The Oka carbonatite complex, Quebec: geology and evidence for silicate-carbonate liquid immiscibility

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Abstract

Associated carbonate and silicate igneous rocks from the northern intrusive center of the Oka complex (Quebec, Canada) probably formed from immiscible carbonate and silicate magmas in situ, as their structure, petrography, isotope ratios and mineral chemistry are consistent with immiscibility. The intrusion consists of intimately intermixed rocks derived from carbonate magma (some jacupirangites, okaites, melilite-bearing carbonatites) and silicate magma (some jacupirangites, melilitites). Some jacupirangites and okaites are crystal cumulates. Inferred order of crystallization is: jacupirangites; nepheline okaites and nepheline melilitite; okaites and melilitites; and melilitite-diopside carbonatite and diopside melilitite. Carbonate-rich ocelli are common in melilitites, and silicate-rich ocelli are present in rocks derived from carbonate magma. Rare earth elements are six times more abundant in carbonatite than in melilitite, consistent with REE partition experiments on immiscible carbonate and silicate liquids. Initial strontium isotope ratios (~0.7033) are identical in silicate and carbonate rocks, as required by immiscibility. The strongest evidence for immiscibility comes from identity of compositions of magmatic minerals in rocks derived from contemporaneous immiscible fractions (required by liquid immiscibility, but inconsistent with other modes of fractionation). Fassaite pyroxenes in the jacupirangites (derived from both silicate-rich and carbonate-rich magmas) span identical compositional ranges (9.3-16% Al₂O₃; 3.2-4.7% TiO₂). Diopsidic pyroxenes in carbonatites and melilitites also have identical ranges (1.45-2.0% Al₂O₃; 0.31-0.45% TiO₂). Perovskites with magmatic compositions are preserved only in nepheline okaites and nepheline melilitite; here, the perovskites are of identical composition. Other minerals (calcite, melilite, nepheline) do not preserve magmatic compositions.

Introduction

Silicate-carbonate liquid immiscibility has been considered an important process in carbonatite genesis (e.g., Koster van Groos and Wyllie, 1963; Middlemost, 1974; LeBas, 1977), primarily because of the common association of carbonatites and alkaline silicate rocks. Experimental studies indicate that immiscible silicate and carbonate magmas are likely in geologically reasonable compositions (Koster van Groos and Wyllie, 1966, 1968, 1973; Koster van Groos, 1975b; Verwoerd, 1978; Freestone and Hamilton, 1980; Harris and Koster van Groos, 1981), and immiscible silicate and carbonate magmas may be preserved in melt inclusions (Rankin and LeBas, 1974) and in pyroclastics (Hay, 1978; Mariano and Roeder, 1983). The case for liquid immiscibility in the genesis of plutonic carbonatite complexes is less certain because magmatic and sub-solidus reactions may have destroyed much of the evidence that is available in experimental charges or volcanic rocks.

The Oka pluton, a Cretaceous intrusive complex of the Monteregian Hills petrographic province (Gold, 1963, 1972; Gold et al., 1967; Gold and Vallée, 1969; Fig. 1) includes a wide range of silicate and carbonate rocks, is fairly well exposed, and has not been deformed or metamorphosed. Many workers (e.g., Eby, 1975; Koster van Groos, 1975b; Cullers and Medaris, 1977; Verwoerd, 1978) have inferred that immiscibility between silicate and carbonate magmas was important at Oka, but these studies did not specifically state which lithologies formed from contemporaneous immiscible magmas. Nor did any of these studies consider more than one line of evidence to support their conclusions. This paper evaluates liquid immiscibility in the Oka complex using all pertinent field, petrographic and chemical data, and delineates which lithologies may have been formed contemporaneously by in situ immiscibility between silicate and carbonate magmas.

Geology

The geology of the Oka complex is described by Gold (1963, 1972) and Gold and Vallée (1969); their work forms...
the basis for Figure 1 and the following description. The Oka complex is a composite pluton emplaced into metamorphic rocks and anorthosite 40 km west of Montreal, Quebec. The complex includes two distinct intrusive centers, which give the complex its "figure-eight" plan. Both intrusive centers include an earlier outer annulus of alkalic silicate-rich rocks and carbonatite (labelled "moderate-alkali carbonatite" on Fig. 1; Fig. 2), and a later central plug of carbonatite (labelled "low-alkali carbonatite" on Fig. 1). The complex is cut by anorthitic breccia pipes and dikes of dolomitic carbonatite (Treiman and Essene, 1984).

Petrography

Jacupirangites are defined as rocks consisting predominantly of titanaugite (see Williams et al., 1954, p. 82 for definition of type jacupirangite); at Oka, the pyroxene is a titaniferous fassaite (Peacor, 1967). Two varieties of jacupirangite occur at Oka; coarse- and fine-grained. The coarse-grained jacupirangite (C-Jac, Fig. 3a) consists of centimeter-sized euhedral fassaite crystals and minor magnetite in a cumulate texture with interstitial melilite, calcite, and hauyne (probably altered nepheline). The fassaite is partially altered to an aggregate of phlogopite, melilite, and perovskite; alteration is most intense near calcite. Fine-

Table 1. Rock types, sample numbers, abbreviations and average modal mineralogies of Oka rocks

<table>
<thead>
<tr>
<th>Rock Type and Number</th>
<th>Abbrev.</th>
<th>Modal Mineralogy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coarse Jacupirangite</td>
<td>C-Jac</td>
<td>Fassaite 60-80%, melilite 5-20%, phlogopite 1-5%, calcite 1-10%, perovskite 1-3%, hauyne 1-3%, pyrrhotite tr., apatite tr.</td>
</tr>
<tr>
<td>Fine Jacupirangite</td>
<td>F-Jac</td>
<td>As above.</td>
</tr>
<tr>
<td>Nepheline Osilite</td>
<td>Ne-Ok</td>
<td>Melilite 30-50%, nepheline 30-50%, phlogopite 10%, magnetite 5%, calcite 5%, apatite 2%, perovskite 1%, pyrrhotite tr.</td>
</tr>
<tr>
<td>Nepheline Melilitite</td>
<td>Ne-Mel</td>
<td>Nepheline 50%, melilite 30%, perovskite 5%, magnetite 5%, phlogopite 5%, calcite 3%, apatite tr., pyrrhotite tr.</td>
</tr>
<tr>
<td>Okosite</td>
<td>Ok</td>
<td>Melilite 30-70%, calcite 15-50%, magnetite 5-10%, perovskite 1-5%, apatite 2-5%, phlogopite 1-5%, pyrrhotite tr., diopside tr.</td>
</tr>
<tr>
<td>Melilitite-carbonatite</td>
<td>Me-C</td>
<td>As above but with calcite &gt; melilite.</td>
</tr>
<tr>
<td>Melilitite</td>
<td>Mel</td>
<td>Melilite 70%, phlogopite 15%, hauyne 5%, perovskite 3%, magnetite 3%, pyrrhotite 2%, apatite 2%, grossular tr.</td>
</tr>
<tr>
<td>Melilitite-diopside Carbonatite</td>
<td>Mel-Di-C</td>
<td>Calcite 50-60%, melilite 3-50%, diopside 25%, apatite 5-20%, nioelite 2-5%, magnetite 1-3%, phlogopite 1-3%, perovskite 1-3%, pyrochlore tr.-3%, pyrrhotite 1%, natalolite tr., noman tr., grossular tr.</td>
</tr>
<tr>
<td>Diopside Nepheline</td>
<td>Di-Ne</td>
<td>Diopside 30-60%, melilite 20-50%, phlogopite 5-15%, grossular 2-10%, perovskite 2%, magnetite 0-3%, pyrrhotite 2-5%, apatite tr., pyrochlore tr., sphene tr.</td>
</tr>
<tr>
<td>Ijolite</td>
<td>Ij</td>
<td>Diopside 20-60%, melilite 0-20%, phlogopite 5-15%, grossular 2-10%, perovskite 2%, magnetite 0-3%, pyrrhotite 2-5%, apatite tr., pyrochlore tr., sphene tr.</td>
</tr>
<tr>
<td>Monticellite-carbonatite</td>
<td>Mo-di-C</td>
<td>Calcite 75%, monticellite 7%, melilite 3%, phlogopite 5%, diopside 5%, apatite 2%, pyrochlore 2%, perovskite 1%, pyrrhotite tr.</td>
</tr>
</tbody>
</table>
grained jacupirangite (F-Jac; Fig. 3b, c) consists of euhedral to subhedral fassaite (1 mm long) with interstitial, poikilitic melilite, phlogopite, hauyne (probably altered nepheline) and perovskite. Within the fine-grained jacupirangite are ocelli of coarse-grained, calcite-rich rock (Fig. 3b) which resemble nepheline okaite (see below). Relict fassaite within the ocelli suggest that they were originally coarse-grained jacupirangite.

Okaite and nepheline okaite (Ok and Ne-Ok) are coarse-grained rocks (Figs. 3d, e; 1 cm grain size) composed of euhedral to anhedral silicate (melilitite, nepheline, hauyne) and oxide (perovskite) minerals with interstitial calcite, apatite, and phlogopite. The proportion of interstitial calcite varies from only a few percent (Fig. 3e) to near 50% (Fig. 3d). The latter rocks are essentially melilite carbonatites; an arbitrary cutoff between okaite and melilite carbonatite is 50% (volume) calcite. Hauyne in the okaites commonly occurs between nepheline and calcite, and is probably reaction product. In the classification of Streckeisen (1980), the okaites would be called varieties of melilitolites.

Melilitite (Mel), nepheline melilitite (Ne-Mel), diopside melilitite (Di-Mel), and ijolite (Ij) are fine-grained (0.1–1 mm grain size) silicate-rich rocks with hypidiomorphic granular textures (Figs. 3f, g). Mineral distribution is commonly irregular, with patchy concentrations of melilitite or pyroxene. Hauyne, commonly present, is probably an alteration of nepheline. In the classification of Streckeisen (1980), these rocks would be called varieties of micro-melilitolites. As used at Oka, “ijolite” encompasses a wide range of mafic rock types, fine and coarse grained, some without nepheline, and some which are almost certainly altered okaites. The ijolites referred to here are fine-grained, hypidiomorphic rocks, commonly intermixed with carbonatite (Fig. 2), and are best exposed on the south-west side of the complex. Ijolite is texturally identical to the melilitolites discussed above, and differs only by addition of nepheline, garnet and commonly wollastonite or pectolite, elimination of magnetite, and partial or complete elimination of melilitite.

Carbonatites are all coarse-grained, calcite-rich rocks (sovites), containing euhedral to subhedral oxide and silicate minerals in a hypidiomorphic matrix of calcite. At least four mineralogic varieties are distinguished. Monticellite-diopside carbonatite (Mo-Di-C) occurs as dikes cutting other rocks of the complex and as the central stock in the northern intrusive center. Melilitite carbonatite (Me-C) is gradational to okaite (Fig. 3d) by decrease in calcite content. Melilitite-diopside carbonatite (Me-Di-C; Fig. 3h) contains diopsidic pyroxene, commonly adhering to melilitite, but also as discrete grains. Melilitite is commonly altered. The two most common alteration assemblages are: diopside + grossular + calcite + natrolite; and vesuvianite + calcite + diopside + natrolite. The grossular-bearing alteration is earlier than the vesuvianite alteration. Late carbonatites have no melilitite, but contain diopside that is chemically similar to that in the melilitite-diopside carbonatite.

**Interpretation**

The outer annulus of the northern intrusive center at Oka, here informally called the Jacupirangite-Okaite-Melilitite-Ijolite-Carbonatite (JOMIC) suite, consists of intermixed silicate-rich and carbonate-rich rocks in incomplete concentric bands. The banding shown on the map (Fig. 1) is a gross simplification of the structure; see Gold and Vallée (1969) for a more detailed map of the complex as a whole, and Gold (1972) for maps of individual outcrops. Lithologic units shown on Figure 1 are really zones where that rock type is the most common lithology. For example, the outcrop of Figure 2 is in a zone mapped as ijolite, although much of the exposure is carbonatite. Despite the structural complexity, there is some regularity to the distribution of rock types. Jacupirangites, okaite, and melilitite are more common to the north and east in the annulus; carbonatite and ijolite are predominant to the south and west.

Regular gradations in mineralogy and mineral chemistry among rocks of the JOMIC suite suggest that they are products of a single magma. Based on intrusive contacts and mineralogic changes (Table 2), the sequence of emplacement is interpreted to be: jacupirangites; nepheline okaite and nepheline melilitite; okaite, melilitite carbonatite and melilitite; diopside melilitite and melilitite-diopside carbonatite; and diopside-carbonatite (without melilitite). It is not certain whether ijolite is magmatic and contemporaneous with the diopside carbonatite or is an alteration of the diopside melilitite. Ijolite could form from diopside melilitite by replacement of magmatic magnetite by garnet + perovskite and by replacement (partial or complete) of magmatic melilitite by diopside + nepheline ± wollastonite ± calcite. Textures indicative of these reactions are observed in the okaites and carbonatites, where they have not progressed to completion.

**Liquid immiscibility in the JOMIC pluton**

The JOMIC intrusion is a good candidate for an investigation of silicate-carbonate liquid immiscibility because it consists of closely associated silicate-rich and carbonate-rich rocks. Among the silicate-rich lithologies, it is essential to contrast those derived from silicate-rich magmas from those formed by crystal accumulation from carbonate-rich magmas.

The only criteria for distinguishing silicate-rich crystal cumulates from solidified silicate magmas are textural. Rocks such as the coarse-grained jacupirangites and okaites (Figs. 3a, d; 3e lower half) may be interpreted as cumulates from carbonate-rich magma because they contain large, euhedral crystals of silicate and oxide minerals with interstitial space-filling carbonate. Other silicate-rich lithologies from the Oka complex are fine-grained, have little carbonate, and are either phenocrystic with poikilitic silicate minerals (fine-grained jacupirangite; Fig. 3b) or are hypidiomorphic granular in texture (melilitites and ijolite; Figs. 3e upper half, 3f, g). These rock types are interpreted to have formed from silicate-rich magmas.
Within this spectrum of rock types which may have been derived from contemporaneous immiscible silicate-rich and carbonate-rich melts, four pairs of lithologies can be chosen, based on mineralogy, as follows (Table 2, rock derived from carbonate magma given first): coarse-grained and fine-grained jacupirangites; nepheline okaites and nepheline mellilitites; okaites and mellilitites; and mellilitite-diopside carbonatite and pyroxene mellilitite. Because all of the pairs of lithologies are interpreted as products of a single magma, evidence from one pair is permissive evidence for the rest.

Immiscibility between magmas, even if not observed directly, should leave structural and chemical imprints on rocks derived from the magmas; such imprints may be used as tests for an origin by liquid immiscibility. Among the proposed imprints and tests are: textures and field relations; major element rock chemistry; trace element and light isotope chemistry; radio-isotope ratios (including radiochronology); and mineral chemistry. These tests are applied to the above pairs of lithologies from the rourc pluton, and applicable tests are satisfied in so far as data and preservation of magmatic characteristics allows. Not all of these tests are of equal value, and the limitations of each test will be discussed.

**Textures**

Immiscible melts will have a phase boundary, a meniscus, between them; the presence of menisci or relict menisci constitutes a permissive test for in situ formation as immiscible melts. Phase relations in immiscible silicate-carbonate melt systems (e.g., Freestone and Hamilton, 1980) show decreasing mutual solubility with decreasing temperature, so that cooling of immiscible silicate and carbonate melts will produce an emulsion of droplets of one melt within drops of the other melt. On cooling, such an emulsion should produce an ocellar texture.

Many rocks of the rourc pluton do exhibit ocellar textures that may reasonably be interpreted as relics of immiscible magmas. Rocks solidified from silicate-rich melt (fine-grained jacupirangite, mellilitite, iolite) commonly contain ellipsoidal ocelli (Figs. 3b, e, f, g) which are richer in calcite and coarser grained than their silicate-rich hosts. The compositions and textures of the ocelli resemble those of the carbonatites.

Evidence for silicate-rich ocelli in the carbonatites is limited. The best examples are the mellilitite stringers in cumulate okaites (Fig. 3e); okaites are interpreted as cumulates from carbonate magma. The mellilitite stringers do not cross-cut the okaites, but fit among its large crystals. The
stringers contain carbonate-rich ocelli, and are thus like other rocks derived from silicate magma. A possible interpretation of this okaite is that it formed from a carbonate magma as a cumulate of silicate crystals and silicate magma droplets.

The intimate mixture of ijolite and carbonatite in some portions of the JOMIC pluoton (Fig. 2) may be interpreted as representing large ocelli of ijolite magma in carbonatite magma, but field relations are not definitive. Rock in this area is strongly deformed (note foliation in Fig. 2 bending around ijolite; Gold, 1963, 1972), and the ijolite-carbonatite contacts are replaced by phlogopite.

Thus, rock textures and structures in the JOMIC pluoton are consistent with in situ immiscibility between silicate and carbonate magmas. Ocellar or emulsion textures are not preserved in all rock types, particularly in the carbonatites, but are common enough to suggest that liquid immiscibility was an important process in the JOMIC pluoton. The rarity in the carbonatites of textures indicative of silicate magma droplets may be ascribed to two possible effects: carbonatite magma has low viscosity (Treiman and Schedl, 1983), so silicate droplets may settle quickly; and carbonatite magma may dissolve silicate, and thus produce few immiscible droplets on cooling.

**Bulk composition**

If the bulk compositions of associated carbonatite and silicate rocks are similar to compositions of known immiscible liquids, one has permissive evidence for a liquid immiscibility relationship between the rock types (e.g., Koster van Groos, 1975a; Verwoerd, 1978; Freestone and Hamilton, 1980). This test of liquid immiscibility is difficult to apply because: igneous rocks may not retain the compositions of their parental magmas, and definitely have lost their magmatic volatiles; and comparable compositions which show immiscibility may not have been studied, either in nature or the laboratory. For the JOMIC pluoton, both problems prevent use of bulk compositions as a test of silicate-carbonate liquid immiscibility.

Rocks of the JOMIC pluoton clearly do not retain their magmatic volatiles (possibly including alkali carbonates) and many are interpreted as crystal cumulates. Examples include the coarse-grained jacupirangite (Fig. 3a) and the okaites (Figs. 3d, e). The melilitie-diopside carbonatite (Fig. 3h) may represent a magma composition, or be a calcite-rich cumulate (cf. Nesbitt and Kelly, 1977; Twyman and Gittins, 1982). Lacking magmatic inclusions, one cannot tell if the melilitie-diopside carbonatite retains a magmatic composition.

The test of bulk compositions is difficult to apply to the JOMIC rocks because no relevant experiments have been reported and no relevant natural samples have been studied. Most experimental studies of silicate-carbonate liquid immiscibility have involved feldspar-bearing synthetic systems (Koster van Groos and Wyllie, 1966, 1968, 1973; Harris and Koster van Groos, 1983), or syenitic or rhyolitic natural compositions (Koster van Groos, 1975a, b; Freestone and Hamilton, 1980). Two studies which found silicate-carbonate liquid immiscibility in systems containing ijolitic or nephelinitic magma also used Na-rich carbonate, and are inapplicable to the JOMIC rocks because their carbonate melt fractions contained 12-19 wt.% Na₂O, while the JOMIC melilites and ijolite contain only 3-5 wt.% Na₂O (Verwoerd, 1978; Freestone and Hamilton, 1980; Table 3).

Natural samples showing possible immiscibility between carbonatite and ijolite or melilitie are not sufficiently well-known for use as references in tests of liquid immiscibility. Most of these have the same status as the JOMIC rocks: unproven. A few pyroclastic rocks contain spheres of melilitie in a carbonatite matrix (LeBas, 1977; Hay, 1978; Mariano and Roeder, 1983), but necessary bulk and magma compositions are unavailable.

**Trace elements and light isotopes**

As with major elements, trace elements and light isotopes may be fractionated in characteristic patterns between immiscible melts and, thus, form a test of origin by liquid immiscibility. In principle all trace elements and light isotopes might serve as tests for immiscibility, but so far only data on rare earth element partitioning are applicable to the JOMIC rocks. Partition of Sr between immiscible silicate and carbonate melts has been studied, but only for silica-rich compositions inapplicable to the JOMIC rocks (Koster van Groos, 1975b). Light isotope ratios are available for rocks of the JOMIC pluoton (Deines and Gold, 1973); no partition data are known.

Rare earth element distributions between the silicate and carbonatite rocks of the JOMIC pluoton are consistent with derivation by liquid immiscibility, although this test is weak. The rare earth elements (REE) are partitioned into carbonate melt over coexisting silicate melt, with D(REE) ~ 2-3 (Cullers and Medaris, 1977; Wendlandt and Harris- on, 1980). Carbonatites and ijolites of the JOMIC pluoton have D(REE) ~ 5-6 (Eby, 1975; Table 3, Fig. 4). Similarity of REE distribution between the JOMIC rocks and the REE partition between known immiscible fractions may be taken as evidence of silicate-carbonate liquid immiscibility in the JOMIC pluoton (Cullers and Medaris, 1977).

But REE partition as a test of liquid immiscibility has two serious problems. First, it is not certain that any of the JOMIC carbonatites retain magmatic compositions. Second, most carbonatites in the world have similar REE content (Loubet et al., 1972) and could be interpreted as immiscible.

### Table 2. Silicate mineralogy of JOMIC rocks*, in inferred order of emplacement

<table>
<thead>
<tr>
<th>Rock Type</th>
<th>Minerals</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Tltnaeguiite</td>
</tr>
<tr>
<td>C-Jsc, P-Jsc</td>
<td>X</td>
</tr>
<tr>
<td>M-Mg, M-Mel</td>
<td>X</td>
</tr>
<tr>
<td>M-C, C, M-Mel</td>
<td>X</td>
</tr>
<tr>
<td>M-Md-C, M-Md-Mel</td>
<td>X</td>
</tr>
<tr>
<td>Ls</td>
<td>X</td>
</tr>
</tbody>
</table>

Parentheses indicate presence in some samples.

* All samples contain phlogopite.

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**References**

- Cullers and Medaris, 1977
- Deines and Gold, 1973
- Gold, 1963, 1972
- Freestone and Hamilton, 1980
- Harris and Koster van Groos, 1983
- Hay, 1978
- Koster van Groos, 1975a, b
- LeBas, 1977
- Mariano and Roeder, 1983
fractions with most ijolites. As an example, the monticellite-diopside carbonatite at Oka (see above) is younger than the ijolite pluton, has different bulk composition (e.g., Fe/Mg, Table 3), different minerals (Table 1), and different mineral compositions (see below). Yet REE data are consistent with liquid immiscibility between this carbonatite and the ijolite ijolite (Fig. 4).

**Radiogenic isotopes**

Radiogenic isotope ratios in associated silicate and carbonate rocks provide a necessary condition for an origin as immiscible liquid fractions: at the time of unmixing, immiscible fractions must have had the same ratios of parent and daughter isotopes. Identity of isotope ratios and crystallization ages merely shows that the rocks may be cogenetic, but non-identity of isotope ratios shows that the rocks were not cogenetic and could not have originated as immiscible fractions. Initial isotope ratios should be independent of crystal fractionation.

Only Rb/Sr isotope data are available for silicate and carbonate rocks at Oka, and do indicate that the rocks are cogenetic. Powell et al. (1966) found that ijolite and carbonatite from Oka (no petrography given) both have initial $^{87}$Sr/$^{86}$Sr = 0.7032, and Grunenfelder et al. (1982) report the same initial ratio for carbonatites. Preliminary analyses of silicate-rich lithologies described here (790K9, 790K10, 80B9-12) give initial Sr isotope ratios of 0.7032-0.7034.

Although further isotopic work remains to be done, available data are consistent with liquid immiscibility in the ijolite pluton.

**Mineral assemblages**

Rocks crystallized from immiscible magmas must have identical liquidus minerals, so long as equilibrium was maintained (Bowen, 1928, p. 15-16), and mineral assemblages thus constitute a test for origin as immiscible fractions. A weakness of this test is that only vanishingly small blages thus constitute a test for origin as immiscible fractures. Although this test is that only vanishingly small quantities of a mineral need be present, and might not be encountered or recognized in thin section.

Mineral assemblages in the pairs of suspect rock types (the jacupirangites; nepheline okaite and nepheline melilitite; okaite, melilitite carbonatite, melilitite; melilitide-diopside carbonatite and diopside melilitite) are essentially identical (Table 1), consistent with origins as immiscible liquid fractions. A few minerals are exceptions. Diopside in okaite and nepheline okaite occurs only as reaction rims on melilitite, and may have formed after the coexisting melilitites had solidified. Magnetite in the melilitide-diopside carbonatite is commonly rimmed by andradite + perovskite; in the ijolites this reaction has gone to completion leaving...
Fig. 4. Rare-earth element contents of Oka rocks, normalized to CI chondrites. Analyses by instrumental neutron activation. Key: melilite-diopside carbonatite, slanted hatchures; ijolite, vertical hatchure; and monticellite-diopside carbonatite, horizontal hatchure. High Yb values are probably in error.

Mineral compositions

Just as rocks crystallized from equilibrium immiscible fractions must have the same liquidus minerals, so must the liquidus minerals in each rock have identical compositions (Bowen, 1928, p. 15–16). This test has been used as evidence for silicate-carbonate immiscibility by LeBas and Handley (1979). Identity of mineral compositions provides a powerful test for origin by liquid immiscibility because: (1) minerals present are easily analyzed (e.g. by microprobe); (2) chemical analysis of complex minerals (like pyroxenes) tests many independent chemical variables; (3) minerals from crystal cumulates can be used; (4) one is not dependent on finding rare mineral grains; and (5) other petrogenetic processes (particularly crystal fractionation) are unlikely to produce magmas of differing bulk compositions with minerals of identical composition. However, a test by identity of mineral compositions is susceptible to the effects of failure to maintain magmatic equilibrium, and post-magmatic chemical reaction, re-equilibration, and alteration.

To apply the test of mineral composition, one must compare only minerals which have retained their magmatic compositions. Clinopyroxenes, melilite, and perovskite are reasonable choices, and compositions of all three may be consistent with liquid immiscibility in the JOMIC pluton. Both melilite and perovskite show evidence of post-magmatic re-equilibration, and identity of clinopyroxene compositions forms the best evidence for liquid immiscibility.

Clinopyroxene is the best JOMIC mineral to use for comparison of compositions because it retains magmatic oscillatory zoning (Fig. 5) and is known to have very low interdiffusion coefficients (Brady and McCallister, 1983; Rietmeijer, 1983). Clinopyroxenes in the JOMIC rocks include fassaite in the jacupirangites and diopside in the melilite-diopside carbonatite, diopside melilitite, and ijolite.

Fassaite compositions from the coarse- and fine-grained jacupirangites are essentially identical (Table 4; Figs. 6, 7), supporting the hypothesis that these rocks originated from immiscible melts. The pyroxenes contain oscillatory growth zones (Fig. 5a, b), and thus have a range of compositions. Compositions of pyroxenes from the melilite-diopside carbonatite, diopside melilitite, and ijolite are also essentially identical (Table 4; Figs. 6, 7), consistent with an origin as immiscible liquid fractions. A few ijolite pyroxenes are strongly enriched in Na and Fe and were not plotted on the figures; these analyses are from partially altered rocks with much phlogopite and garnet. Trace element content of pyroxenes should be as useful as their major-element composition for testing liquid immiscibility. The REE and Sc data of Eby (1973, 1975) for Oka pyroxenes are not, on the surface, consistent with liquid immiscibility between silicate and carbonate melts because pyroxenes from the carbonatite are richer in REE and Sc than those from the ijolites and mellitegites. However, all of Eby’s analyzed ijolite and mellitegite samples were strongly altered, containing 15–57% volume of alteration products (including analcime, natrolite, cancrinite, nosean and gonnardite). Trace element analyses of pyroxenes from less-altered rocks would be most useful.

Melilites in the JOMIC rocks are essentially of constant composition, and are primarily solid solutions between sodamellite (NaCaAlSi$_2$O$_8$) and akermanite components (Table 5; Watkinson, 1972; Yoder, 1973). Chemical zoning is seen as a gradual change in birefringence, with zone boundaries parallel to grain boundaries. Oscillatory zoning is not visible, although it might be present. It is more likely that melilite compositions reflect sub-solidus (and/or late magmatic) diffusional zonation. Thus, melilite compositions are consistent with liquid immiscibility in the JOMIC pluton, but weakly so because they may not be magmatic.

Perovskite may be useful in the test of mineral compositions because it can accept many substituents in readily measurable proportions (Table 6). However, perovskite in the JOMIC rocks is either unzoned or indistinctly zoned, suggesting that diffusional re-equilibration may be significant.

Perovskite composition in nepheline melilitite and neph-
Perovskites from the melilite carbonate and melilite-diopside carbonatite vary widely from sample to sample (Table 6). The best match among perovskite analyses is between the carbonatite 790K25 and the high-Nb variety in the ijolite 790K25. These samples are from adjacent units in the field, and the similarity of compositions may imply that these two rocks crystallized from contemporaneous immiscible melts, or that perovskites in the two samples equilibrated with each other after magmatic emplacement.

Perovskite in the coarse-grained jacupirangite is compositionally distinct from that in the fine-grained jacupirangite (Table 6). In neither rock is the perovskite zoned, and in both rocks it crystallized quite late. It seems reasonable that the two samples might have been out of chemical contact when perovskite grew and/or equilibrated.

Other minerals in the OOMIC pluton (including calcite, magnetite, nepheline, and pyrochlore) are not zoned, and thus may have experienced post-magmatic chemical effects.
carbonate-rich magmas. In situ immiscibility between these
radiogenic isotope ratios, REE abundances, mineral assem-
blage sequences of rocks derived from silicate-rich and
carbonatite pluton or in the magma chamber presently exposed.

Among the proposed tests of liquid immiscibility, little
faith may be placed in those that rely on bulk rock compo-
sitions, including bulk trace element abundances. Many of
the silicate-rich lithologies at Oka are crystal cumulates
from carbonate-rich magmas, and thus reflect neither the
compositions of silicate-rich melt nor carbonate-rich melt.

The carbonatites may also be crystal cumulates (cf. Nesbitt
and Kelly, 1977; Twyman and Gittins, 1982). Use of REE
abundances has an added pitfall: most carbonatites world-
wide have similar REE abundances (Loubet et al., 1972).
Thus, unrelated ijolite or melilitite and carbonatite may
have the REE distributions of immiscible magmas.

Comparison of mineral compositions is the most useful
and powerful test for origin as immiscible liquid fractions
(Bowen, 1928). Comparison of minerals with complex solid
solutions tests many independent chemical parameters, and
chemical analysis of these minerals is easy and precise with
modern methods. Mineral compositions consistent with
petrogenesis by liquid immiscibility, i.e., identical composi-
tions in rocks derived from silicate and carbonate
magmas, are extremely unlikely if the rocks are unrelated
or are related by fractional crystallization, partial melting,
assimilation, etc. Thus, if mineral compositions and tex-
tures are consistent with two rock types originating as
immiscible melt fractions, essentially no other petrogenetic
processes can be invoked.

To use mineral compositions as tests of immiscibility, the
minerals must retain magmatic compositions. Presence of
small-scale oscillatory zoning is taken as evidence of mag-
matic compositions (Fig. 5); homogeneous minerals may

Table 4. Chemical analyses of pyroxenes from the Oka complex, and
normalizations to four cations

<table>
<thead>
<tr>
<th></th>
<th>O-Jac</th>
<th>P-Jac</th>
<th>I</th>
<th>M-Mel-C</th>
<th>M-Mel-C</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO2</td>
<td>42.76</td>
<td>42.76</td>
<td>46.51</td>
<td>43.52</td>
<td>52.93</td>
</tr>
<tr>
<td>TiO2</td>
<td>4.66</td>
<td>4.66</td>
<td>3.64</td>
<td>3.96</td>
<td>0.70</td>
</tr>
<tr>
<td>Cr2O3</td>
<td>0.03</td>
<td>0.03</td>
<td>0.01</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>Al2O3</td>
<td>13.47</td>
<td>11.15</td>
<td>11.97</td>
<td>10.68</td>
<td>9.46</td>
</tr>
<tr>
<td>Fe2O3</td>
<td>7.82</td>
<td>6.01</td>
<td>6.23</td>
<td>5.63</td>
<td>4.65</td>
</tr>
<tr>
<td>MnO</td>
<td>0.13</td>
<td>0.10</td>
<td>0.09</td>
<td>0.17</td>
<td>0.15</td>
</tr>
<tr>
<td>MgO</td>
<td>9.71</td>
<td>10.87</td>
<td>10.35</td>
<td>10.69</td>
<td>14.88</td>
</tr>
<tr>
<td>CaO</td>
<td>24.66</td>
<td>24.36</td>
<td>24.42</td>
<td>24.34</td>
<td>24.95</td>
</tr>
<tr>
<td>Na2O</td>
<td>0.29</td>
<td>0.27</td>
<td>0.27</td>
<td>0.33</td>
<td>0.29</td>
</tr>
<tr>
<td>Total</td>
<td>99.29</td>
<td>98.79</td>
<td>98.77</td>
<td>99.61</td>
<td>99.64</td>
</tr>
</tbody>
</table>

The O-Jac pluton of the Oka complex includes an evol-
utionary sequence of rocks derived from silicate-rich and
carbonate-rich magmas. In situ immiscibility between these
magmas is suggested by, or consistent with, their textures,
radiogenic isotope ratios, REE abundances, mineral assem-
blage, and mineral compositions (especially clinopyrox-

Discussion

The JOMIC pluton of the Oka complex includes an evol-
utionary sequence of rocks derived from silicate-rich and
carbonate-rich magmas. In situ immiscibility between these
magmas is suggested by, or consistent with, their textures,
radiogenic isotope ratios, REE abundances, mineral assem-
blages, and mineral compositions (especially clinopyrox-

Fig. 6. Compositions of pyroxenes from the JOMIC pluton plot-
ted as molar Ca, Fe and Mg normalized to 1 cation; end members
do not represent wollastonite, enstatite and ferrosilite components.

Pyroxenes from the coarse-grained and fine-grained jacupirangites
(open and filled triangles respectively) plot together, and the py-
roxenes from the melilitie-diopside carbonatite, diopside-melilitie
and ijolite (open and closed circles respectively) plot together. Py-
roxenes from the monticellite-diopside carbonatite (open squares)
are shown for comparison.

Fig. 7. Compositions of pyroxenes from the JOMIC pluton, plotted
as ranges of minor-element contents (weight percent). Average
is shown as central line within each bar. Key: J, coarse-grained
jacupirangite; j, fine-grained jacupirangite; I, ijolite and diopside-
melilitie; C, melilitie-diopside carbonatite; L, monticellite-diopside
carbonatite.
### Table 5. Chemical analyses of melilites from the JoMrc intrusion, and normalizations to five cations

<table>
<thead>
<tr>
<th></th>
<th>Mel 79O10</th>
<th>Ok 79O14</th>
<th>Ne-Mel 79O13</th>
<th>Ne-Ok 79O8</th>
<th>IJ 79O25</th>
<th>Ne-Dt-C 8089-4</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeO</td>
<td>2.53</td>
<td>2.34</td>
<td>1.63</td>
<td>1.62</td>
<td>2.34</td>
<td>2.83</td>
</tr>
<tr>
<td>MgO</td>
<td>1.14</td>
<td>0.26</td>
<td>0.67</td>
<td>0.55</td>
<td>0.42</td>
<td>1.24</td>
</tr>
<tr>
<td>CrO</td>
<td>7.19</td>
<td>7.44</td>
<td>6.94</td>
<td>7.20</td>
<td>6.55</td>
<td>5.93</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>33.07</td>
<td>33.38</td>
<td>32.85</td>
<td>32.09</td>
<td>33.69</td>
<td>33.81</td>
</tr>
<tr>
<td>TiO₂</td>
<td>4.15</td>
<td>3.99</td>
<td>5.75</td>
<td>5.63</td>
<td>4.87</td>
<td>4.98</td>
</tr>
<tr>
<td>Nb₂O₅</td>
<td>0.00</td>
<td>0.11</td>
<td>0.02</td>
<td>0.00</td>
<td>0.05</td>
<td>0.06</td>
</tr>
<tr>
<td>Total</td>
<td>99.58</td>
<td>101.31</td>
<td>100.75</td>
<td>99.31</td>
<td>99.30</td>
<td>101.17</td>
</tr>
</tbody>
</table>

K 0.000 0.007 0.001 0.004 0.003 0.003 0.002 0.002 0.000 0.000 0.000 0.000
Na 0.364 0.348 0.494 0.489 0.494 0.430 0.526 0.479 0.491 0.445 0.479 0.337
Ca 1.600 1.607 1.556 1.539 1.619 1.613 1.507 1.562 1.521 1.484 1.521 1.642
Fe 0.096 0.088 0.072 0.061 0.128 0.116 0.078 0.121 0.106 0.073 0.113 0.109
Mg 0.005 0.010 0.025 0.021 0.025 0.019 0.016 0.019 0.048 0.059 0.047 0.041
Al 0.492 0.454 0.461 0.444 0.463 0.482 0.558 0.483 0.467 0.449 0.459 0.357
Si 1.956 1.987 1.933 1.960 1.909 1.898 1.911 1.910 1.975 2.085 2.006 2.044

Analyses by electron microprobe, crystal spectrometer. Standards included mineral and synthetic glasses. See Table 1 for samples and abbreviations.

### Table 6. Chemical composition of perovskites from the JoMrc intrusion, and normalizations to two cations

<table>
<thead>
<tr>
<th></th>
<th>C-Jac 79O9</th>
<th>F-Jac 79O12</th>
<th>Ne-Ok 79O8</th>
<th>Ne-Mel 79O8</th>
<th>Ne-Dt-C 8089-7</th>
<th>IJ 79O25</th>
<th>Ne-Dt-C 8081-2</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>0.04</td>
<td>0.06</td>
<td>0.00</td>
<td>0.04</td>
<td>0.05</td>
<td>0.01</td>
<td>0.05</td>
</tr>
<tr>
<td>TiO₂</td>
<td>53.13</td>
<td>54.68</td>
<td>51.52</td>
<td>50.72</td>
<td>27.60</td>
<td>21.07</td>
<td>26.67</td>
</tr>
<tr>
<td>ZrO₂</td>
<td>0.24</td>
<td>0.02</td>
<td>0.09</td>
<td>0.05</td>
<td>0.15</td>
<td>0.26</td>
<td>0.11</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>0.50</td>
<td>0.22</td>
<td>0.43</td>
<td>0.39</td>
<td>0.11</td>
<td>0.13</td>
<td>0.28</td>
</tr>
<tr>
<td>FeO</td>
<td>1.65</td>
<td>1.28</td>
<td>2.08</td>
<td>2.14</td>
<td>1.57</td>
<td>4.01</td>
<td>4.42</td>
</tr>
<tr>
<td>Nb₂O₅</td>
<td>0.09</td>
<td>0.05</td>
<td>0.07</td>
<td>0.07</td>
<td>0.09</td>
<td>0.22</td>
<td>0.13</td>
</tr>
<tr>
<td>CaO</td>
<td>34.56</td>
<td>36.67</td>
<td>36.10</td>
<td>35.40</td>
<td>26.46</td>
<td>22.71</td>
<td>28.19</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.16</td>
<td>0.20</td>
<td>0.38</td>
<td>0.43</td>
<td>4.14</td>
<td>4.62</td>
<td>4.30</td>
</tr>
<tr>
<td>La₂O₃</td>
<td>1.61</td>
<td>1.04</td>
<td>1.18</td>
<td>0.83</td>
<td>0.81</td>
<td>0.57</td>
<td>0.59</td>
</tr>
<tr>
<td>CeO₂</td>
<td>2.70</td>
<td>0.69</td>
<td>1.91</td>
<td>2.14</td>
<td>2.19</td>
<td>1.60</td>
<td>0.69</td>
</tr>
<tr>
<td>Nd₂O₃</td>
<td>0.73</td>
<td>0.21</td>
<td>0.61</td>
<td>0.63</td>
<td>0.55</td>
<td>0.48</td>
<td>0.30</td>
</tr>
<tr>
<td>Sm₂O₃</td>
<td>0.82</td>
<td>1.93</td>
<td>3.26</td>
<td>3.53</td>
<td>28.38</td>
<td>35.66</td>
<td>32.44</td>
</tr>
<tr>
<td>Eu₂O₃</td>
<td>0.01</td>
<td>0.00</td>
<td>0.08</td>
<td>0.12</td>
<td>1.27</td>
<td>3.27</td>
<td>1.29</td>
</tr>
<tr>
<td>Total</td>
<td>96.34</td>
<td>96.51</td>
<td>97.68</td>
<td>96.70</td>
<td>97.47</td>
<td>96.35</td>
<td>98.99</td>
</tr>
</tbody>
</table>

See Table 1 for rock types and abbreviations.

Analyses by electron microprobe, crystal spectrometer. Standards include natural and synthetic glasses and: metals for Nb, Ta, and Zr; synthetic glass (from A. Chodos) for U; and synthetic glasses (Drake and Weil, 1972) for the REE.
have magmatic compositions but should be used with caution. In rocks of the JOMIC pluton, only clinopyroxene and perovskite have small-scale zoning and are useful for testing for liquid immiscibility. However, few JOMIC rocks have zoned perovskites, and clinopyroxenes in some ijolites are unzoned and apparently re-equilibrated. In other carbonatites with less post-magmatic equilibration or alteration, other minerals may be useful as indicators of immiscibility. Apatite has been used in hypabyssal rocks (LeBas and Handley, 1979) and pyrochlore and magnetite may retain magmatic zoning (Petruk and Owens, 1975; McMahon and Haggerty, 1979).

Silicate-carbonate liquid immiscibility in the Oka complex has been suggested by many investigators (Eby, 1975; Koster van Groos, 1975b; Cullers and Medaris, 1977; Verwoerd, 1978). Their studies have each relied on single lines of evidence and have not considered textures, field relations, or the possibilities of magmatic evolution or multiple intrusions at Oka. The present study has attempted to integrate all available field, chemical, and isotopic data on the Oka complex, delineate a pluton (JOMIC) which contains silicate-rich and carbonate-rich lithologies, and apply all appropriate tests for liquid immiscibility to the rocks of that pluton. All available data are consistent with silicate-carbonate liquid immiscibility in the evolution of the JOMIC pluton.

It must be emphasized that these conclusions do not imply that carbonatites and ijolites everywhere originated as immiscible liquid fractions. In most complexes, it is clear that the carbonatites intrude the alkaline silicate rocks (e.g., Temple and Grogan, 1965; Erickson and Blade, 1963; LeBas, 1977), precluding derivation as in situ immiscible fractions. Before carbonatite and ijolite are concluded to have formed as immiscible liquid fractions, a study integrating field observations, petrography, trace and isotope chemistry, and mineral chemistry should be undertaken.

Acknowledgments

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Appendix
Sample locations
See Figure 1 and Table 1 for general locations and abbreviations for rock names. All figures listed below are to Gold (1972).

79OK8 – Me-O – Huereau Hill, Stop 2b, Fig. 5.
79OK9 – Me-Jac – Huereau Hill, 20 m SW of Stop 2b, Fig. 5.
79OK10 – Me-H – Huereau Hill, Stop 2b, Fig. 5.
79OK12 – C-Jac – Huereau Hill, 35 m E of Stop 2b, Fig. 5.
79OK13 – Me-H – Huereau Hill, 25 m E of Stop 2b, Fig. 5.
79OK14 – Hu – Huereau Hill, Stop 2b, Fig. 5.
79OK23 – Di-C – Bond Zone, Trench 9, westernmost carbonatite, Fig. 8.
79OK25 – I-J – Bond Zone, Trench 9, westernmost ijolite, Fig. 8.
79OK28 – I-J – Bond Zone, Trench 1, westernmost ijolite, Fig. 8.
80A2-8 – Di-C – Open Pit A2, SW wall of adit, upper hornblende level, Fig. 7.
80A2-40 – Di-C – Open Pit A2, Lower hornblende level, II0E from pit center, Fig. 6.
80A1-2 – Di-H – Bond Zone, Trench 1, 20 m S of W end of trench, Fig. 8.
80A9-4 – Di-C – Bond Zone, Trench 9, 7 m from SW end of trench, Fig. 8.
80A9-7 – Di-C – Bond Zone, Trench 9, 12 m from SW end of trench, Fig. 8.
80A9-12 – I-J – Bond Zone, Trench 9, 20 m from SW end of trench, Fig. 8.
80A11-3 – Di-C – Bond Zone, Trench 11, 5 m from SW end of trench, Fig. 8.