Mineralogy of vesicles in an olivine leucitite at Cosgrove, Victoria, Australia

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SUMMARY. An olivine–titanomagnetite–apatite–clino-pyroxene–mica–nepheline–feldspar assemblage occurs in late-stage vesicles in a small outcrop of olivine leucitite at Cosgrove, Victoria. The vesicles were formed by exsolution of volatiles at an early stage in the cooling history of the lava. Subsequently, a volatile-rich residual liquid filled cavities and fractures, giving rise to a coarse-grained pegmatoid rock type similar in over-all mineralogy to the vesicles. The volatiles facilitating crystallization in both the vesicles and the pegmatoid were probably enriched in F, CO_2, and P. A number of geothermometers applied to the vesicle assemblage failed to agree on likely crystallization temperatures.

THE olivine leucitite at Cosgrove, 175 km north of Melbourne, Victoria, is the southernmost outcrop of a belt of Tertiary leucite-bearing lavas in south-eastern Australia (Birch, 1976, 1978; Cundari, 1973). While only a very small volume of lava is preserved in outcrop, the Cosgrove occurrence is of interest owing to the development of a coarse-grained pegmatoid phase and a spatially related vesicular phase of the lava. These have been exposed in the westernmost of three quarries on the outcrop. The mineralogy and geochemistry of the lava and its pegmatoidal differentiate have been described previously by Birch (1976, 1978) and this paper concentrates on the mineralogy of the vesicles.

Representative samples have been lodged in the collection of the National Museum of Victoria, as the restricted occurrence of the pegmatoid and vesicles will result in their imminent removal by the quarrying process.

Geological notes

The Cosgrove leucitite forms a low rise about 800 m by 400 m in area and is approximately 30 m thick. Only one flow is apparent. No contacts with other rock units are exposed but the age of the flow has been established at 6.8 m.y. (late Miocene) by fission-track dating of apatite crystals in the vesicles (Gleadow, pers. comm., in Birch, 1976). The fresh

lava is dark grey and aphyric, and relatively uniform over the entire occurrence. Olivine (generally iddingsitized) and clinopyroxene are phenocrystic in a groundmass of diopside–sahlite, leucite, titanomagnetite, ilmenite, nepheline, and biotite.

The pegmatoid phase occurs as irregular patches, veinlets, and veins up to 40 cm thick. Several large boulders up to 1 × 1 × 0.5 m were discovered initially on the quarry floor. Clinopyroxene is the dominant mineral, as randomly oriented, dark-brown crystals up to 5 mm long. Their composition lies mainly in the range sahlite–Ti sahlite–Na sahlite, but small amounts of green Ti-aegirine-augite and Ti-aegirine are present (Birch, 1978). Nepheline, alkali feldspar, apatite, and titanomagnetite are common, with minor ilmenite, amphibole, sodalite, and aenigmatite. Analcime has partially replaced nepheline (or possibly completely replaced leucite). Analogous pegmatoids are known from Begargo Hill, in the NSW leucitite belt (Cundari, 1973).

Mineralogy of the vesicles

Vesicles within the leucitite are of two types. The more common type occurs as irregular cavernous regions, up to about 10 cm across, lined with crystals, and frequently associated with coarse-grained pegmatoid patches and veins within the lava (fig. 1). The other type forms subspherical cavities usually less than 2 cm across (fig. 2). These are usually associated with thin pegmatoid veinlets and patches of similar mineralogy to the irregular vesicles.

Irregular vesicles

In the irregular vesicles the main minerals present are clinopyroxene, apatite, and titanomagnetite.

Clinopyroxene occurs as tabular crystals flattened on {010}, up to 4 mm long and dark
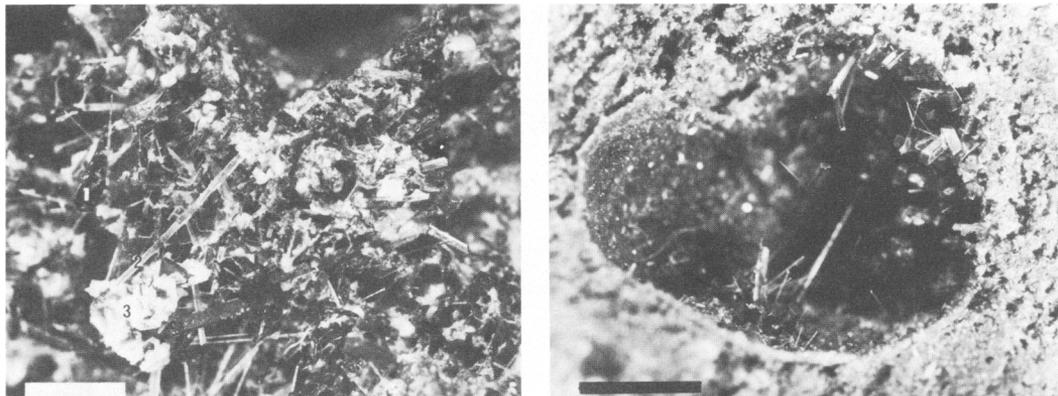
FIGS. 1 and 2. FIG. 1 (*left*). The assemblage in a typical irregular vesicle in the Cosgrove leucitite. Minerals shown are clinopyroxene (1), apatite (2), and analcime-nepheline (3). Titanomagnetite crystals are also present. Scale bar is 2.5 mm. (National Museum of Victoria Specimen M 32259). FIG. 2 (*right*). A typical subspherical vesicle in the Cosgrove leucitite showing prismatic diopside crystals. The cavity is bordered by small, granular diopside crystals. Scale bar is 2.5 mm. (N.M.V. Specimen M 32258).

purplish-brown in colour. Some zoning is evident, mainly of the hourglass or sector variety, but some concentric zoning is observed with the development of thin greenish rims. Crystals frequently have a hollow termination.

Microprobe analyses (Table I) show compositions ranging from diopside to high Ti-Al diopside-sahlites. The high Ti contents (1.9–4.6 wt % TiO_2) are presumably responsible for the dark-brown colour. Core-rim zonation shows increasing Si, Mg, and Ca and decreasing Ti, Al, Fe, and Na. In compositional terms the vug clinopyroxenes are similar to those in

FIG. 3. Compositions of clinopyroxenes from the Cosgrove occurrence, plotted in the alkali pyroxene triangle $Mg-(Fe^{2+} + Mn)-Na$. The pyroxenes in the vesicles do not show the Na enrichment of those in the pegmatoid.

the pegmatoid except for the pronounced sodium enrichment in some of the latter pyroxenes (fig. 3).

Apatite is a conspicuous phase in the vesicles, forming abundant, colourless, prismatic crystals up to 5 mm long. Microprobe analysis of apatite from both the pegmatoid phase and the vesicles reveals similar compositions, with F contents of about 4 wt % and SrO contents of about 0.5 wt % (Table II). Microanalysis of apatite from the pegmatoid resulted in CO_2 and H_2O contents of 2.6 and 1.9 wt % respectively. Thus the apatite is a carbonate fluorapatite, with a significant OH content. The relatively high Si and Sr contents reflect the advanced stage reached in the differentiation process (Nash, 1972). The average refractive index is 1.635 ± 0.005. Lattice constants of $a = 9.408$ Å and $c = 6.889$ Å were determined independently by B. M. Gatehouse (Chemistry Department, Monash University) and W. G. Mumme (Division of Mineral Chemistry, CSIRO). None of these parameters define the composition of apatite uniquely.

Titanomagnetite forms small, black octahedra up to 1 mm across. They are locally abundant, and are frequently perched on clinopyroxene crystals. Examination of polished grains under reflected light reveals exsolution lamellae of ulvöspinel in a titaniferous magnetite, both phases confirmed by X-ray diffraction. Bulk compositions of the grains are close to the ulvöspinel end member (Table III). Although microprobe analyses of composite grains should always be presented with some hesitation, the fact that recalculation of structural formulae on an ilmenite, rather than ulvöspinel, basis gives totals close to 100 %, suggests a degree of trans-
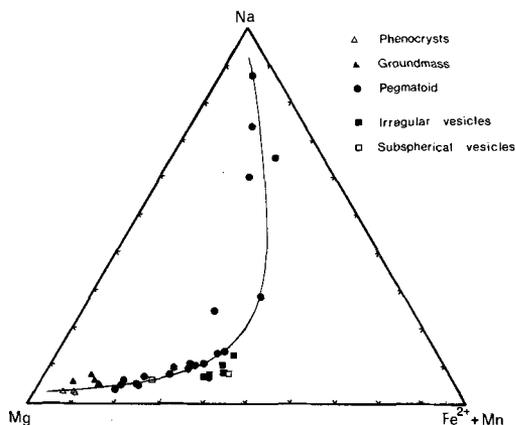
TABLE I. *Electron microprobe analyses of clinopyroxene crystals from vesicles in the Cosgrove leucitite*

	I	2	3	4	5
SiO_2	50.79	51.27	47.11	53.59	52.73
TiO_2	3.07	1.86	4.60	0.21	0.35
Al_2O_3	2.75	2.07	4.75	0.04	0.12
Fe_2O_3	—	1.33	1.62	3.00	2.85
FeO	5.87	6.08	5.81	4.02	6.36
MnO	0.13	0.18	0.12	0.10	0.11
MgO	14.07	13.73	12.16	15.00	13.06
CaO	23.00	22.39	22.57	24.52	24.28
Na_2O	0.55	0.76	0.85	0.43	0.54
Total	100.23	99.67	99.59	100.91	100.40

Structural formulae based on 4 cations and 6 oxygens

Si	1.878	1.911	1.773	1.967	1.967
Al^{iv}	0.120	0.089	0.211	0.002	0.005
Al^{vi}	—	0.002	—	—	—
Ti	0.085	0.052	0.130	0.006	0.010
Fe^{3+}	—	0.037	0.046	0.083	0.080
Fe^{2+}	0.182	0.189	0.183	0.123	0.199
Mn	0.004	0.006	0.004	0.003	0.003
Mg	0.776	0.763	0.682	0.821	0.726
Ca	0.911	0.894	0.910	0.964	0.970
Na	0.039	0.055	0.062	0.031	0.039

Atomic Percentages

Mg	77.5	75.3	73.3	83.9	75.1
Fe^{2+} + Mn	18.6	19.2	20.1	12.9	20.9
Na	3.9	5.4	6.7	3.2	4.0

1. Diopside crystal from irregular vesicle (CLCV 31). 2. Diopside–sahlite from irregular vesicle (CLCV 36). 3. Diopside–sahlite from irregular vesicle (CLCV 32). 4. Green diopside in subspherical vesicle (CLCV 33). 5. Green sahlite in subspherical vesicle (CLCV 34).

formation from the β- to the γ-spinel (titano-maghemite).

Nepheline forms stumpy, clear or white, and translucent crystals up to 4 mm long with a hexagonal basal section about 1 mm across (fig. 4). The crystals however are usually skeletal with hollow cores, having been altered to analcime. The microprobe analyses in Table IV show a relatively small range in composition with Ne/Kp ranging from 5.0–7.4.

Mica occurs rarely as reddish brown, single or groups of crystals less than 1 mm across. Microprobe analyses (Table II) show it to be a barian biotite (Mg/(Mg + Fe) = 0.56) with a high Ti content (10.5 wt % TiO_2) with 1.6 % F. It is indistinguishable in composition from the biotite flakes occurring in the groundmass of the lava (Birch, 1978).

Sanidine forms small, colourless, tabular crystals in some of the cavities. Microprobe analyses gave

TABLE II. *Analyses of apatite and mica from the vesicles in the Cosgrove leucitite*

	I	2	3
SiO_2	0.73	0.48	33.18
TiO_2	0.07	0.03	10.82
Al_2O_3	—	—	—
FeO*	0.24	0.12	15.09
MnO	0.04	0.02	0.18
MgO	0.22	0.17	10.84
CaO	54.54	55.10	—
SrO	0.47	0.57	—
BaO	—	—	5.95
Na_2O	—	—	0.76
K_2O	—	—	6.78
P_2O_5	37.59	38.79	—
F	4.22	4.02	1.60
Cl	0.33	0.27	0.02
H_2O^+ } H_2O^- }	1.89	n.d.	3.12†
CO_2	2.60	n.d.	
$-O \equiv F, Cl$	1.85	1.73	0.68
Total	101.09	97.84	101.01

Structural formula on basis of 26 (O, Cl, F)

P	5.147 }	
Si	0.118 } 5.84	
C	0.574 }	
Ti	0.009 }	
Fe	0.032 }	
Mn	0.005 } 9.59	
Mg	0.053 }	
Ca	9.451 }	
Sr	0.044 }	
F	2.158 }	
Cl	0.090 } 4.29	
OH	2.039 }	

Structural formula assuming (OH, F, Cl) = 4.00

|		
Si	5.120 }	
Ti	1.256 }	
Al	2.428 }	
Fe	1.947 }	
Mn	0.024 } 15.191	
Mg	2.494 }	
Na	0.227 }	
K	1.335 }	
Ba	0.360 }	
F	0.781 }	
Cl	0.005 } 4.00	
OH	3.214 }	

* Total iron as Fe^{2+}. † Calculated. n.d. Not determined.
1. Average composition of apatite in pegmatoid. 2. Average composition of apatite in vesicles. 3. Barian biotite in vesicle.

Note on analytical methods. Compositions of the apatites and mica were determined by electron microprobe, with the exception of CO_2 and H_2O in the pegmatoid apatite, which were determined by microanalytical techniques on a small separate. The similarity of the pegmatoid and vug apatite compositions suggests a similar F/(F + OH) value for each, and this was assumed when using the apatite–biotite geothermometer (see text). The OH content of the vesicle mica was calculated by assuming stoichiometry.

compositions ranging from Or_{55} Ab_{45} to Or_{76} Ab_{24} (fig. 5). A sanidine of composition Or_{71} Ab_{29} (mole %) was found to coexist with nepheline of average composition Q_{24} Ne_{67} Kp_9 (mole %) (Table IV). X-ray diffraction investigation revealed partial ordering and evidence for development of cryptoperthites.

Olivine is observed only very rarely as small, red-brown crystals which are always iddingsitized.

Other minerals: Analcime forms irregular white aggregates in seams and skeletal replacements of nepheline. Occasional globules of calcite and clusters of gypsum crystals on apatite needles have been observed. Several unidentified Mn-rich phases have also been detected and are under investigation.
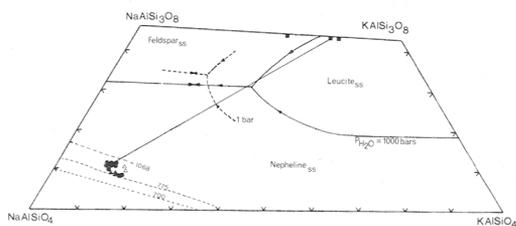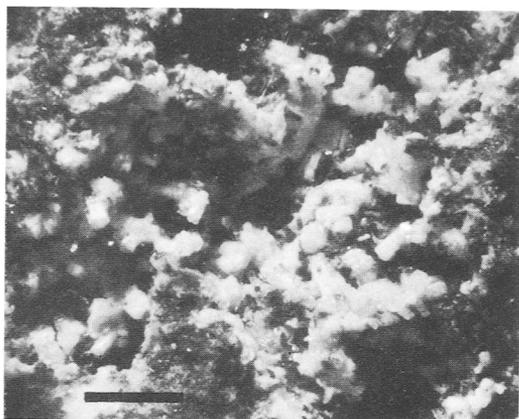
FIGS. 4 and 5. FIG. 4 (*above*). Nepheline crystals in the irregular vesicles in the Cosgrove leucitite. Scale bar is 2.5 mm. (N.M.V. Specimen M 34521). FIG. 5 (*below*). Compositions of nepheline (solid circles) and feldspars (solid squares) from the vesicles plotted in the $NaAlSiO_4$-$KAlSiO_4$-SiO_2 system. Solid boundary curves are for the hydrous system at 1 kbar. The 700 and 775 °C isotherms in the hydrous system at 1 kbar and the 1068 °C isotherm in the dry system are shown by short dashed lines (after Hamilton, 1961). Coexisting phases are joined by the solid line. (Solid and open triangles indicate nepheline compositions in the leucitite groundmass and the pegmatoid respectively.)

TABLE III. *Electron microprobe analyses of titanomagnetite crystals in vesicles in the Cosgrove leucitite*

	I	2
SiO_2	0.13	0.09
TiO_2	28.55	27.30
Al_2O_3	1.06	0.61
FeO	62.41	63.63
MnO	0.60	0.74
MgO	2.09	1.99
CaO	0.01	—
Cr_2O_3	0.37	0.29
NiO	0.06	0.11
Na_2O	0.10	0.11
Total	95.38	94.88

Structural formula on ulvöspinel basis

USP	84.38	80.50
Mt	15.62	19.50
Fe_2O_3	10.63	13.28
FeO	52.84	51.68
Total	96.45	96.00

Structural formula on ilmenite basis

Ilm	55.69	53.36
Hm	44.31	46.66
Fe_2O_3	45.68	47.95
FeO	21.31	20.48
Total	99.96	99.47

1. Titanomagnetite in vesicle (CLOV 47). 2. Titanomagnetite in vesicle (CLOV 48).

Sub-spherical vesicles

The mineralogy of these is more simple. Diopside is the major mineral, forming transparent, grass-green, prismatic crystals up to 4 mm long (fig. 2). They are comparable in composition to the purplish-brown clinopyroxenes in the irregular vesicles in terms of the Mg:Fe:Ca ratios, but Ti and Al contents are extremely low (Table I). No hour-glass zoning is observed but there is a suggestion of weak concentric zonation with the opposite trends to those shown by the titaniferous diopsides.

Small apatite, titanomagnetite, and sanidine (Or_{55} Ab_{45}) crystals occur rarely in these vesicles.

Discussion

Alkaline lavas show a propensity to form late-stage pegmatoidal veins and patches, often associated with the development of vesicles containing

TABLE IV. *Electron microprobe analyses of nepheline and K-feldspar in vesicles in the Cosgrove leucitite*

	1	2	3	4	5	6
SiO_2	47.28	45.96	47.95	64.00	64.96	64.24
TiO_2	0.02	0.04	0.16	0.13	0.17	0.02
Al_2O_3	30.50	31.49	30.23	18.70	19.28	19.05
Fe_2O_3	0.62	0.53	0.98	0.52	0.19	0.36
MgO	0.06	0.06	0.08	0.02	0.03	0.03
CaO	—	—	—	—	—	0.03
Na_2O	16.82	16.51	16.51	3.24	5.32	2.65
K_2O	3.10	4.46	3.35	12.22	9.35	12.22
BaO	—	—	0.04	0.19	—	1.19
Total	98.40	99.05	99.30	99.04	99.29	99.79

Structural formula based on 32 oxygen

	1	2	3	4	5	6
Si	9.027	8.796	9.09	11.845	11.835	11.849
Al	6.866	7.105	6.74	4.080	4.141	4.141
Fe^{3+}	0.089	0.077	0.14	0.073	0.024	0.003
Ti	0.003	0.006	0.02	0.018	0.026	0.049
Mg	0.017	0.016	0.02	0.006	0.008	0.008
Ca	—	—	—	—	—	0.006
Na	6.228	6.127	6.06	1.163	1.877	0.948
K	0.755	1.088	0.81	2.885	2.174	2.875
Ba	—	—	0.00	0.013	—	0.086
	22.985	23.115	22.88	20.083	20.085	19.965

Mole %					Mole %		
Q	22.6	18.0	24.4	Or	71.0	53.6	73.4
Ne	69.0	69.7	66.7	Ab	28.7	46.4	24.2
Kp	8.4	12.4	8.8	An	—	—	0.2
				Cel	0.3	—	2.2

1. Nepheline in vesicle (CLNV 2). 2. Nepheline in vesicle (CLNV 6). 3. Average composition of nepheline coexisting with feldspar in analysis 4 (CLNV 8, 9). 4. K-feldspar in vesicle coexisting with nepheline in analysis 3 (CLFV 3). 5. K-feldspar in vesicle (CLFV 2). 6. K-feldspar in vesicle (CLFV 1).

a high-temperature mineral assemblage. The formation of a volatile-rich residuum during cooling of the lava at or near the surface may be due to normal differentiation accompanying extensive crystallization of olivine, pyroxene, and Fe-Ti oxides, or possibly to a liquid immiscibility effect (Kogarko, 1974; McKenzie and White, 1970). Evidence for the latter process is lacking at Cosgrove as gradational textures between coarse-grained pegmatoid patches and the aphyric lava are often observed. At Cosgrove, the irregular vesicles resulted from the release of volatiles at an early stage in the cooling history of the erupted lava. The close association between vesicles and pegmatoid may in part be due to the release of volatiles having provided channelways through which the residual liquid could migrate.

The mechanism by which abundant, well-formed crystals of clinopyroxene, titanomagnetite, and apatite line vesicle walls, as at Cosgrove, is not well understood, although it is clear that the presence of volatiles at high temperatures in the cavities is a requirement. That crystallization under these circumstances was rapid is indicated by the hollow terminations observed on many of the clinopyroxene crystals. It is likely that a sudden release of volatiles from the lava would have resulted in an increase in the solidus temperature of the residual melt, particularly in a narrow zone around each vesicle. This may have facilitated the nucleation of the observed phases and their growth into the volatile-filled cavities. There is no evidence for the volatile release having caused quenching of the melt around the vesicles—neither glass nor reduced crystal grain size is observed.

Since titanomagnetite crystals may be found perched on clinopyroxene and as clinopyroxene may grow on apatite, the general crystallization

sequence at Cosgrove was probably (olivine)-apatite–clinopyroxene–titanomagnetite, followed by mica, nepheline, and feldspar. Considerable overlap in both time and temperature undoubtedly occurred however.

Geothermometry

There are a number of geothermometers which may theoretically be applied to the Cosgrove vesicle assemblage. Two of the available geothermometers are based on coexisting nepheline and feldspar—those of Powell and Powell (1977) and Perchuk and Ryabchikov (1968).

Using equation 10 of Powell and Powell, a crystallization temperature of 660 °C is obtained for the co-existing nepheline and K-feldspar compositions given in Table IV. ($x_{K.Ne} = 0.09$, $x_{Na.Fsp} = 0.29$, $x_{Na.Ne} = 0.67$, $x_{K.Fsp} = 0.71$.) These data are considerably outside the range of compositions used in establishing the geothermometer. This is shown on fig. 6, a series of calibration curves for T ranging from 600 °C to 800 °C. The $x_{K.Ne}$ and $x_{SiO_2.Ne}$ points used to plot these curves were obtained, for a value of 0.29 for $x_{Na.Fsp}$, by interpolation of the data in Table V of Powell and Powell. The Cosgrove data point appears to be close to a linear extension of the 700 °C curve, not unreasonably distant from the 660 °C result obtained by direct calculation.

While the Powell and Powell geothermometer takes into account the excess silica content
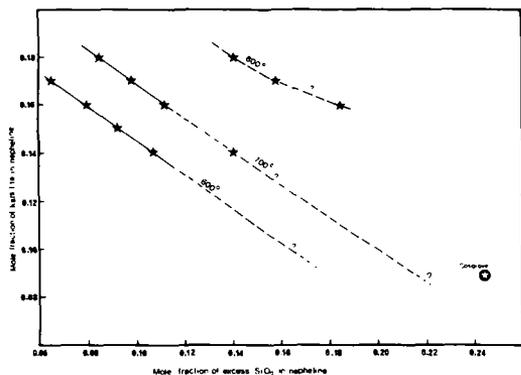

FIG. 6. Nepheline–alkali feldspar geothermometric curves for temperatures of 600 °C, 700 °C, and 800 °C adapted from data of Powell and Powell (1977). The $x_{SiO_2.Ne}$ value for each point on the curves was obtained by interpolation of data in Table 5 of Powell and Powell (1977) for a feldspar composition given by $x_{Ab = 0.29}$ (see text). The data point for co-existing nepheline–feldspar from Cosgrove plots close to the linear extension of the 700 °C curve (see text).

of nepheline, the geothermometer of Perchuk and Ryabchikov (1968, fig. 7) neglects this factor. Applying the geothermometer to the same Cosgrove nepheline/K-feldspar data gives a temperature estimate of less than 400 °C (a temperature of 700 °C is obtained by plotting the absolute, i.e. non-normalized nepheline data). Thus, whether or not excess silica in nepheline is taken into account exerts a profound influence on the estimate of temperature—this may apply in particular to very high-silica nephelines, such as those from Cosgrove.

In the system $NaAlSiO_4$–$KAlSiO_4$–SiO_2–(H_2O) the vesicle nephelines plot between the experimentally determined isotherms for 775 °C (hydrous system at 1 kbar) and 1068 °C (dry system at 1 atm.) (Hamilton, 1961). Due to the differing experimental water pressures, it is only possible to estimate a crystallization temperature of around 900 °C for the nepheline composition used previously for the Powell geothermometer (Q_{12} Ne_{77} Kp_{11}, weight %).

These temperature estimates may be compared with that obtained using an apatite–biotite geothermometer developed by Stormer and Carmichael (1971) and based on the partitioning of fluorine and hydroxyl. Using $F/(F+OH)$ values of 0.51 and 0.20 for the apatite and mica respectively (see notes to Table II), an apatite–biotite crystallization temperature of 900 °C is obtained using the curves presented in Fig. 3.4 of Carmichael et al. (1974).

It is therefore clear that application of the various geothermometers available has failed to produce consistent and reliable estimates of crystallization temperatures. It is difficult to decide whether this is due to weaknesses (of varying degree) in the geothermometers themselves, or to the imprudence of applying them to rapidly crystallized and hence, possibly, non-equilibrated vesicle assemblages. Crystallization temperatures in the range 700–900 °C are probably geologically reasonable for the Cosgrove occurrence.

Volatiles

The composition of the volatiles involved in the crystallization of the vesicle assemblage may only be guessed at, as the fugitive constituents need not enter the crystallizing phases, but rather, facilitate crystal growth (Kogarko, 1974). However, the existence of relatively abundant carbonate-fluorapatite in the irregular vesicles indicates that F, CO_2, H_2O, and P were essential constituents, while small amounts of Cl were undoubtedly present.

The behaviour of F, OH, and Cl in silicate melts

depends considerably on the melt composition. Thus, F is more soluble than Cl in granitic melts (Fuge, 1977; Kilinc and Burnham, 1972), while the reverse is the case for basaltic liquids (Fuge, 1977). In the presence of F and OH, Cl will not enter any hydroxy minerals crystallizing, but will concentrate in the melt fraction (Fuge, 1977; Carmichael et al., 1974). Late-stage F-bearing mica is present in the groundmass of the Cosgrove leucitite, while apatite is probably represented as inclusions in the leucite (this is suggested by a bulk rock P_2O_5 content of 1.2 wt. %). Thus, F and OH were probably entering apatite and mica at the expense of Cl. Subsequent to the expulsion of the volatiles giving rise to the vesicles, the mineral assemblage indicates that F and OH continued to enter apatite and mica, this time in the vesicles. Any Cl was probably expelled in an aqueous fluid phase.

In the residual liquid represented by the pegmatoid, F again entered the relatively abundant apatite, while any remaining Cl entered the late crystallizing sodalite and possibly a late-stage Na- and Cl-rich aqueous solution. The similarity of apatite compositions in the vesicles and pegmatoid (Table II) suggests that essentially the same reservoir of volatiles, in terms of the F to Cl ratio, was available in both environments. The essential difference is that volatiles in the pegmatoid were trapped in crystal interstices, thus explaining the presence of amphibole, sodalite, aenigmatite, and aegirine in the pegmatoid, and their absence in the open vesicles. The action of a late-stage brine with a high Na/K ratio may have been responsible for analcimization of any original leucite in the pegmatoid and some of the nepheline in the pegmatoid and vesicles. This influence probably also extended to such sub-solidus reactions as the oxidation and exsolution of titanomagnetite and the partial ordering and exsolution of sanidine.

Volatiles facilitating crystallization within the subspherical vesicles containing the green diopside crystals may have been relatively depleted in F and P, thereby explaining the near absence of apatite. It is difficult to account for the occurrence of the two types of vesicle clinopyroxene other than by a difference in volatile composition.

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