

## Myth and reality in the carbonatite - silicate rock “association”

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**ABSTRACT.** — The very common «association» of carbonatite with alkalic silicate rocks is generally assumed to be a genetic relationship in which carbonatite was derived from silicate magma, or that both groups of rocks shared a common parent magma. It has become a dominant theory of carbonatite genesis on which much experimental petrology study has been based. We suggest that this assumption is unwarranted and misleading. The «association» is spatial rather than genetic and is caused by two separately generated magmas having used the same system of conduits to reach the crust from their generation sites in the mantle. Experimental petrology, field relations and isotope studies do not definitively confirm any mode of origin to the exclusion of others. Experimental petrology allows the development of carbonatites (largely as cumulates) by fractional crystallization of silicate magma and, under some conditions, of carbonatite magma derived by liquid immiscibility from a silicate magma, but it also allows the development of carbonatite magma by partial melting of carbonate-bearing mantle peridotite, unaccompanied by silicate magma generation. Most isotope data give no definitive proof that silicate magma is parental to carbonatite magma, - nor are they inconsistent with such a relationship. Furthermore, where the data are consistent with a derivative relationship they also show, quite definitively, that not all of the silicate rocks could be parental to carbonatite. However, there are South African and Zimbabwean carbonatite complexes in which isotopic data show that at least some carbonatites were generated at considerably greater

depths than the silicate rocks that accompany them in the same intrusive complex, thus ruling out a directly derivative relationship between carbonatites and silicate rocks, either by fractionation or by liquid immiscibility. We believe that, in general, there is no liquid line of descent relationship between silicate magma and carbonatite magma, except in very minor amounts. It would be more meaningful to refer to carbonatites and their «accompanying», rather than to their «associated» alkalic silicate rocks.

**RIASSUNTO.** — L'associazione, molto comune, di rocce carbonatitiche e silicatiche alcaline ha, generalmente, correlazioni genetiche. Ne deriva che le carbonatiti derivano da magmi silicatici, oppure che entrambi i due gruppi di rocce hanno un comune magma capostipite. Questa teoria sulla petrogenesi delle rocce carbonatitiche è diventata dominante ed è stata supportata da molti studi di petrologia sperimentale. Noi suggeriamo che questa assunzione è non garantita ed è fuorviante. L'«associazione» ha una connotazione spaziale piuttosto che genetica, ed è causata da magmi che hanno usato lo stesso sistema di condotti per risalire attraverso la crosta dalla loro sorgente mantellica. La petrologia sperimentale, le relazioni spaziali e gli studi isotopici non confermano inequivocabilmente nessun modo di origine che escluda gli altri. La petrologia sperimentale permette la formazione di carbonatiti (prevalentemente come cumulati) dalla cristallizzazione frazionata a partire una magma silicatico ed, in particolari condizioni, di carbonatiti da immiscibilità allo stato liquido da un magma silicatico, ma permette anche la formazione di un magma carbonatitico per fusione parziale da

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mantello peridotitico contenete carbonato, non accompagnato però dalla formazione di magma silicatico. La maggior parte dei dati isotopici non offrono una prova definitiva che i magmi silicatici siano consanguinei con i magmi carbonatitici – ma non sono neanche inconsistenti con questo tipo di relazione. Inoltre, laddove i dati indicano questo tipo di relazione derivativa mostrano anche, abbastanza definitivamente, che non tutte le rocce silicatiche possono essere imparentate alle carbonatiti. Per contro, ci sono complessi carbonatitici del Sud Africa e dello Zimbabwe in cui i dati isotopici mostrano che almeno alcune carbonatiti si sono formate a una profondità considerevolmente maggiore delle rocce silicatiche che le accompagnano nello stesso complesso intrusivo, escludendo quindi una diretta relazione derivativa tra carbonatiti e rocce silicatiche, sia attraverso processi di cristallizzazione frazionata, che immiscibilità allo stato liquido. Noi crediamo che, in generale, non ci sia una linea di discendenza allo stato liquido tra magma silicatico e magma carbonatitico, fatta eccezione per piccoli quantità. Sarebbe quindi più corretto riferirsi alle carbonatiti ed i loro magmi alcalino silicatici che le «accompagnano» piuttosto che vi sono «associati».

**KEY WORDS:** *Carbonatite - alkalic rock association, carbonatite genesis, field relations, experimental data, isotopic data.*

## INTRODUCTION

Carbonatites occur commonly, but by no means universally, in the company of a wide variety of feldspathoidal igneous rocks and pyroxenites, and the term «carbonatite - alkalic silicate rock association» has become firmly established. It is widely assumed that there is a derivative relationship in which silicate magma is parental to carbonatite magma, but a wide variety of hypotheses have been summarised by: Kjarsgaard and Hamilton (1989), Wyllie (1989), Brooker (1998), Harmer and Gittins (1998), Lee and Wyllie (1998), Wyllie and Lee (1998), Bart (1999).

How has this concept become so firmly entrenched as to be the principle on which much of the experimental approach to carbonatite genesis has been based? The answer is to be found in the history of

carbonatite studies, where a magmatic origin of carbonatite was not widely accepted until the late 1950s. This was also a period when little was known about the composition and mineralogy of the mantle and, consequently, of magma genesis; therefore, when the idea of a carbonatite magma became accepted there was almost no parallel concept of where it might have come from. For a long time there was no clear acceptance that the magmas originated in the mantle.

It was natural at first to think of carbonatites as originating from crustal limestone. That this was not a viable option came both from Sr isotope studies, beginning in the 1950s, and also much earlier from the careful mineralogical observations of petrologists such as Campbell Smith who noted that carbonatites carry a suite of minerals quite unlike any known limestones. This was later supported by geochemical analytical studies that again emphasized their unique compositions. Yet nobody had any clear idea of how so much carbonate (largely calcite and dolomite) could come from an igneous source.

Given this dilemma, it is hardly surprising that field mapping led petrologists to the obvious fact that carbonatites occur commonly, although not universally, in small intrusions accompanied by alkalic silicate rocks. This led inevitably to the assumption that the two rock types constitute a single magmatic system, however different they may be mineralogically and chemically, and that they are related through a liquid line of descent, or at least share a common parental magma. This theme is common in igneous petrology textbooks and in other carbonatite writings, typical examples being:

«Carbonatites are habitually associated, both at the regional scale and at individual eruptive centres, with nephelinites and allied ultrabasic alkaline rocks such as ijolite and alkali pyroxenite. .... The logical inference is that the carbonatite and silicate magmas, in spite of their contrasted compositions, must be genetically related». (Carmichael *et al.*, 1974, p. 520);

«... granting that this silicate - carbonate assemblage represents a unified magmatic system, how and where did the parent magma form .... ?» (Best, 1982, p. 205);

«The association, both regionally and locally, of carbonatites with alkali-rich, silica-poor igneous rocks implies a strong genetic relationship.» (Ehlers and Blatt, 1982, p. 242).

«The continuity between the mineralogy of the carbonatites and that of the silicate rocks supports the suggestion that they have been derived from the latter.» (King, 1965, p. 96)

Thus, it became axiomatic to view carbonatites and accompanying alkalic silicate rocks as products of a single parent magma, and the «carbonatite - alkalic silicate rock association» was born.

However, their mere coexistence is not a convincing reason for assuming that they were born of the same parent at the same time and in the same place. They are very different rocks both chemically and mineralogically. Many carbonatites are unaccompanied by silicate rocks, and vastly more intrusions of alkalic silicate rocks are unaccompanied by carbonatite. The genetic 'association' seems to us to be a false deduction.

#### DISCUSSION OF THE ARGUMENTS ADVANCED IN SUPPORT OF A GENETIC ASSOCIATION

The arguments draw upon experimental studies, field relations, and isotope geochemical data, - but selectively! Indeed, not only are many of the supporting arguments weak, but some are internally inconsistent, self contradictory, or involve circular reasoning.

#### *Experimental data*

Design of experiments to study carbonatite origin has been guided extensively by the assumption of a genetic «association». Most experimental studies assume that CO<sub>2</sub> - rich mafic, alkalic, silica-undersaturated silicate magma develops into one of the following: (i) a

carbonatite magma; (ii) a silicate magma capable of crystallizing carbonates which then accumulate to form carbonatite; or (iii) a magma that eventually reaches a solvus and splits into two immiscible liquids, - one being carbonate-rich and the other silicate-rich.

Some experimental results allow for the crystallization of calcite, and/or dolomite, from silicate liquids (e.g. many of the studies of Lee and Wyllie), - a process that would lead to carbonatites being largely cumulate rocks as a result of fractional crystallization. There is also experimental support for the immiscible development of some types of carbonate liquid from silicate liquids in the crust. Immiscibility in the mantle, however, is a dubious possibility. It is argued against by Lee and Wyllie (1996, 1997, 1998) but supported by Brooker (1998), each assuming strikingly different proportions of CO<sub>2</sub> at the melting site.

Thus, there is experimental support for carbonatite magma being generated either by fractionation of a silicate magma or by liquid immiscibility, the latter occurring most likely in the crust rather than the mantle. However, there is a danger implicit in either process for each requires development of the two magmas at the same time and in the same place. They are theories that both explain and require carbonatites and silicate rocks to occur together in a single intrusive complex, and they contain more than a hint of circular reasoning.

In contrast to these studies, other experiments show that carbonate liquid can be generated by partial melting of carbonate-bearing mantle peridotite without any development at all of alkalic silicate magmas. Similarly, alkalic silicate rocks can be generated without any involvement of carbonate liquids. Thus, carbonate and silicate magmas can be generated in the mantle quite independently of each other. They are likely to utilise the same conduit or «plumbing system» to rise from their birth place into the crust where they will crystallize in a very limited volume, so forming an intrusive complex in which carbonatite and alkalic silicate rocks are close neighbours. This does not, however,

necessarily mean that they are consanguineous. Such «separate development» has been advocated by Harmer and Gittins (1997, 1998); Harmer et al. (1998); and Harmer (1999).

In summary, experimental petrology alone does not unambiguously prove any one of the following:

(i) fractional crystallization of a carbonate-bearing alkalic silicate magma to generate a carbonatite cumulate;

(ii) liquid immiscibility;

(iii) independent development of carbonate and silicate magmas within the mantle, some of which are able to reach the crust and crystallize there

There is evidence for all three, and the operation of one would not negate the validity of the others.

#### *Field (spatial) relations*

In complexes containing carbonate rocks and silicate rocks it is usually the carbonate rocks that have intruded the silicate rocks, and from this a genetic association has been deduced. The case has been put very clearly by Barker (1989, pp. 52-53) and may be summarised thus:

«Times of emplacement of carbonatites, relative to associated silicate rocks in the same complex, are fairly consistent. In general, the sequence from periphery to core, and from oldest to youngest, is nepheline syenite (if present), to nepheline-clinopyroxene rocks to carbonatites»

but:

«Carbonatite liquids have such low solidus temperatures, densities, and viscosities that they should form earlier, and ascend faster, than any cogenetic silicate magma. Nevertheless, carbonatites are emplaced later than most of the silicate rocks with which they are associated.»

and

«The tardiness of carbonatite liquids in arriving at the level where their products are observed, strongly suggests that they have not come all the way from the upper mantle as independent and

primary liquids (which should form early and rise rapidly), but have separated from parental carbonated silicate liquids closer to the surface.»

In short, the two magmas must have developed almost simultaneously in the crust so that the more buoyant carbonatite magma would not have very far to travel and could more readily intrude the silicate rocks.

A popular corollary has been that liquid immiscibility is the most feasible method by which this could happen, and so its proponents have embraced field relations to narrow down the possibilities allowed by experimental petrology. They argue for liquid immiscibility at crustal depths operating on a mantle-derived magma.

Their case is unconvincing, for if the carbonatite in the «association» is not derived from a silicate magma the problem vanishes. The field relations require crustal liquid immiscibility only if the carbonatite-silicate rock 'association' is a genetic one, - and that is largely an assumption. Theirs is a circular argument in which the proof requires the assumption, and the assumption provides the proof!

One also needs to examine the extent to which the low viscosity and density of carbonatite magma governs its rate of rise. Is the buoyancy argument valid at all? We can accept that carbonatite magma has relatively low density and viscosity. We also know that minerals crystallizing from carbonate-rich liquids sink readily during sealed capsule experiments. However, these are static experiments in which crystals sink at most a few millimetres through a stationary liquid. It is fallacious to extrapolate from these experiments to a rising magma. While the density and viscosity of a carbonate liquid are undoubtedly low, the liquid soon begins to crystallize as it rises, and so is no longer simply a liquid. The viscosity of a crystal mush with interstitial liquid is vastly greater than that of a carbonatite liquid, and such a magma will encounter considerable frictional resistance with the walls of the cracks and passageways through which it is moving. It is simply not safe to assume that a carbonatite magma rises

through mantle and crust more rapidly than does a silicate magma.

#### *Isotopic compositions: previous studies*

There are extensive isotope data on carbonatites and accompanying silicate rocks from a wide range of localities and ages. Bell (1998) has surveyed the «association» and finds no preponderance of evidence in favour of one particular theory of carbonatite and alkalic silicate rock genesis to the exclusion of other possibilities. The data neither support nor refute, with any certainty, the development of carbonatite magma from silicate magma, but neither are they inconsistent with such processes. They do, however, impose the severe limitation that even if carbonatite magma can be developed from silicate magma, it could only be from some of the silicate magmas, - not all of them. This is because «The silicate rocks ... show a much greater variation in isotopic ratios than their associated carbonatites» (Bell, 1998, p.1995). Consequently, Bell (1998, p. 1992) has suggested that «... each intrusive event involved distinct mantle-derived melts...». Thus, while most of the isotopic data assembled so far, offer limited support for a derivative association of carbonatite and silicate rocks, they clearly rule out any universal relationship.

#### *Isotopic compositions: newer data*

Data on the dolomitic Dorowa and Shawa carbonatite complexes in the Buhera district of south eastern Zimbabwe (Harmer *et al.*, 1998) show that the carbonatites and accompanying nephelinites represent magmas that were derived from different portions of the sub-cratonic mantle of southern Africa, and that the Shawa carbonatites must have been derived from a greater depth than the silicate rocks. Data from the Spitskop complex of South Africa lead to similar conclusions. In the Buhera and Spitskop complexes the carbonatite and silicate rocks can not have been derived from a single parental magma by either

fractional crystallization or liquid immiscibility. The only feasible alternative is the development of discrete magmas as a result of completely separate partial melting events. Carbonatite magma, discrete from any silicate magma, must have existed in the mantle at depths greater than those at which nephelinitic magmas were being developed.

#### FURTHER PROBLEMS WITH THE GENETIC ASSOCIATION CONCEPT

There are additional problems that the genetic association concept fails to explain. There are many carbonatite intrusions that are either completely devoid of accompanying silicate rocks or in which silicate rocks form only a minuscule part (Harmer and Gittins, 1997). These «carbonatite-only» complexes invariably invite comment about the lack of accompanying silicate rocks. It seems to be assumed that carbonatite - silicate intrusive complexes fit an easily explicable pattern whereas the «carbonatite-only» complexes are a mystery and may even have an entirely different origin. More commonly, they have simply been ignored.

Then we must consider the fact that feldspathoidal silicate rock intrusions in which there is no carbonatite greatly exceed the number of carbonatite-alkalic silicate rock complexes, and yet this absence of carbonatite is not considered odd or remarkable. Furthermore, at least in the miassic intrusions, the feldspathoidal and other alkalic silicate rocks are very similar to those in carbonatite - silicate rock complexes. The commonest rocks in both types are (a) in the plutonic complexes: nepheline syenites (under many names) and the jacupirangite - melteigite - ijolite - urtite series; and (b) in the volcanic complexes: nephelinites and phonolites. Why should alkalic silicate rock complexes without carbonatite seem normal while carbonatite complexes without silicate rocks are considered unusual? Is an alkalic rock complex without carbonatite an anomaly, - one that failed to develop

carbonatite, - or do alkalic silicate rocks and carbonatites not have a derivative relationship?

If the two fundamentally different types of carbonatite complex (carbonatite only, and carbonatite - silicate rocks intrusions) are given their due recognition, then the «association» concept fails to explain a large number of carbonatite complexes, and different schemes of carbonatite genesis are required. Carbonatites in one type of intrusion could not be formed in the same way as carbonatites in another type. While possible, this seems unlikely and unnecessary. But what is beyond doubt is that the concept of a genetic association can not explain both types of carbonatite complex.

#### CONCLUSIONS

At the outset we questioned whether it is reasonable to deduce, from close proximity in the field alone, that carbonatites and their accompanying silicate rocks are derived one from the other or jointly derived from a common parent magma. We also commented on the paucity of convincing evidence in support of the contention.

The conflicting hypotheses of carbonatite magma genesis have troubled enough petrologists that a theme, paraphrasing H. H. Read's famous dictum, has begun to emerge suggesting that there are carbonatites and carbonatites. Bell *et al.* (1998) have cautioned that «... the recognition of a number of distinct carbonatite - silicate rock associations demands that each complex be evaluated on its merits.». While this has the comfort of a truism, it also contains a hint of what might be called Desperation Petrology for, while it would be unwise to suggest that all carbonatites formed in identical fashion, the fact remains that there is very little that «separate development» does not explain. The derivative relationship of the «genetic association», however, leaves a great deal unexplained. Indeed, there have always been hints of doubt even in the same literature that has supported the «genetic association».

For example:

«Although carbonatites are habitually associated with alkaline mafic rocks, the two types - the one a carbonate, the other a silicate phase assemblage - tend to retain sharply their respective identities. There is nothing in mutual relationships in the field to indicate derivation at the present site from a common parental magma ....» (Carmichael *et al.*, 1974, p. 519).

«Now that the magmatic nature of carbonatites is generally recognized, there are still alternative genetic models. In one, carbonatite and nephelinite magmas are independently generated, although perhaps at much the same level of the mantle.» (Carmichael *et al.*, 1974, p. 522).

The idea of separate magma development is not new, but experimental verification of its feasibility came well after 1974.

We suggest that the data from the Buhera and Spitskop carbonatite complexes are applicable fairly widely. Indeed, the wide variability of isotopic composition in the silicate rocks of most complexes, and their partial overlap with carbonatites, is readily explicable by separate development of carbonate and silicate magmas, but not by other methods of genesis.

Experimental petrological study of carbonatite genesis has been hampered by assuming a consanguineous relationship between carbonatite and accompanying silicate rocks. For example, experiments designed to explain the unique chemical concentrations of certain elements in carbonatites have been based on the dual assumptions of the 'genetic association' and of liquid immiscibility, and so they have produced data only on element partitioning between immiscible carbonate and silicate liquids. If liquid immiscibility is not the means by which carbonatite magmas are generated these data are of very little value. Furthermore, the carbonate liquid employed in the experiments is far more alkali-rich than any acceptable carbonatite magma in Nature, with the exception of the natrocarbonatite of Oldoinyo Lengai. In similar vein, the large body of published phase equilibrium data on carbonate and silicate rocks does not, in

general, explain the genesis of carbonatites. What it does explain is the almost universal presence of small amounts of calcite in most alkalic rocks. However small an amount of CO<sub>2</sub> there is in a silicate magma it is inevitable that it will eventually concentrate to the stage where calcite can crystallize. The extrapolation from this to a body of carbonatite is a long one indeed.

The existence of a genetic 'carbonatite - alkalic silicate rock association' has been assumed rather than proved conclusively. We believe that, in general, this association is a purely spatial one caused by separately-developed magmas employing the same system of conduits to rise from the mantle into the crust. In the Spitskop complex of South Africa and the Buhera complexes of Zimbabwe there can be no possibility of carbonatite magma being derived through fractionation or liquid immiscibility from the silicate magma that produced the alkalic silicate rocks. The two magmas must have developed separately, but the data are more widely applicable.

Carbonatite magma is generated by partial melting of carbonate-bearing peridotite within the mantle and this is independent of any silicate magma generation which might or might not occur. Liquid immiscibility is, in general, inconsistent with the isotopic geochemistry of carbonatites. Uncritical acceptance of it as a viable means of carbonatite magma generation has led to much clouding of carbonatite study. Because the term «association» has taken on a strong genetic connotation that is now almost ineradicable, it would be better to refer to carbonatites and their «accompanying» rather than «associated» silicate rocks.

#### ACKNOWLEDGMENTS

We are grateful for support from the Foundation for Research Development, the Council for Geoscience in South Africa, and the Natural Science and Engineering Research Council of Canada. JG is particularly grateful to Professor Lord Lewis, former Warden of Robinson College, Cambridge, and the College Council for the tenure of a Bye Fellowship

on several occasions, and to the Department of Earth Sciences in the University of Cambridge for sabbatical leave facilities

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