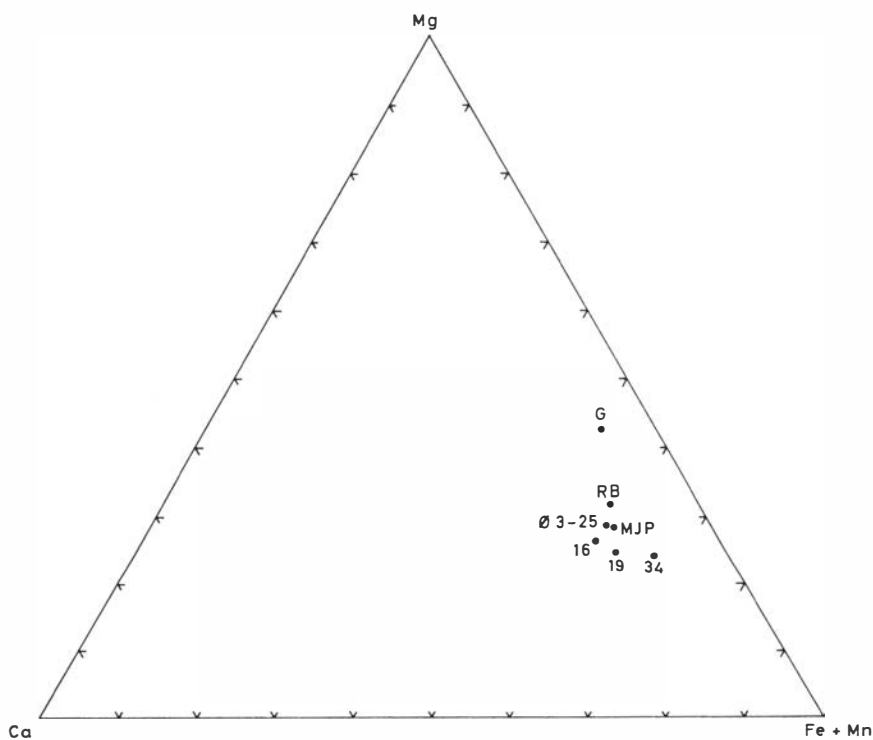


Fig. 1. Divalent ions — Mg : Ca : Fe²⁺ + Mn.

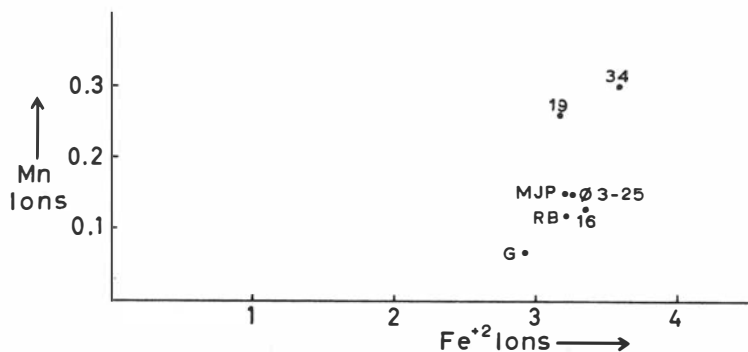


Fig. 2. Co-existing Mn and Fe²⁺ ion contents.

seen that all garnets consist predominantly of almandine-pyrope. The almandine content is consistently over 50%, but is higher in the granitic garnet and lowest in pelitic specimens. Amphibolitic garnets are intermediate in this respect. Likewise, the spessartine content is over 5% in 61/34 and generally appears to 'follow' ferrous iron closely. Fig. 3 gives the R^{3+} site content; the possible effect of chromium is

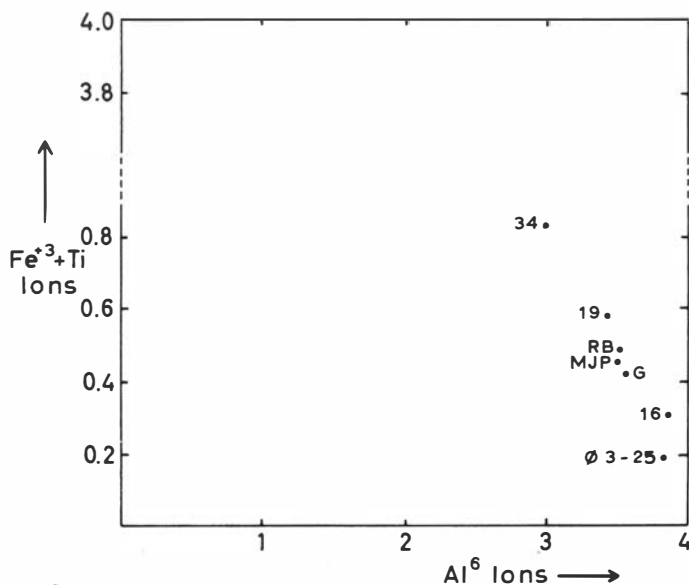


Fig. 3. R^{3+} site content.

an unknown factor here. Pelitic and amphibolitic garnets contain variable, but small, amounts of the andradite molecule, whereas 61/34 shows nearly 20% of this end-member; it is possible that some of the latter may be due to oxidation during sample preparation. Substitution of Al for Si is either absent or very slight as in 61/G.

TRACE COMPONENTS

The zinc and copper contents of the garnets are given in Table 2. Fig. 4 shows the zinc content of coexisting garnet-hornblende and garnet-biotite pairs, and Fig. 5 the copper data of coexisting garnet and hornblende only. Qualitative emission spectrographic determi-

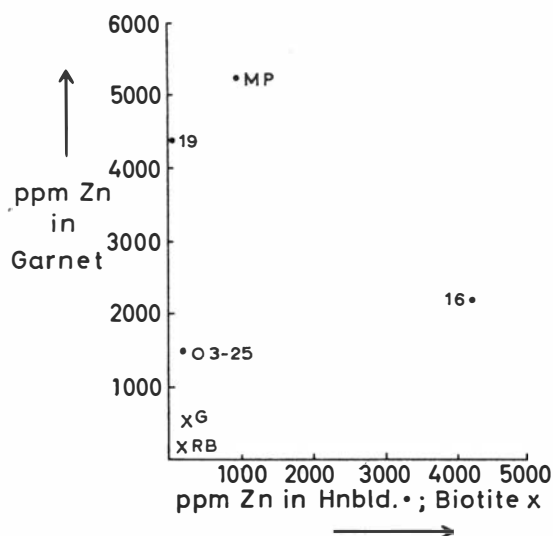


Fig. 4. Zinc (ppm) in co-existing garnet-hornblende and garnet-biotite pairs.

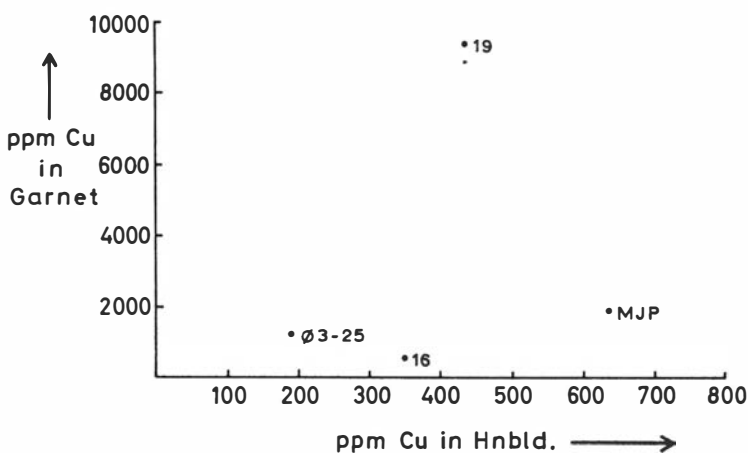


Fig. 5. Copper (ppm) in co-existing garnet-hornblende pairs.

nations showed the presence of relatively large amounts of sodium, potassium, and lithium in the specimens, but no quantitative data have been obtained.

Optical and X-ray diffraction data

The values obtained for the refractive index are given in Table 2, and Fig. 6 shows these data plotted against the almandine-pyrope content which has been recalculated to 100%.

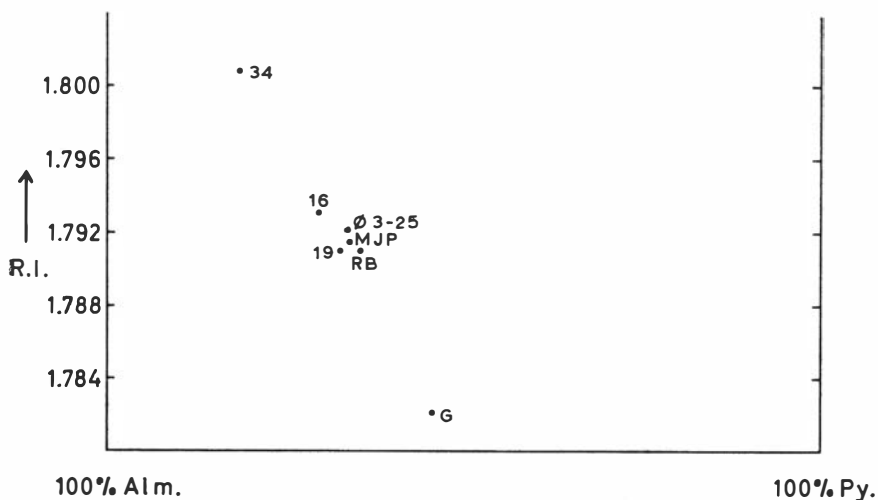


Fig. 6. Refractive indices — almandine-pyrope content recalculated to 100%.

Table 4. *d hkl data; 2θ 10–70°*

hkl	Ø3/25	61/19	61/G	RB	61/16	MJP
222		3.332			3.349	3.335
400	2.887	2.888	2.879		2.887	2.892
420	2.576	2.581	2.571		2.588	2.586
332	2.460	2.470	2.456		2.464	2.464
422	2.357	2.359	2.353		2.363	2.361
510	2.264	2.267	2.257		2.267	2.267
521	2.108	2.110	2.102		2.113	2.111
440	2.040	2.042				2.042
611	1.873	1.876	1.866		1.876	1.877
444	1.667	1.669	1.663		1.670	1.670
640	1.603	1.604	1.599	1.606	1.604	1.603
642	1.544	1.546	1.540	1.547	1.546	1.546
800	1.445	1.448	1.441		1.445	1.445
840					1.293	

The isometric structure of garnet ensures a simple X-ray diffraction pattern which can be indexed easily. Each unit cell contains eight formula units (i.e. $Z = 8$); the somewhat complex cell structure is well illustrated in DEER *et al.* (1962). Table 4 gives the d hkl data from which values for a have been calculated (Table 2) using the method advocated by MIYASHIRO (1959) and CHINNER *et al.* (1960). This procedure justified values quoted only to the second decimal place. Fig. 7 gives the cell edge length (a) plotted against the almandine-pyrope content recalculated as for Fig. 6 above. Fig. 8 reproduces

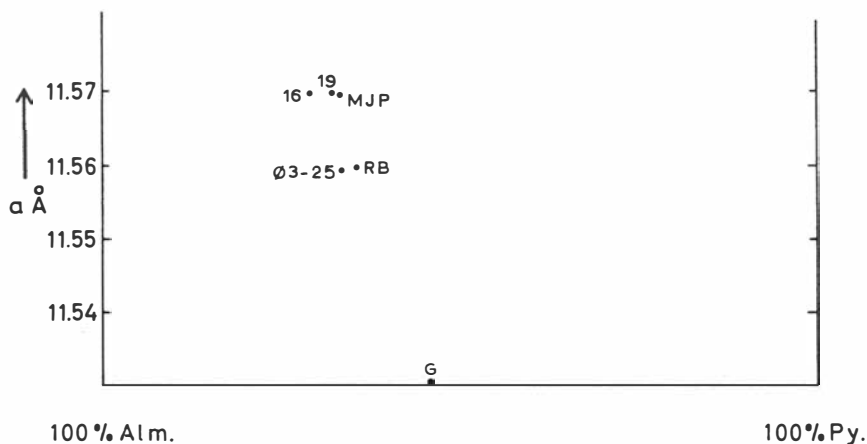


Fig. 7. Cell-edge length — almandine-pyrope content recalculated to 100%.

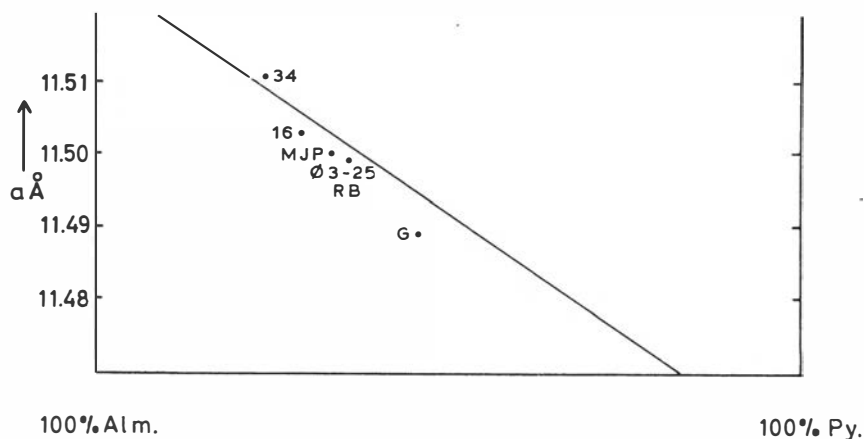


Fig. 8. As for Fig. 7 with effect of andradite content statistically removed.

the same data, but here a has been recalculated so as to remove the 'influence' of the andradite, spessartine, and grossular contents. The oblique line on this graph joins the pure a values of almandine and pyrope. Deviations from this line may be attributed to errors inherent in the recalculation procedure. Comparison of Figs. 7 and 8 illustrates clearly the effect of the relatively large cell-edge length of the andradite molecule. Fig. 9 is a plot of a values against the corresponding refractive indices.

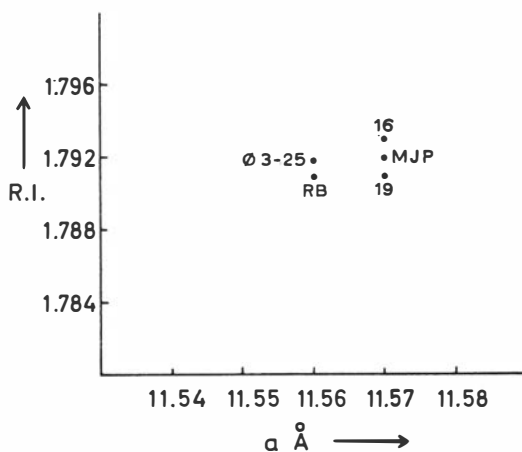


Fig. 9. Refractive indices — cell-edge length.

Infrared analysis

Six garnet specimens have been analysed over the frequency range 5,000 to 650 cm^{-1} by infrared spectroscopy. Table 5 lists the principle absorption bands noted. The spectra in the range 1300 to 650 cm^{-1} are shown in Fig. 10; this includes the principle Si-O bond stretching frequency.

Silicate infrared analysis has been seriously attempted only in recent years, e.g. KELLER and PICKETT (1959), HUNT *et al.* (1950), MILLER and WILKINS (1952), KELLER *et al.* (1952). The complex structure of most silicate minerals coupled with the variable ionic-covalent character of the structural bonding has made interpretation of obtained spectra exceedingly difficult. However, the Si-O bond stretching-vibration frequency occurs within the range normally stud-

Table 5. *Infrared absorption data*

Ø3/25	61/19	61/G	RB	61/16	MJP
	690		695	695	
	810		805		
	885		880	885	880
910	900	920	900	905	905
975	965	1000	965	980±	970
1080	1090±	1090	1085	1090	1090
	1260				1260

ied and shifts in these peaks have been correlated with varying degrees of substitution of Al into the silicon-oxygen tetrahedra of feldspars (THOMPSON and WADSWORTH 1957, MILKEY 1960). This technique now awaits extension to other mineral groups. Further research is also required on silicate absorption patterns in the longer wavelength range, since many characteristic bands occur outside the range normally scanned.

Discussion

All garnet specimens are believed to have been taken from lithologies formed within the K-feldspar-sillimanite subfacies of the almandine-amphibolite facies. Little experimental work has been recorded, but YODER (1955) has determined the upper stability limits of almandine, and at temperatures below 785° C almandine would appear to be stable at atmospheric pressure; this reaction temperature increases with pressure. A consistent variation of chemical content with metamorphic grade has been discussed in several recent publications. MIYASHIRO (1953) found that the spessartine content of pelitic garnets decreased with increase in grade. ENGEL and ENGEL (1960) confirmed this trend and noted in addition that it was accompanied by an increase in the ratio of pyrope to almandine; LAMBERT (1959) recorded a decrease in the calcium content with metamorphic grade. MIYASHIRO emphasized that Fe²⁺ and Mg would normally be considered unstable in the octahedral sites, but that higher temperatures would ensure greater stability for the pyrope-almandine molecules. Thus STURT

(1962) suggested that the ratio $\text{CaO} + \text{MnO} : \text{FeO} + \text{MgO}$ provided a valuable indicator of metamorphic grade. However, it should be noted that Sturt's work takes no account of the antipathetic relationship between ferrous iron and magnesium at high temperatures. The garnets considered here conform well to the general pattern, being predominantly solid solutions of almandine and pyrope, but with not inconsiderable amounts of ugrandite. A complete evaluation of the relationships between pyralspite and ugrandite is most desirable. Insufficient specimens have been studied to permit more than a very generalized picture of the chemical variations between the various lithologies. The granite garnet is highest in both ferrous and ferric iron and in manganese. The two specimens from pelites are richest in magnesium and the amphibolitic garnets in calcium, but the variations, though consistent, are very slight.

In the last decade, a great deal of information has been published dealing with the trace element contents of co-existing minerals (e.g. RAMBERG and DE VORE 1951, NICKEL 1954, KRETZ 1959, 1960, TUREKIAN and PHINNEY 1962). Briefly, the partition of an ion between co-existing phases depends only upon the controlling physical conditions during crystallization. Thus, under fixed pressure conditions, the partition coefficient (i.e. Nerst's Law) is a function of the obtaining temperature. It must be emphasized that all other physical conditions should be fixed, an ideal condition which can seldom be approached in natural geological systems. As an extension of this concept, the partition of an ion into two separate structural sites within a single mineral phase could be considered. The quality and quantity of trace element data obtained for co-existing minerals of this work are insufficient to provide much of significance. In general, the garnets are greatly enriched in both zinc and copper compared with the co-existing hornblende phase, but there is no trend as would be expected for rocks from a unique metamorphic subfacies. The zinc content of the pelitic assemblages (garnet-biotite pairs) is much less than that of the amphibolites. This is to be expected, since the latter are probably of igneous origin.

It is somewhat easier to determine a few major physical parameters of members of the garnet series than to perform a complete chemical analysis, and several workers have published graphs which correlate the major oxide contents with combinations of cell-edge length, re-

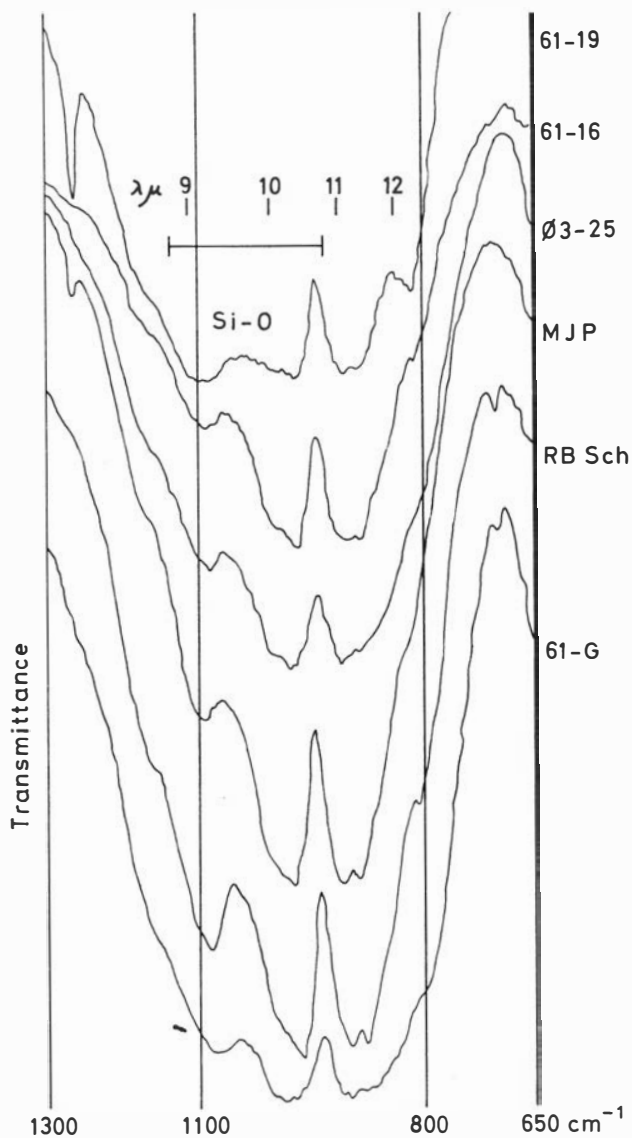


Fig. 10. Infrared absorption traces over range 1300-650 cm^{-1}

fractive index, and density (e.g. SKINNER 1956, SRIRAMADAS 1957, WINCHELL 1958). Such data, however, can be only a first approximation, and an accurate analysis is always desirable. Figs. 7 and 8 clearly demonstrate the effects of the various constituent molecules upon the physical properties of the garnet family. Both the relatively large cell size and refractive index of andradite result from substitution of ferric iron (radius 0.64 Å) for aluminium (radius 0.51 Å) in the garnet structure. On the assumption that the cell-edge value is an additive function of the molecular proportions of the end-members, GNEVUSHEV *et al.* (1956) have constructed formulae which enable a to be approximately calculated from chemical analyses.

The Si–O stretching frequency has been stated by MILKEY (1960) to be in the wavelength range 9.1 to 10.8 μ ; this is indicated in Fig. 10. The garnets show three major absorption peaks in this region as has been previously noted by GNEVUSHEV and FEDOROVA (1962). These workers further showed that the wavelength value of the centre peak (i.e. the 960–1000 cm^{-1} frequency of Fig. 10) could be directly correlated with the ionic radius of the R^{2+} ion. Specimen 61/G is the most pyrope-rich garnet and thus shows a reverse trend to that noted by the above workers.

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