

CARBONATITES—INTO THE TWENTY-FIRST CENTURY

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INTRODUCTION

At the 1997 annual meeting of the GAC–MAC in Ottawa, a two and a half day carbonatite symposium was held in honour of John Gittins, who recently retired from the Geology Department at the University of Toronto. John, long associated with carbonatites, was co-editor with Frank Tuttle of *Carbonatites*, a book which provided a comprehensive and detailed summary of carbonatite research up to 1966 (Tuttle & Gittins, 1966). During his research career John studied a number of diverse problems pertaining to carbonatites, especially those involving mineralogical and phase equilibrium studies. His uncanny knack of homing in on some of the fundamental problems associated with carbonatite petrogenesis led to the publication of a number of stimulating papers. John is one of those rare individuals who can integrate both field and experimental studies. The paper he co-authored with Cooper and Tuttle on the calcium–alkali carbonate ternary system was one of the first papers to show the complete phase relationships among the different carbonates that could potentially precipitate out of a carbonatitic liquid at low pressures (Cooper *et al.*, 1975). Subsequently, his attention turned to problems inherent in the use of calcite–dolomite geothermometry and particularly its application to carbonatites (Gittins, 1979a). John became a staunch advocate of high-temperature dolomitic and calcitic carbonatite parental magmas, and suggested fractionation of such magmas as one way of generating alkali-rich melts at Oldoinyo Lengai (e.g. Gittins & McKie, 1979; Twyman & Gittins, 1987). His later concerns about the compositions of fluid phases accompanying carbonatite magmatism (Gittins *et al.*, 1990), led him to emphasize the anhydrous

nature of carbonatitic melts, and the role of fluorine in lowering liquidus temperatures (Jago & Gittins, 1991). Several of his papers were overviews of carbonatite and alkalic rock research, and although published some time ago, these papers still provide perceptive insights into magma generation and subsequent evolution (Gittins, 1966, 1979b, 1989). Because of the breadth of his knowledge about carbonatites, John with his incisive comments could be a formidable adversary, and he was never one to avoid a healthy debate. John's insistence on clear thinking led to the publication of several skilfully crafted critiques that kept many of us on our toes and many of us honest.

The papers in this special issue of the *Journal of Petrology* represent a cross-section of the findings presented at the Ottawa meeting and provide a comprehensive update of carbonatite research. Below are formulated five questions that we think outline the most outstanding current problems in carbonatite research and that we think provide a basis for summarizing the contributions contained in this special issue:

- (1) Are the parental melts to carbonatites derived from the lithosphere or asthenosphere, or do they represent mixtures of both?
- (2) Are carbonatites generated as partial melts derived directly from the mantle, or are they the products of magmatic differentiation of carbonated silicate melts?
- (3) How easily can such parental melts migrate through the mantle, and how do such melts affect upper-mantle chemistry and mineralogy (modal and cryptic metasomatism)?
- (4) What is the relationship among the silicate rocks, such as melilitite, nephelinite, ijolite, syenite, and phonolite, and spatially associated carbonatites?
- (5) How do carbonatite magmas fractionate and evolve?

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These questions are discussed below.

LITHOSPHERE VERSUS ASTHENOSPHERE

One of the key questions is the site of generation of the parental melts to carbonatites. Clearly, the source has to contain carbonates but whether the carbonates are primary or metasomatic is still open to debate.

The general feeling, at present, is that metasomatism is restricted only to the lithosphere, which traps fluid or melt incursions generated from the convecting asthenosphere. Several workers have made the suggestion that carbonatites are related to plume activity (e.g. Gerlach *et al.*, 1988; Bell & Simonetti, 1996), and that plume involvement can be marked by a HIMU-like signature first detected in ocean-island basalts (OIBs) (e.g. Hart, 1988) and noted in some mantle xenoliths from East Africa (Rudnick *et al.*, 1993). On the basis of isotope signatures, more than one source needs to be invoked for generating the parental melts to carbonatite. In East Africa, the two main mantle components involved in carbonatite generation are HIMU and EMI (Bell, this volume).

The relationship between plume activity and carbonatitic magmatism is strongly supported by Nd, Pb and Sr isotope data obtained from Indian carbonatites. The isotope data from both carbonatites and alkaline rocks from western India, which are coeval with or slightly younger than the Deccan volcanism, are attributed to three mantle sources (Simonetti *et al.*, this volume), of which one is lithosphere and the others are associated with Indian mid-ocean ridge basalt (MORB) and the Reunion plume. The Sung Valley carbonatites (134 Ma) in the Shillong Plateau in northeast India are similar in age to the breakup of Gondwanaland, and the enriched isotope signatures associated with these carbonatites are correlated by Veena *et al.* (this volume) with igneous and metasomatic activity related to the Kerguelen–Heard plume.

Carbonatite occurrences related to collision-type tectonic activity on a global scale are scarce, as most carbonatites are typically found in rift-related, continental settings. In an Nd, Pb, and Sr isotope study of both pre- (300 Ma) and syn-orogenic (30 Ma) carbonatites within the Indus Suture Zone of north-western Pakistan, Tilton *et al.* (this volume) point to similarities between the isotope systematics of the former and those for East African carbonatites. Tilton *et al.* suggest that the lithospheric source for the 30 Ma Pakistan carbonatites was probably transported with the Indian plate during migration from East Africa to the collision with the Asian continent.

Most carbonatites still preserve their mantle Nd and Sr isotope signatures, and many volcanic silicate rocks associated with carbonatites show little sign of contamination, unlike many plutonic rocks. Bell's review of the isotope systematics of silicate rocks from individual complexes (this volume) emphasizes the role of more than one source in generating the silicate rocks, the role of discrete partial melting events even within sources below individual volcanoes and the similarity of data between oceanic island volcanic rocks and carbonatites (e.g. Nelson *et al.*, 1988). The latter is one of the most interesting features to emerge from the Nd, Pb and Sr isotope signatures of carbonatites. In particular, complexes <200 Ma old contain isotope ratios that lie between HIMU and EMI, signatures that clearly indicate a mantle origin (e.g. Tilton & Bell, 1994). Nd and Sr isotope signatures of fenites from Oldoinyo Lengai led Kramm & Sondern (this volume) to argue that some of the silicate rocks, e.g. ijolites and phonolites, may have formed by palingenesis from crustal wall rocks by high-grade fenitization.

The isotopic evidence available at present thus suggests that the parental melts to carbonatites are generated during asthenosphere–lithosphere interaction. Whether the interaction between these two sources is solely the result of plume activity or whether other mantle perturbations, such as lithospheric delamination, can produce similar features needs to be evaluated using geochemical, geophysical and field information.

PARENTAL MAGMAS AND THEIR PRODUCTS

During the last decade, significant evidence from phase equilibrium experiments has shown that carbonatites can be generated by primary mantle melting and by the differentiation of carbonated silicate melts, i.e. liquid immiscibility and crystal fractionation.

Carbonate-rich magmas can be generated at depths >70 km by partial melting of carbonate-bearing peridotite (Wyllie & Huang, 1975). New experimental work summarized in several papers examines the production of carbonatite magmas by near-solidus melting of synthetic carbonated peridotite at 3 GPa by Moore & Wood (this volume) and at 6 GPa by Dalton & Presnall (this volume). Wyllie & Lee (this volume) observe that the composition of carbonatitic melts in equilibrium with lherzolite mineral assemblages will invariably be dolomitic, with Ca/(Ca + Mg) ratios of 0.5–0.7 at pressures from 2 GPa to at least 7 GPa.

Liquids in equilibrium with a model lherzolite containing 0.15 wt % CO₂ at 6 GPa show a systematic compositional variation with increasing temperature (Dalton & Presnall, this volume). A continuum of rock

compositions were produced ranging from near-solidus dolomitic carbonatites through ultramafic lamprophyre or kimberlite over the melting range 0–1%. Moore & Wood (this volume) illustrate that at lower pressure (3 GPa), there is a sharp compositional break from carbonatite to carbonated silicate liquid over a 50°C heating interval. These experimental data provide important insights into the production of both carbonated silicate and carbonatitic. Wyllie & Lee argue against the generation of immiscible carbonate and silicate magmas at mantle depths. However, Brooker (this volume) demonstrates that a wide two-liquid field exists at mantle pressures (2.5 GPa), suggesting that immiscibility need not be constrained to shallow levels.

Wyllie & Lee (this volume) and Lee & Wyllie (this volume) examined carbonated silicate melt differentiation. They suggest that one potential residual liquid path involves reaching the silicate–carbonate liquidus field boundary, resulting in the precipitation of silicate and carbonate phases, which could form calcic carbonatites. The recent work of Slagel (1997) in the system phlogopite–calcite at 1.0 and 2.5 GPa provides new experimental data on this liquidus field boundary, further supporting the crystal fractionation model. Other residual liquid paths may lead to a miscibility gap, with the exsolution of an immiscible carbonate-rich melt (Kjarsgaard, this volume; Lee & Wyllie, this volume).

Numerous experimental studies on the immiscibility between silicate and carbonate liquids at crustal pressures (<1.0 GPa) have been published (e.g. Koster van Groos & Wyllie, 1963; Freestone & Hamilton, 1980). In this volume, Kjarsgaard expanded the known compositional range relevant to liquid immiscibility by examining the sub-liquidus phase assemblages of an evolved, carbonated high-CaO nephelinite at 0.2 and 0.5 GPa. Importantly, 0.2 GPa experiments documented two-liquid plus melilite phase assemblages, suggesting a potential genetic link between sövites and coexisting melilite nephelinites.

The most recent carbonatite experimental studies have focused on phase relations and the major element composition of the liquid and solid phases. Relatively few studies have examined the partitioning behaviour of trace elements between immiscible silicate and carbonate liquids (e.g. Hamilton *et al.*, 1989). New two-liquid partitioning data are presented by Veksler *et al.* (this volume) for a wide range of trace elements including rare earth elements (REEs), high field strength elements (HFSEs) and other elements. The silicate and carbonate liquids were produced using a novel centrifuge autoclave technique and then subsequently analysed *in situ* with laser-ablation microprobe inductively coupled plasma mass spectrometry (ICP-MS). It appears that during immiscible separation certain elements that are

normally coherent are decoupled, and included among these are Nb and Ta.

CARBONATITIC MELT MIGRATION AND MANTLE METASOMATISM

Carbonatitic melts produced by very low degrees of partial melting in the mantle can form interconnected melts at fractions lower than 0.05 wt % in carbonated olivine-rich mantle (with grain size of the order 1 mm) because of low dihedral wetting angles and low viscosities (Minarik & Watson, 1995). Ascent rates from fluid flow calculations have been estimated at 20–65 m/s (Genge *et al.*, 1995). Such melts, however, may or may not reach the surface. Although carbonatitic melts, like silicate melts, can undergo thermal death before they reach crustal levels, they may also undergo chemical death because of their highly reactive nature. Dalton & Wood (1993) have previously shown that the high reactivity of primary carbonatitic melts can transform lherzolite to wehrlite.

Unusual trace element abundances in mantle-derived xenolith material were suggested by Green & Wallace (1988) to be the result of metasomatic activity brought about by ephemeral carbonatitic liquids. However, close comparison of data from mantle xenoliths and carbonatitic melts suggests that the story may be considerably more complicated than simple incursions of carbonatitic melts into the mantle and subsequent interaction with mantle minerals. Carbonate phases in mantle lherzolite xenoliths from Spitsbergen and Mongolia (Ionov, this volume) cannot reflect quenched carbonatitic liquids but are probably crystal phases precipitated from carbonate-rich melts. Moreover, estimates of the absolute incompatible element abundances of the metasomatizing agent cannot supply the extremely strong enrichments invoked by some models of carbonatite infiltration. Similarly, Yaxley *et al.* (this volume) discuss the role of carbonate metasomatism for a variety of mantle xenoliths from the Newer Volcanics, southeastern Australia, and conclude that this event was associated with the loss (from the metasomatized volume) of a fugitive, Si-, Al-, alkali, and large ion lithophile element (LILE)-enriched silicate melt.

THE SILICATE–CARBONATITE ROCK ASSOCIATION

Almost all carbonatites are associated with alkali silicate rocks that are usually of nephelinitic or melilititic affinity or their plutonic equivalents. Thus it is difficult to divorce the origin of one from the origin of the other. A number of distinct carbonatite–silicate rock associations occur

that include melilitite–sövite, nephelinite–sövite, pyroxenite–sövite, and olivine-rich ultrabasites–dolomitic carbonatite (Hornig-Kjarsgaard, this volume; Bell, this volume). Carbonatites, too, are spatially related to Group 1 kimberlites (Premier, South Africa) and ultramafic lamprophyres (Alno, Sweden; Kandalaksha, Russia). These diverse associations argue against any simple model for magma generation.

The relationships between carbonatites and their associated silicate rocks are complex and are still not completely understood. Whether both melts were generated from the same parental magma, or whether both were generated independently of one another, still remains one of the fundamental problems in carbonatite petrogenesis. In an attempt to resolve this problem, Harmer & Gittins (this volume), utilize Nd–Sr data to support the model that most carbonatites are of primary mantle derivation and that the divergent Nd and Sr of the associated silicate rocks reflect different mantle source regions.

A further problem that has always concerned carbonatite researchers is the timing of the emplacement of carbonatites relative to that of their associated silicate rocks (e.g. Barker, 1989). In almost all cases, emplacement of the carbonatite is later than that of most of the silicate rocks. This is shown dramatically for the Grays Bay complex, which intruded Archean rocks of the Slave structural province in northwestern Canada (Villeneuve & Relf, this volume). High-precision U/Pb ages obtained for the syenites of 2606 Ma are older by at least 6.5 my than the associated carbonatites, showing that the same plumbing system was used over an extensive period of time. Experiments, relevant to this problem (Minarik, this volume), show that carbonate melt has a higher melt–solid interfacial energy than coexisting silicate melt, thus restricting migration of the carbonate liquid either until the silicate melt had solidified or by separating from the silicate melts within liquid dominated reservoirs.

Primitive carbonated silicate magmas, such as olivine nephelinites and olivine melilitites, are able to differentiate towards more evolved liquids. Studies in this volume illustrate a variety of evolutionary paths for such fractionating liquids. Veksler *et al.* (this volume) examined mineral melt inclusions from silicate and carbonatite samples from the Gardiner complex. Results of this study support an immiscible origin for the carbonatites. Ivanikov *et al.* (this volume) found that major and trace element data for a variety of silicate dykes, which range from olivine melilitite nephelinites through to nephelinites, from Kandalaksha, Russia, are consistent with an origin via closed-system fractional crystallization. They attributed the limited volume of associated carbonatite to an origin by late-stage liquid immiscibility.

Crystal fractionation offers yet another way of generating carbonatitic liquids. The sequence of rocks, forming a perfect concentric zonation, at the Salmgorskii ring complex, Kola Peninsula, Russia, is in keeping with fractional crystallization (Korobeinikov *et al.*, this volume). As part of the study on Gardiner primary melt inclusions, Veksler *et al.* (this volume) also examined silicate and carbonatite samples from Kovdor (Russia) and propose on the basis of their findings that the carbonatites at Kovdor formed by fractional crystallization. Kjarsgaard's (this volume) examination of natural ultrabasic silicate–calciocarbonatite pairs (Premier, Blue Hills) further lends credence to the viability of generating calciocarbonatites by fractional crystallization.

Degassing can also play an important role in magma differentiation by suppressing the generation of carbonatitic liquids. Dawson (this volume) argues on the basis of detailed petrological studies on silicate volcanic rocks from Oldoinyo Lengai that an important factor in controlling magma differentiation is loss of CO₂. He suggests that CO₂ degassing inhibits exsolution of carbonatitic liquids from parental carbonated wollastonite nephelinite melts, which concomitantly concentrates alkalis, Ba, Sr and the halogens into the more evolved melt composition that generates combeite nephelinites.

FRACTIONATION AND EVOLUTION OF CARBONATITE MAGMAS

Calcitic carbonatites can be produced in a variety of ways, including direct precipitation from a primary dolomitic melt at low pressure (Fanelli *et al.*, 1986; Harmer & Gittins, 1997). Primary calciocarbonatite magmas can also be produced between depths of 70 and 40 km (Wyllie & Lee, this volume) but only if a carbonatitic melt is in equilibrium with wehrlite, produced from lherzolite during reaction with rising dolomitic melts at depths of about 70 km. Lee & Wyllie (this volume) further argue that calciocarbonatite melts produced by differentiation processes (liquid immiscibility or differentiated residual liquids) in the crust or mantle can contain no more than 80% CaCO₃. Kjarsgaard (this volume) considers the composition of the immiscible carbonate liquids produced in his experiments to be ideal analogs for sövite parent magmas, noting the high CaO contents coupled with significant SiO₂ levels required to precipitate silicate mineral phases.

An aspect of carbonatite genesis that is starting to emerge is the role of silica activity in an evolving carbonatitic liquid. Parental calcitic or dolomitic carbonatite melts generated by any method contain silicate components in solution (generally >5 wt % SiO₂). Most carbonatites contain silicate phases, including olivine, pyroxene, mica, amphibole and feldspathoids, indicating

that sufficient silica must be present in the parental liquids to precipitate such silicate phases. New experimental results (Petibon *et al.*, this volume) from 100 and 20 MPa melting on a silicate-bearing natrocarbonatite recently erupted from Oldoinyo Lengai were used to model fractional crystallization of nepheline + sodic pyroxene + melanite, leading to the generation of silicate-free natrocarbonatites.

At Dicker Willem, an Eocene age subvolcanic intrusion, the parental liquid is considered to be a nepheline-bearing sövite with between 10 and 20% SiO₂. Fractionation of sodic diopside, melanite, nepheline, magnetite, pyrochlore apatite, and Sr calcite can produce the alvikites and late-stage ferroan dolomitic carbonatites (Cooper & Reid, this volume). Hornig-Kjarsgaard (this volume) utilizes detailed carbonatite petrography and mineralogy in conjunction with REE data from mineral separates to evaluate discrete carbonatites within a number of individual complexes. Evidence can be found for single and multiple parental carbonatitic melts within individual complexes.

SUMMARY

The most important findings to emerge from the contributions contained in this volume are as follows:

- (1) Carbonatites and primitive silicate volcanic rocks are mantle derived.
- (2) On the basis of phase equilibrium experiments, carbonatitic melts can be generated by (a) primary mantle melting, (b) liquid immiscibility, and (c) crystal fractionation. Clearly, there are 'carbonatites and carbonatites'.
- (3) Integrated petrological studies support all of the three mechanisms outlined above for the generation of carbonatitic melts.
- (4) Most carbonatites are associated with silicate rocks. There are, however, a number of distinct carbonatite–silicate rock associations, which are different from one another.

These findings have helped to resolve some of the problems that were outlined and discussed in the volume *Carbonatites: Genesis and Evolution* (Bell, 1989), the last major comprehensive survey of carbonatite research. But other problems, as we have seen, are still being vigorously debated.

With the realization that carbonatites can be produced by different processes, what are now needed are criteria that can be used to separate one type of carbonatite from another. Eggler (1989) listed a set of specific criteria that could be used to recognize primary, mantle-derived carbonatites. This approach needs to be taken further. The problem is complicated by the fact that most carbonatites that are seen at crustal levels are plutonic rocks,

probably cumulates, that have lost fluids and volatiles during magma migration and emplacement. The low viscosity of many carbonatitic melts aids crystal cumulation, and further suggests that even extrusive carbonatites may not approximate liquid compositions (Le Bas *et al.*, 1997). Because of these complexities, it is very likely that few carbonatites have chemical compositions that closely approximate those of their parental liquids. The identification of parental carbonatite magmas and their differentiation paths could be addressed by analysis of mineral phases using laser ablation ICP-MS, PIXE or conventional methods on mineral separates, in the hope that such results may help to more closely identify the different paths that are taken during magmatic evolution. There is a clear need too for further experimental data on the precipitation of various mineral phases from carbonatitic melts.

The intimate relationships of both silicate and carbonate rocks in most carbonatite complexes require that petrogenetic studies of both suites are needed to fully evaluate their origins. The complexities shown by the silicate rocks alone are well illustrated by the isotopic disequilibrium documented between phenocryst phases and their host rocks (e.g. Simonetti & Bell, 1993). In addition, the recognition of a number of distinct carbonatite–silicate rock associations demands that each complex be evaluated on its own merits. Integrated studies, including detailed field mapping, mineralogy, chemical and isotope studies of individual complexes should help reveal the way in which different carbonatites can form.

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