

CARBONATITES AND CARBONATITES AND CARBONATITES

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ABSTRACT

Carbonatites are redefined using a mineralogical-genetic classification and divided into two groups: primary carbonatites, and carbothermal residua. Attention is drawn to the fact that *carbonatite* is both a petrographic term applicable to a particular rock-type as well as a group name applied to a complex of related carbonate and silicate rocks in a magmatic or extrusive complex. Primary carbonatites, in terms of mineralogical-genetic classifications, rather than simple modal classifications, can be divided into a group of *bona fide* magmatic carbonatites formed from diverse mantle-derived magmas, *i.e.*, carbonatites associated with the melilitite, nephelinite, aillikite and kimberlite clans, with the latter best being termed calcite kimberlites. Each magma type and associated carbonatites are considered to be genetically distinct, and formed at different depths in the upper mantle by different degrees of partial melting. Carbonatites associated with the melilitite and nephelinite clans can have a multiplicity of origins, and may be formed by fractional melting, fractional crystallization or liquid immiscibility. Calcite kimberlites are small-volume late-forming differentiates that are not related to other carbonatites or their parental magmas. The origin and genetic relationships of the Oldoinyo Lengai natrocarbonatite cannot be unambiguously determined, although these rocks are regarded as a distinct variety of primary carbonatite. Carbonate-rich rocks associated with diverse potassic or sodic peralkaline saturated to undersaturated magmas derived predominantly from metasomatized lithospheric mantle, together with REE-carbonate-rich rocks of undetermined genesis, are best termed carbothermal residua rather than carbonatite. There can be mineralogical (or modal) convergence between these rocks and low-pressure REE-rich derivatives of *bona fide* primary carbonatites. Carbonate-rich rocks formed by pneumatolytic reactions or anatectic melting of crustal rocks should not be considered to be carbonatites.

Keywords: carbonatite, melilitite, nephelinite, ijolite, melilitolite, aillikite, nepheline syenite, liquid immiscibility, carbothermal residue, upper mantle.

SOMMAIRE

Le clan des carbonatites est ici redéfini dans le contexte d'une classification minéralogique et génétique, et subdivisé en deux groupes: carbonatites primaires et résidus carbothermaux. On souligne le fait que "carbonatite" est à la fois un terme pétrographique applicable à un type de roche particulier, et un terme faisant référence à un groupe de roches, et appliqué à des roches carbonatées and silicatées génétiquement liées dans un complexe magmatique ou extrusif. On peut diviser les carbonatites primaires, en termes de classifications minéralogiques et génétiques plutôt que de simples classifications modales, en un groupe de carbonatites véritablement magmatiques formées à partir de divers magmas mantelliques, *i.e.*, carbonatites associées aux clans de mélilitite, néphéline, aillikite et kimberlite, avec ces dernières manifestations préférablement appelées kimberlites à calcite. Chaque type de magma et les carbonatites associées seraient génétiquement distincts, et formées à des profondeurs différentes du manteau supérieur selon des taux distincts de fusion partielle. Les carbonatites associées aux suites de mélilitite et de néphéline peuvent avoir une multiplicité d'origines, impliquant possiblement fusion fractionnée, cristallisation fractionnée ou immiscibilité liquide. Les kimberlites à calcite seraient des différenciés tardifs à volume limité qui ne sont pas apparentées aux autres carbonatites ou à leur magmas parents. L'origine et les relations génétiques de la natrocarbonatite de Oldoinyo Lengai ne peuvent pas être précisées sans ambiguïté, quoique ces roches sont considérées comme variété distincte de carbonatite primaire. Les roches riches en carbonates associées aux divers complexes hyperalkalins potassiques ou sodiques saturés ou sous-saturés et dérivés surtout à partir du manteau lithosphérique ayant subi une métasomatose, de même que les roches enrichies en carbonates de terres rares, d'origine indéterminée, seraient plutôt des résidus carbothermaux que des carbonatites. Il pourrait y avoir convergence minéralogique (ou modale) entre de telles roches et les produits véritables à faible pression de carbonatites primaires enrichis en terres rares. On ne devrait pas inclure les roches riches en carbonates formées par réactions pneumatolytiques ou par anatexie de roches de la croûte dans le clan des carbonatites.

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Mots-clés: carbonatite, mélilitite, néphéline, ijolite, mélilitolite, aillikite, syénite néphélinique, immiscibilité liquide, résidu carbothermal, manteau supérieur.

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INTRODUCTION

The rocks now known as carbonatites were originally described by Bose (1884) from the Lower Narbada Valley of India, but it was not until the investigations of Högbom (1895) at Alnö, Sweden, and of Brögger (1921) at Fen, in Norway, that a magmatic origin was postulated for the carbonate-bearing rocks found in these alkaline complexes. Not everyone agreed with this concept; in particular, the highly influential petrologists Reginald Daly (1933) and James Shand (1943) remained adamant that these “igneous limestones” were merely megaxenoliths of sedimentary material. Significantly, the widely used glossary “A Descriptive Petrography of the Igneous Rocks” by Johannsen (1938) did not include carbonatite as a rock type, although associated exotica such as “tveitåsite” were included! In his explanation for excluding rocks with a large percentage of carbonate, Johannsen (1938, page v) stated that we may “dismiss these carbonate-bearing rocks as non-igneous”.

This petrological divide remained until the seminal experimental work of Wyllie & Tuttle (1960), who showed that calcite could crystallize as a liquidus phase at temperatures as low as ~650°C at 0.1 GPa. This study sounded the death knell of the limestone-syntexis hypothesis (Shand 1943) for the genesis of undersaturated alkaline rocks, and the work ushered in a decade of renewed interest in carbonatites in general (Heinrich 1966, Tuttle & Gittins 1966), highlighted by the discovery of the natrocarbonatite lavas at Oldoinyo Lengai, Tanzania (Guest 1956, Dawson 1962). In the 1950s and 1960s, many new localities of carbonatite were described, but no real advances in understanding their genesis were made. During the subsequent 40 years, carbonatite studies have progressed significantly in only two areas; experimental and isotopic studies. Experimental studies of synthetic carbonated upper mantle source-rocks have led to insights into the genesis of primary carbonatitic magmas, and those of carbonated nephelinite compositions heralded the promotion of liquid immiscibility as a potential genetic process (Koster van Groos & Wyllie 1963, Kjarsgaard & Hamilton 1989, Kjarsgaard 1998). Isotopic studies have confirmed that asthenospheric or lithospheric mantle sources (or both) have played a role in the genesis of many carbonatites but have only served to confuse further their relationships to associated silicate rocks. Thus, these data show quite clearly that carbonatites could have come from the same sources as some of the associated silicate rocks, but that equally clearly, not all silicate rocks could have the same origin as the carbonatites (Bell 1989, Bell & Blenkinsop 1989, Gittins & Harmer 2003). Although these studies advanced some aspects of our knowledge of carbonatite genesis, there has been little substantial progress in our understanding of the origins of large bodies of intrusive calcite carbonatite such as are found at the Fen complex. In fact,

the genetic questions highlighted by Heinrich (1966) nearly 40 years ago are as unresolved today as when initially proposed, and not significantly different from those listed in the recent summaries of Bell (1998) and Harmer & Gittins (1997, 1998).

Why has there been no substantial progress in our understanding of carbonatites? In part, this might be due to a consideration of *all* carbonatites as a single rock-type or of a derivation from a common parental magma, rather than as a diverse group of rocks with a multiplicity of origins. One objective of this paper is to consider the latter hypothesis. Another reason is that there are very few studies of carbonatite complexes combining geology, mineralogy and isotope geochemistry into an integrated petrogenetic scheme. A major hindrance to any genetic study is the paucity of extrusive carbonatites that might be representative of parental magmas. In this context, vigorous debate has raged regarding the role of the unique natrocarbonatites of Oldoinyo Lengai and their relationships to plutonic calcite carbonatite. For example, compare the antithetical views of Le Bas (1981) and Twyman & Gittins (1987).

Not all of the concepts outlined in this work are necessarily new. Thus Gittins (1978) has noted that “under certain circumstances small amounts of carbonate can crystallize from a wide variety of magmatic compositions” and criticized the “tendency to refer to any such carbonate as carbonatite” (Gittins 1978, p. 110). Subsequently, Gittins & Harmer (2003, p. 24–25) have commented that “no matter how small an amount of CO₂ 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.” These comments have direct relevance to some of the “carbonatites” discussed below.

This work is a development of these and other concepts, but there are **no** claims that it is a definitive study of the origins of carbonatites. It is presented to stimulate and renew discussion of the overall problem of “carbonatite” petrogenesis. The approach taken is to regard not all “carbonatites” as petrographically synonymous but as petrologically distinctive rocks with respect to their antecedents and descendants.

CARBONATITES DEFINED

Carbonatites are defined in the IUGS system of classification as igneous rocks composed of more than 50 modal per cent primary (*i.e.*, magmatic) carbonate (*sensu lato*) and containing less than 20 wt.% SiO₂ (Le Maitre 2002). Varieties of carbonatite are named on the basis of the dominant carbonate mineral, *e.g.*, calcite carbonatite, dolomite carbonatite, *etc.* (Woolley & Kempe 1989). The IUGS system is *non-genetic*. Whereas it is useful in the classification of common rock-types, it is totally inadequate for more exotic rock-types such as kimberlite, lamproite, orangeite, and

diverse undersaturated potassic volcanic and plutonic rocks. For these types, Mitchell (1995) and Woolley *et al.* (1996) have devised mineralogical-genetic classifications that are based upon the principle that a spectrum of modally diverse yet genetically related rocks can be derived from a single magma-type. They can be recognized by the presence of typomorphic or characteristic assemblages of major, minor and accessory minerals. Examples are the kimberlite and lamproite clans (Mitchell 1995). In this type of classification, it is *not* necessary to know the origin of a particular magma-type. Indeed, genetic hypotheses advanced for a particular magma-type will change depending upon the current petrological paradigm or the way the empirical petrological and mineralogical data are interpreted. One important aspect of mineralogical-genetic classifications is that genetically unrelated but modally similar rocks can be derived from a variety of parental magmas.

The deficiencies of the IUGS system are particularly apparent in the definition of carbonatite. Regardless of origin, rocks composed of more than 50 modal per cent carbonate that can reasonably be inferred to be of igneous origin are to be called carbonatite. Thus, the term can embrace a spectrum from plutonic calcite- or dolomite-rich rocks, to calcite veins formed by hydrothermal processes in the late stages of crystallization of many types of magma, to the “albite-carbonatite” veins of the Great Glen in Scotland (Garson *et al.* 1984) or even the late veins of calcite in the Calton Hill analcime basanite (Tomkeieff 1928). Such grouping of modally similar but genetically diverse rock-types into a single pigeon-hole inevitably leads to the formulation of false and misleading petrogenetic hypotheses; a prime example is provided by the alleged “kimberlite – carbonatite – alkaline rock” relationship that was discredited by Mitchell (1979, 1986).

In this paper, carbonatites (*sensu lato*) are defined as any rock containing greater than an *arbitrary* 30 vol.% primary igneous carbonate regardless of silica content. The objective of this definition is to recognize that a carbonatite-forming magma will, through differentiation, generate a suite of genetically related rocks in which the carbonate content varies significantly. Thus, the precise amount of carbonate present is less important than the fact that a suite of carbonate-bearing rocks is derived from same magma. It recognizes that in the field, there is a wide variation in modal percentages of the constituent minerals over small distances, and that a given geographic domain can consist of a wide variety of genetically related rock-types. Thus in these terms, a rock composed of 30 vol.% calcite and 70 vol.% pyroxene is no less of a carbonatite than a close neighbor that has 51 vol.% calcite and 49 vol.% pyroxene. Many plutonic carbonatite complexes display such modal variation on a centimeter-to-decimeter scale. For example, at the Fen complex (Brøgger 1921, Andersen 1988), there is a spectrum of genetically related rocks

of decreasing content of modal calcite ranging from calcite carbonatite (søvite), through pyroxene-carbonate rocks (kåsenite, *i.e.*, pyroxene søvite) and calcite-rich pyroxenites (hollaite, *i.e.*, calcite pyroxenite), to calcite-poor ijolitic rocks (calcite-bearing melteigite or ijolite). On one hand, it would obviously not be appropriate to refer to the calcite-poor pyroxenites as “carbonatite”, but their genetic relationship to calcite-rich rocks at this locality is inescapable. On the other hand, establishing an arbitrary modal limit for the carbonate fraction is also not really desirable. Unfortunately, there is no satisfactory solution to this dilemma without some form of petrographic “pigeon-holing”.

Silicate-bearing rocks, such as pyroxene calcite carbonatite, are commonly termed “silicocarbonatite” (Brøgger 1921, Pecora 1956), but this an unsatisfactory term, partly because it does not specify the silicate mineral present (pyroxenes, amphiboles, micas, olivine and humite-group minerals are common), but also because it tends to imply that these rocks might be genetically unrelated to the carbonatites that commonly accompany them in close proximity. Similar objections to the term have been made by Gittins *et al.* (2005). Allowing a lower limit of 30 vol.% carbonate rather than 50 vol.% enables these rocks to be adequately described by adding mineralogical prefixes to the carbonatite stem-name in ascending order of modal abundance, *e.g.*, phlogopite dolomite carbonatite or olivine phlogopite calcite carbonatite. For rocks with less than 30 vol.% carbonate, but that are part of the same suite, the terms would be calcite phlogopite or calcite pyroxenite. The further advantage of this approach is that it allows these rocks to be included in the carbonatite suite to which they belong.

There is an analogous situation in the carbonatites that contain widely varied amounts of phosphates and oxides. The 30 vol.% carbonate limit permits the recognition in the Jacupiranga complex of Brazil, of a carbonatite suite made up of perovskite calcite carbonatite, magnetite apatite dolomite carbonatite, and magnetite apatite olivine calcite carbonatite (phoscorite). With respect to phoscorite, at the type locality at Phalaborwa, South Africa (Ericksson 1989), there is a modal range from 100 vol.% magnetite through apatite calcite carbonatite to 100 vol.% olivine. Within a particular domain, individual rock-types could be called magnetite, carbonatite or olivine (dunite), but together they constitute a magnetite – apatite – olivine carbonatite. Such gross modal variations within relatively small areas are the rule rather than the exception in most plutonic carbonatite complexes. Thus, *a given carbonatite complex can be considered as a package of rocks that are modally diverse, but that have a common magmatic origin.* The term *carbonatite* can therefore be used as a name for a specific rock-type, but also as a group name for a suite of genetically related rock-types that include a wide range of carbonatites. For mapping

or descriptive purposes, individual domains within the “package” can be given specific modal names, provided the size of these domains is specified.

Recognition of many aspects of the taxonomy that have been discussed above is important both scientifically and economically, for failure to recognize a rock as belonging to a carbonatite suite might result in missed economic potential. From a scientific point of view, incorrect classification ultimately leads to incorrect petrogenetic hypotheses and even failure to describe rocks as carbonatite if the IUGS system is strictly followed. The latter point is important in an age where literature searching is based on the rapid scanning of databases that depend on keywords. Hence, it becomes disturbingly easy to miss important references if the IUGS system, or even careless classification, has been followed in describing rocks. Apart from purely scientific objections, use of a single term for “carbonatite” can result in misdirected efforts in exploration, in the search for rare metals and other mineral deposits, as each genetic type of “carbonatite” has particular geochemical characteristics. Thus, carbothermal fluids typically concentrate the rare-earth elements (REE), Sr, Ba, *etc.* and not Nb, whereas intrusive carbonatites associated with rocks of the melilitite clan are typically important sources of Nb, Ti and P, but not the REE. Note in particular that none of these carbonate-rich rocks are economic sources of Ta or Zr.

In the above discussion, I have so far only considered “carbonatite” *sensu lato*. Rocks described in the literature as “carbonatites” can be dominated by calcite, dolomite, siderite, Ba–Ca carbonates, Sr–Ba–REE carbonates, Na–Ca carbonates and rare-earth fluorocarbonates. The simple carbonates are in most occurrences considered to be primary magmatic minerals, whereas the complex carbonates could be of either primary magmatic or late-stage carbothermal in origin. This question is discussed further below.

PETROLOGICAL CLANS AND CARBONATITES

The relationships emphasized in the preceding section echo the concept of petrological clans, first enunciated by Daly (1914), which states that a particular magma-type can be produced repeatedly in space and time, and that a group of rocks with diverse mineralogy and texture can be formed by the differentiation of this parent magma. Detailed discussion of the application of this concept to alkaline rocks can be found in Mitchell (1995, 1996). Details of the geology of world-wide occurrences of carbonatite (*sensu lato*) can be found in Woolley (1987, 2001), Kogarko *et al.* (1995), Gittins (1966), and Heinrich (1966). Note that the geology of, and original references to, carbonatite and other alkaline complex localities mentioned there can be found in these compilations. In them, it is apparent that carbonatites (*sensu lato*) are associated with an extremely wide

variety of alkaline rocks, *e.g.*, with ijolite at the Fen Complex (Norway), uncompagrite at Iron Hill (USA), potassic syenite in the Bearpaw Mountains (USA), and peralkaline sodic agpaitic syenite at Khibina (Russia). Importantly, many occurrences of carbonatite have no association with undersaturated feldspathoidal alkaline rocks *e.g.*, Sarfartoq (Greenland), Bulhoek (South Africa), Newania (India), and not all alkaline rocks are associated with carbonatites, *e.g.*, nepheline syenite at Mt. Brome (Canada) or Kangerdlugssuaq (Greenland), lamproite (Leucite Hills), katungite (Toro–Ankole), and leucite (Vesuvius). The significance of these associations has been briefly discussed by Mitchell (2005) and Woolley (2003) with respect to Nb mineralization and genetic associations, respectively. Woolley (2003), using the IUGS criterion for the definition of carbonatite, suggested that six series (or magma types) of rocks can be distinguished as being associated with “carbonatites” (see below). However, Gittins & Harmer (2003) have disputed the significance of most associations of carbonatites and alkaline rocks in the same complex, suggesting that the association is spatial rather than genetic, and results from two separate magmas using the same conduit to reach the crust from their site of generation in the mantle.

In terms of the “petrological clan” concept and using a mineralogical-genetic approach to classification (Mitchell 1995, 1996), two broad groups of “carbonatite” (*sensu lato*) can be recognized. These are: (1) calcite or dolomite carbonatites (or both) that are primary and genetically related to nephelinite, melilitite, kimberlite and other mantle-derived magmas, and (2) carbothermal residua derived from a wide variety of magmas. Carbothermal is a term referring to low-temperature fluids derived from a fractionated magma dominated by CO₂ but also containing fluorine and H₂O in variable proportions. Addressing the question of whether or not carbothermal residua should be termed carbonatite at all is one of the major objectives of this work.

PRIMARY CARBONATITES

Nephelinite-clan carbonatites

This is perhaps the most common association and is represented by carbonatites associated with intrusive (melteigite – ijolite – urtite suite, nepheline syenite) and extrusive rocks (nephelinite, phonolite suite) of the nephelinite clan, *e.g.*, at Fen (Norway), Alnö (Sweden), Tororo and Napak (Uganda), and Shombole (Kenya). Notably absent are large volumes of melilitite-bearing rocks. If present, these are confined to *minor* early melilitite nephelinite lavas and tuffs, as found at Napak, Sadiman or Kerimasi, or they occur as spatially close, small-volume hypabyssal intrusions (Loolmurwak, Armykon Hill). Phlogopite, perovskite and olivine

pyroxenites and dunites are typically absent. Carbonatites range from calcite, through dolomite to siderite or ankerite-bearing types. Dolomite carbonatites can precede calcite types or *vice versa*, and calcite and dolomite can be present in the same rock. Late-stage events involving a carbothermal fluid can result in the formation of Sr-, Ba- and REE-bearing carbonates (Ngwenya 1994, Wall & Mariano 1996). Formation of these in some instances might involve interaction of pre-existing carbonatites with groundwater, *i.e.*, hematite – calcite – dolomite carbonatite (rødberg) and ankerite carbonatite at Fen (Andersen 1984, 1986), or they might represent residual fractions of the parental magmas, *e.g.*, at Kangankunde, Malawi (Wall & Mariano 1996).

This association was the first in which a direct relationship between nephelinitic volcanism and the plutonic carbonatite–ijolite series was recognized. At Napak, one of a series of Miocene volcanoes in eastern Uganda, nephelinitic agglomerates and tuffs form a dissected stratovolcano, the eroded core of which contains a plug of carbonatite surrounded by modally heterogeneous rocks of the melteigite – ijolite – urtite suite. These core rocks were considered by King (1949) and King *et al.* (1972) to occupy a magma chamber that was the original feeder vent to the volcanic edifice. Similar plutonic ijolitic central plugs, although without carbonatite, have been described from the nearby volcanoes of Moroto and Yelele (King *et al.* 1972). The carbonatite–nephelinite association is further strengthened by the occurrence of extrusive carbonatite as tuffs and lavas at the Shombole (Kenya) and Hanang–Balan-gida (Tanzania) nephelinitic volcanoes.

As a consequence of the above observations, many petrologists regard most plutonic nepheline syenite – ijolite – carbonatite complexes *e.g.*, Chisyana, Tororo (Uganda), Fen (Norway), Spitzkop (South Africa), Lackner Lake (Canada), to represent the roots of now eroded nephelinite–phonolite volcanoes.

Melilitite-clan carbonatites

This association includes carbonatites associated with plutonic rocks (melilitolites, *i.e.*, okaite, turjaite, afrikandite, uncompahgrite, perovskite pyroxenite, dunite, *etc.*) of the melilitite clan (Mitchell 1996), *e.g.*, Araxa, Catalão and Tapira (Brazil), Kovdor, Turja (Kola, Russia), Kugda, Romanikha (Maimecha–Kotui, Russia), Iron Hill (USA), Oka (Canada), Gardiner (Greenland). Also included in this group are “carbonatites” and ultramafic lamprophyres associated with the hypabyssal facies of the melilitite clan (Mitchell 1996), *i.e.*, alnöite, polzenite, *etc.*

Unlike the nephelinite clan, the melilitite clan has few carbonatites among its extrusive members. For example, none is known in the melilite-bearing volcanic centers of the Western Rift Valley of Africa, such as at Nyiragongo (Congo). In the Eastern Rift Valley, an asso-

ciation is observed principally at the dissected volcanic center of Kisingiri–Rangwa (Le Bas 1977). At Rangwa, melilitolites (uncompahgrite) occur as a significant component of the ijolite–carbonatite complex forming the roots of the Kisingiri volcano. However, melilitites appear to be absent from the associated volcanic rocks, and melilite-bearing rocks are represented by melilite nephelinites, which comprise less than ~5 vol.% of nephelinite volcanic suite (Le Bas 1977). Note that at Napak, a similar relationship is found, and minor amounts of melilitolite occur at the margin of the central ijolite complex, with small amounts of melilite nephelinite found at the base of the nephelinite volcanic series (King & Sutherland 1966). Melilitites occur as lavas and plugs in the Natron–Engaruka Depression, although these are not in direct association with the carbonatites occurring at Kerimasi and Oldoinyo Lengai (Dawson & Powell 1970). Apart from the above occurrences, it should be stressed that the overwhelming majority of extrusive rocks and intrusive complexes of the nephelinite clan, with or without carbonatite, do not contain melilitites or melilitolites. All of the melilite-bearing extrusive rocks in nephelinite-clan volcanoes appear to be early, volumetrically minor members of the volcanic suite and undoubtedly represent small-volume initial partial melts (see below).

It is primarily by their analogous geological structure to plutonic complexes of the nephelinite–carbonatite suite that melilitolite–carbonatite complexes are considered to occupy the root zones of former melilitite-clan volcanoes. The Osečná complex of northern Bohemia (Ulrych *et al.* 1988) is the only well-documented link between plutonic and subvolcanic (hypabyssal) members of the melilitite clan (Mitchell 1996).

The plutonic facies of the melilitite clan include a wide variety of ultramafic rocks (melilitolites) composed principally of calcite, melilite, olivine, phlogopite, clinopyroxene and perovskite. Nepheline-bearing rocks, if present, occur in only small volumes, and might be of rheomorphic origin. The paucity of nepheline syenite and ijolite–urtite is a feature that distinguishes melilitolite complexes from plutonic rocks of the nephelinite clan. Some phlogopite-rich rocks can be regarded as examples of the plutonic lamprophyric facies (Mitchell 1994a) of the melilitite clan. With decreasing grain-size, there is a complete gradation to hypabyssal rocks (micromelilitolites), and ultimately to alnöite and aillikite.

Three regions of the world are characterized by extensive development of melilitolite-bearing complexes: Kola–Kandalaksha (Russia), Maimecha–Kotui (Russia), and Minas Gerais (Brazil). Elsewhere, the complexes occur as isolated intrusions *e.g.*, Iron Hill, or at the margins of alkaline rock provinces dominated by nepheline syenite complexes *e.g.*, the Oka – Como – Ile Cadieux intrusions of the Monteregian Hills (Quebec). Unfortunately, many melilitolite complexes

are poorly exposed, especially those in Brazil, and have been insufficiently investigated. Melilitolite-bearing and associated complexes contain ultramafic rocks enriched in magnetite, perovskite, apatite and phlogopite, and are major economic sources of Ti, Nb, Fe, REE, mica and phosphate.

A wide variety of carbonatites occur typically as the latest-crystallizing members of melilitolite–pyroxenite complexes, and have been suggested to form by fractional crystallization [Iron Hill: Nash (1972), Turiy: Dunworth & Bell (2001)], liquid immiscibility [Gardiner: Nielson (1980)], and as carbothermal residua [Sebyav: Bulakh *et al.* (1998)].

Kimberlite-clan – calcite kimberlites

“Carbonatites”, in the IUGS modal sense, are associated with *bona fide* kimberlites, *e.g.*, the Benfontein (Dawson & Hawthorne 1973, Mitchell 1994b) and Wesselton Sills (Mitchell 1984), the Premier calcite “carbonatite” dikes (Mitchell 1986), and the Wemindji sill–dike complex (Mitchell & Letendre 2003). Other kimberlites, *e.g.*, Internationalya (Siberia), Jos and Nikos (Canada) contain primary carbonate, which occurs as microphenocrystic prismatic crystals. The latter can be concentrated by flow differentiation to produce rocks that might be termed “carbonatite” in a simple modal classification. These rocks represent late-stage differentiates of kimberlitic magma and are now termed “calcite kimberlites” in recognition of this genetic affinity (Mitchell 1986, 1995). They have NO genetic relationships to carbonatites associated with nephelinite- or melilitite-clan magmas, even though they can contain similar assemblages of carbonate minerals, and can in some instances, *e.g.*, Benfontein, differentiate to residua that contain Sr–REE carbonates (Mitchell 1994b).

The aillikite – carbonatite association

Aillikites consist of widely varying modal proportions of olivine, phlogopite and calcite or dolomite (or both). Other minerals present include characteristic magnesian spinel and perovskite, together with less common clinopyroxene and amphibole, although the latter silicates are more abundant in mela-aillikite (Tappe *et al.* 2004, Mitchell *et al.* 1999). Melilite, or the products of its subsolidus alteration, leucite, and nepheline, are absent. The best examples of the association are found in regions of Canada and Greenland adjacent to the Labrador Sea. Here occur several suites of Late Neoproterozoic dikes (Torngat, Abloviak, Sisimiut, Sarfartoq, Maniitsoq) that range in their mode from mela-aillikite through olivine phlogopite carbonatite to carbonatite (Tappe *et al.* 2004, Dignonnet *et al.* 2000, Mitchell *et al.* 1999). The Torngat–Abloviak swarm alone has intruded an area that is at least 1500 km². These regional dike-swarms are not associated

with mafic plutonic or feldspathoid-bearing alkaline complexes of any type.

The absence of feldspathoids serves to set these rocks apart from modally similar ultramafic lamprophyres associated with melilitolite (Kaiserstuhl, Osečná, Oka) or plutonic nephelinite complexes (Alnö). In these complexes, some rocks exhibit modal gradations from aillikite to others containing melilite and monticellite, *e.g.*, alnöite, polzenite, vescite, *etc.* The modal similarity of some of these rocks to the aillikite–carbonatite suite not associated with alkaline complexes is considered here to be an example of petrographic convergence, each suite being genetically distinct. The regional character and composition of these dike swarms are considered to indicate that they represent distinct primary mantle-derived carbonate-rich magmas (Mitchell *et al.* 1999, Dalton & Presnall 1998), whereas the ultramafic lamprophyres associated with alkaline complexes are undoubtedly local differentiates of melilititic or nephelinitic magmas.

The Sarfartoq complex (Greenland) and the Sisimiut–Maniitsoq dike swarms provide an illustrative example of the aillikite–carbonatite association. Here a large (15 km²) central complex (Sarfartoq) consisting of concentric silicate-bearing (phlogopite, amphibole) magnesiocarbonatite sheets with closely interleaved fenites is surrounded by an extensive (80 × 80 km) swarm of cone sheets and dikes of olivine phlogopite calcite- or dolomite carbonatite. The cone sheets within 25 km of the intrusion are centered on the carbonatite core, whereas the more distant dikes appear to be part of a regional swarm. The cone sheet and dike rocks have for many years been referred to as kimberlite (Larsen & Rex 1992), but have been shown by Mitchell *et al.* (1999) and Tappe *et al.* (2004) to be carbonate-rich ultramafic lamprophyres (*sensu lato*) and carbonatites. The complex and associated dikes are considered represent a primary partial-melting-related sequence (Mitchell *et al.* 1999, Dalton & Presnall 1998), with emplacement of olivine phlogopite carbonatite followed by the emplacement of the central dolomite carbonatite, perhaps as a derivative of those earlier-emplaced magmas.

Other Proterozoic carbonatite complexes, emplaced in Archean country-rock in northern Ontario, are characterized by the presence of pyroxene carbonatite or amphibole carbonatite (or both) and lack nepheline- or melilite-bearing pyroxenites, *e.g.*, Argor, Goldray, Cargill, Big Beaver House, Schryburt Lake. In terms of their mineralogy, they are similar to the aillikite–carbonatite suite and cannot be included with the plutonic complexes of the melilitite- or nephelinite-clan magmas. All of these complexes are exposed in deeply eroded terrains; they may well be the plutonic equivalent of the aillikite–carbonatite dikes of the Labrador Sea region.

*The peralkaline nephelinite –
natrocarbonatite association*

The natrocarbonatite lavas erupted at Oldoinyo Lengai, Tanzania (Dawson 1962) are unique. The lavas consist of phenocrysts of gregoryite $[(\text{Na,K,Ca})_2\text{CO}_3]$ and nyerereite $[\text{Na}_2\text{Ca}(\text{CO}_3)_2]$ set in a matrix composed predominantly of a gregoryite-like carbonate, Ba-rich nyerereite, sodian sylvite, and fluorite. The groundmass also contains significant amounts of a Ba–Ca–Sr–Na-rich carbonate, which Mitchell (1997) regarded as a quenched carbonate solid-solution; it has the potential to break down to olekminskite, alstonite and other Sr–Na–Ba–Ca carbonates, as observed in the subsolidus evolution of carbonates at the Little Murun complex (Konyev *et al.* 1991, Vorobyev & Piskunova 1987). Calcite and dolomite are notably *absent* in unaltered rocks.

Oldoinyo Lengai is a stratovolcano composed principally of peralkaline nephelinite tuffs and lavas, together with highly peralkaline phonolite. Xenoliths derived from the infrastructure of the volcano consist of jacupirangite, ijolite, melteigite and nepheline syenite, including peralkaline varieties (Dawson & Hill 1998). The natrocarbonatite lavas are the youngest manifestation of igneous activity (Guest 1956, Dawson 1962), and have been considered to be derived by either liquid immiscibility (Kjarsgaard 1998, Kjarsgaard & Hamilton 1989) or fractional crystallization from a peralkaline nephelinite parent (Pettibon *et al.* 1998). However, other genetic possibilities can be suggested. Thus, the lavas might represent the products of fractional crystallization of a phonolitic magma (this work), be vapor-phase condensates (Nielson & Veksler 2002) or even formed by the interaction between silicate magma and evaporitic lacustrine sediments (Milton 1968, Lentz 1999a).

Le Bas (1981) has suggested that natrocarbonatites are parental to calcite carbonatites. However, regardless of the actual genetic process, it is now certain that natrocarbonatite is a residual magma and *cannot* be parental to calcite carbonatites as found in carbonatite–ijolite complexes of the nephelinite clan (Twyman & Gittins 1987, Mitchell 1997, Mitchell & Belton 2004). Because of their close association with sodic peralkaline volcanic rocks, the Oldoinyo Lengai natrocarbonatites are considered here as part of a peralkaline nephelinite association, but are recognized as a distinct variety of carbonatite because of their unique mineralogy.

Since the recognition of the Oldoinyo Lengai lavas, there have been claims that extrusive natrocarbonatites are widespread. Pyroclastic rocks occurring at Homa Mountain, Laetolil, Kerimasi, Tinderet, and Kaluwe have been interpreted to represent altered natrocarbonatite (Hay 1983, Hay & O'Neil 1983, Deans & Roberts 1984, Clarke & Roberts 1986, Turner *et al.* 1988). In these rocks, no relics of the alkali–calcium carbonates allegedly present originally have been preserved, and

the evidence that lath-shaped pseudomorphs of calcite represent former nyerereite is at best circumstantial. Many of the pseudomorphs might actually represent altered melilite or even primary calcite. Replacement of nyerereite should also result in a 50% volume decrease and disruption of the original texture; such changes have not been documented (Gittins & Jago 1991, Barker 1996). Even if the pseudomorphs do represent a former nyerereite-like mineral, it does not follow *a priori* that the original rock was Oldoinyo-Lengai-type natrocarbonatite. It is possible that a nyerereite-like sodic–calcic carbonate has formed merely as a subliquidus phase during the crystallization of these magmas. The absence of fluorite, secondary chlorides, and pseudomorphs after gregoryite is strong evidence against the natrocarbonatite-replacement hypothesis.

In summary, the natrocarbonatites of Oldoinyo Lengai appear to be unique. Whether similar material has been erupted in the past and at other volcanic centers is unknown, as their small volume and extremely rapid decomposition do not favor preservation and recognition, even in the older volcanic rocks at the type locality.

Carbonatites without associated silicate rocks

Woolley (2003) has noted that there are at least 68 intrusive and 10 extrusive carbonatite localities with no associated intrusive or extrusive silicate rocks, excluding associated fenites. The simplest explanation for the absence of silicate rocks is non-exposure of the deeper parts of a parental plutonic complex. This situation could be represented by the Kangankunde complex (Malawi), which Garson (1965) considered as a preserved superstructure similar to that postulated to be originally present above the more deeply eroded Chilwa and Tundulu complexes.

An alternative explanation for other occurrences, *e.g.*, Bulhoek (South Africa), Laiwu–Zibo (China; Ying *et al.* 2004), is that there really are no associated silicate rocks belonging to the ijolite – nepheline syenite or melilitolite suites. In some instances, emplacement of carbonatites is followed by that of silicate rocks, *e.g.*, Dorowa, Shawa, Buhera (Zambia), and Spitzkop (South Africa). Gittins & Harmer (2003) claimed that in these cases, there is no genetic relationship, as the two magmas merely utilize the same conduits. If this hypothesis is correct, the carbonatites in these examples might also represent primary partial melts.

Ultimately, each occurrence of carbonatite not obviously associated with silicate rocks must be assessed on its own merits, but it is evident that there are some “carbonatites”, commonly dolomitic, which are emplaced without the ijolite – nepheline syenite or melilitolite suite, and thus must represent distinct magmas. Regardless, these undoubtedly are not “pure” carbonate melts and must contain some silica and alkalis, as suggested by Twyman & Gittins (1987).

CARBOTHERMAL RESIDUA AND PSEUDOCARBONATITES

Potassic-suite "carbonatites"

"Carbonatites" are associated with potassic plutonic rocks at Mountain Pass (California), Rocky Boy (Montana), Little Murun (Yakutia), Dunkeldysky (Tajikistan) and Loch Borralan (Scotland). The bastnäsite-rich carbonatite occurrence at Mountain Pass is well known as a major economic source of rare-earth elements (Olsen *et al.* 1954, Mariano 1989, Haxel 2005). The characteristics of most of these occurrences are their association with diverse saturated to undersaturated potassic syenitic rocks, high abundances of REE-bearing carbonates and barite, and the total absence of members of the ijolite and melilitolite suites. Feldspathoids, where present, are typically leucite, pseudoleucite, and kalsilite. Rocks composed of orthoclase and diverse carbonates are common in this association.

At Mountain Pass (Olsen *et al.* 1954, Haxel 2005) potassic syenite and shonkinite are associated with carbonatite dikes and veins consisting principally of: calcite, barite and bastnäsite, siderite, barite, bastnäsite and quartz, ankerite and barite, ankerite and fluorite, and quartz. The largest of these veins, known as the Sulphide Queen orebody, consists of three distinct units: ferruginous dolomite carbonatite; bastnäsite barite calcite carbonatite, and barite quartz bastnäsite (up to 60 vol.%) carbonatite. The dolomitic carbonatite contains abundant pyrite, magnetite, monazite, bastnäsite and parisite. Notably absent are Nb-, Ti- and Zr-bearing oxide minerals. These carbonatites are commonly considered to be magmatic (Mariano 1989, Jones & Wyllie 1983), but an origin as carbothermal residua derived by the fractional crystallization of a potassic magma is equally appropriate. However, relationships to the potassic syenites are ambiguous, as the carbonatite appears to be 25 Ma younger than the main shonkinite (DeWitt *et al.* 1987). Mineralogically similar barite- and REE-rich "carbonatites" associated with nordmarkite and granite have been described from the Maoniuping deposit (Sichuan, China) by Wang *et al.* (2001).

At the Rocky Boy (Montana) occurrence (Pecora 1962), a stockwork of veins of carbonatite and silicate-bearing carbonatite occur within a "sericitized" potassic syenite. The veins consist of orthoclase, biotite, apatite, carbonates and sulfide in widely varying modal proportions. Reguir & Mitchell (2000) have reported as present in these veins: Sr-rich calcite with exsolved strontianite, siderite, ankerite, dolomite, synchysite-(Ce), bastnäsite, ancylite-(Ce), ancylite-(Nd), calcioancylite-(Ce), burbankite, carbocernaite, and huanghoite. These "carbonatites", which are similar in paragenesis and association to the potassic syenite at Mountain Pass, are interpreted to be fractional crystallization residua

derived from the potassic syenitic parental magma (Reguir & Mitchell 2000).

Sr-Ba-rich calcite carbonatites occurring at the Dunkeldysky complex (Tajikistan) are associated with potassic plutonic rocks that include pseudoleucite syenite, borolanite, and fergusonite. The carbonatites contain: strontian calcite, strontianite, celestine, barite, fluorite, and diverse REE fluorocarbonates (Faiziev *et al.* 1998).

The "carbonatites" of the Little Murun complex (Borisov 1985, Konyev *et al.* 1996, Reguir 2001) are associated with a wide variety of strongly undersaturated potassic rocks including: kalsilite- and pseudoleucite syenite, shonkinite, clinopyroxenite, nepheline syenite, phonolite and trachyte. The carbonatite bodies occur at the southern contact of the complex with the Precambrian basement rocks, in an aureole of aegirine-microcline fenites formed at the expense of Archean granite gneiss and Upper Proterozoic quartzite and sandstone. The carbonatites are closely associated with the charoitite complex and form veins and lenses up to 0.5 m in thickness.

Varieties of "carbonatite" present include calcite carbonatite, Ba-Sr-Ca carbonatite or benstonite carbonatite, and phlogopite calcite carbonatite. Also present are aegirine carbonatite, titanite carbonatite, and quartz calcite carbonatite (torgolite). The "carbonatites" exhibit an extremely wide range in their modal composition and are unique in that they can contain charoite, tinaksite, fedorite, and other exotic silicate minerals. Carbonates in the Ba-Sr-Ca suite are characterized by extremely complex subsolidus intergrowths of Sr-rich calcite, barytocalcite, strontianite, witherite, ancylite, burbankite, olekminskite and kukharenkoite (Vorobyev & Piskunova 1987, Konyev *et al.* 1996, Reguir 2001). The "carbonatite" suite at Little Murun is unique and is currently regarded as the product of late-stage carbo(hydro)thermal fluids that have reacted with metasomatized magmatic and country rocks (Orlova 1988).

Unlike the "carbonatites" described above, the apatite phlogopite calcite carbonatites associated with the potassic Loch Borralan intrusion are not enriched in REE-rich minerals, although they do contain fluorite and sulfides (Young *et al.* 1994). However, the exposure is very limited, and undoubtedly only a small portion of this carbonatite suite has as yet been investigated.

From the above, it is clear that "carbonatites" associated with potassic plutonic rocks are very different in their paragenesis and mineralogy to most of the primary carbonatites associated with the nephelinite and melilitite clans, and it is evident that these magma types play no role in their genesis. Parental magmas to the plutonic potassic complexes are not well characterized, but appear to have been derived from ancient metasomatized lithospheric mantle (Mitchell 1995, 1996).

Sodic-suite "carbonatites"

Carbonatites, or carbo(hydro)thermal residual fluids, are associated with sodic peralkaline syenite at the Khibina complex, Russia (Minakov *et al.* 1981, Zaitsev *et al.* 1998). Although there are only small amounts of these rocks relative to other silicate rocks present, they demonstrate that sodic peralkaline magmas can differentiate to residual fluids that crystallize REE-, Na- and Ca-carbonates. At Khibina, an early series of calcite carbonatite dikes is followed by veins of younger calcite carbonatite, calcite – manganian ankerite and ferroan rhodochrosite – manganian siderite carbonatite, and, ultimately, by carbonate–zeolite rocks. The oldest series of calcite carbonatites do not contain REE minerals, whereas the younger calcite carbonatites, Mn–Fe-rich carbonatites and calcite–zeolite rocks contain up to 17 distinct REE, Sr and Ba carbonates and sulfates, including: burbankite, carbocernaite, ancylite-(Ce), synchysite-(Ce), and kukharenkoite-(Ce). Zaitsev *et al.* (1998) documented complex metasomatic replacements of primary Sr–REE–Ca–Na-rich carbonates (burbankite, carbocernaite) by later Ca–REE fluorocarbonates (bastnäsite, *etc.*) and Ba–REE carbonates (cordylite group). The Khibina carbonatites are interpreted by Zaitsev *et al.* (1998) to be polygenetic, with the earliest calcite carbonatites considered to be as magmatic, the later REE–Sr–Na–Ba carbonatites to be derived from carbohydrothermal fluids, and the youngest calcite–zeolite rocks, to be hydrothermal residua.

The Khibina "carbonatite" appears to be unique, especially with respect to the contemporaneous enrichment of Na, REE and Mn. Other REE-rich carbonatites reputed to be associated with sodic peralkaline nepheline syenite include the Niznesayanskii and Verkhnesayaniskii complexes of East Sayan, Russia (Kogarko *et al.* 1995).

REE–F-rich carbonate rocks not related to undersaturated alkaline rocks

This diverse group of REE–F-rich carbonate-rich rocks are not obviously related to any undersaturated alkaline rocks and includes: the Ravelli–Lemhi area (Idaho–Montana), Goldie (Colorado), Rock Canyon Creek (British Columbia), and Bayan Obo (China). Heinrich (1966) has been the principal advocate regarding the western USA occurrences as "carbonatite". Geological and geochemical aspects of many of these occurrences have been described by Samson & Wood (2005), who noted that diverse genetic models have been proposed for many of these deposits, *e.g.*, Bayan Obo, some of which do not invoke any relationship to alkaline magmatism or carbonatite.

This diverse group of rocks have no common characteristics apart from a significant enrichment in Th, REE and F, which is reflected by the presence of REE fluorocarbonates, fluorite and other fluoride minerals.

In the Ravelli–Lemhi area (Idaho–Montana), veins and dikes of calcite and dolomite with barite and fluorite occur in metasedimentary rock of the Proterozoic Belt Supergroup. The mode and mineralogy of the veins vary widely, and silicates, including quartz, are common constituents. In this area, three types of vein have been recognized: fine-grained foliated dolomite, coarse-grained dolomite with Sr–Ba-rich calcite, and coarse-grained Sr-poor calcite. The main REE minerals are monazite, ancylite, bastnäsite and allanite. Niobium minerals present include niobian rutile (dominant), fersmite and columbite. At these occurrences, there appears to be a gradation from rocks that appear to be unaltered bands of phosphatic dolomite marble to cross-cutting veins. Many of the veins have fenitized margins, and Heinrich (1966) noted that even those bodies that most closely resemble marble in texture and mineralogy contain monazite and carbonates with abnormal Sr contents. Both Heinrich (1966) and Woolley (1987) regarded the veins as intrusive "carbonatites". Similar rocks are found at the nearby Snowbird deposit (Montana). Here a lens-shaped vein-enlargement body is lined with large (7 m) quartz crystals. The interior is filled by coarse-grained calcite, dolomite, fluorite, parisite, pyrite and gersdorffite. The occurrence has been termed a carbonatite pegmatite (Clabaugh & Sewell 1964).

The Rock Canyon Creek fluorite–REE deposit (British Columbia) is hosted by a sequence of Ordovician–Devonian limestones and dolomites. Mineralization is disseminated throughout altered carbonate rocks and consists of fine-grained disseminated vein and breccia-matrix REE-bearing fluorite with lesser barite, synchysite, parisite, pyrite, quartz, rare pyrochlore, rare complex fluorides and niobian rutile. The host carbonates have been altered to ferroan dolomite. Pell (1994) regarded the occurrence as *bona fide* carbonatite and noted that veins of similar mineralogy occur adjacent to the Aley carbonatite complex.

Bayan Obo is a giant polymetallic Cu–REE–Fe ore deposit which Samson & Wood (2005) classified as an Olympic-Dam-type deposit. At Bayan Obo, the orebodies are apparently stratabound and consist of discontinuous mineralized lenses in dolomite. The ores vary greatly in texture and mineralogy, and can be banded, disseminated or massive. Several paragenetic stages have been recognized by Smith *et al.* (2000), which in sequence are: (1) disseminated monazite, magnetite, iron carbonate and bastnäsite, (2) monazite, bastnäsite, apatite, magnetite – hematite – aegirine, fluorite, (3) aegirine – bastnäsite – apatite – calcite – barite – fluorite, (4) fluorite – bastnäsite – parisite, (5) barite – parisite – huanghoite and other Ba–REE fluorocarbonates. Niobium mineralization is also present as niobian rutile, columbite, aeschynite and pyrochlore. Models for the origin of the deposit range from sedimentary authigenic to primary igneous carbonatite mineralization. In support of the latter, Le

Bas *et al.* (1992) and Yang *et al.* (2003) have reported “carbonatite” dikes in the region.

Among petrologists interested in carbonatites, these occurrences have attained an importance that is out of all proportion to their abundance, and their similarities to other REE–fluorite deposits that are either not connected with undersaturated alkaline rocks or are demonstrably associated with granitic complexes is typically ignored. The mineralogy of many of these deposits bears a remarkable resemblance to that of late-stage quartz – calcite – fluorite pegmatites and veins associated with peralkaline A-type granitic magmas, *e.g.*, Coldwell complex (Ontario), Thor Lake (NWT), South Platte Gallinas Mountains (New Mexico). For example, pegmatites associated with ferroaugite syenite of center 1 of the Coldwell complex contain quartz, calcite, fluorite, synchysite, bastnäsite, niobian rutile and fluorite.

An enrichment in REE, Sr and Nb is commonly invoked as evidence indicating a carbonatite affinity; however, similar elevated levels of these elements and REE distribution patterns must be expected for any rock enriched in light REE fluorocarbonates, regardless of origin. Indeed, Tuttle & Gittins (1966) have previously noted that rare-earth mineralization associated with granitic and syenite pegmatites is common in highly metamorphosed terranes and that under extreme conditions of metamorphism, carbonate rocks containing rare-earth minerals could be generated which would bear no genetic relationship to carbonatite–alkaline complexes (see also below). Gittins (1978) has also noted that carbonate veins enriched in Sr and REE should not automatically be considered as carbonatites.

In summary, it appears that designation of the REE-rich carbonate rocks described above as “carbonatite” stems essentially from the assumption that as some carbonatite-forming magmas differentiate to late-stage REE fluorocarbonate-rich residua *e.g.*, Fen (Norway), Tundulu (Malawi), St-Honoré (Quebec), then similar REE fluorocarbonate mineralization in other environments must also be carbonatite-derived. Unfortunately, this syllogism is not typically supported by any geological evidence, and designation of these rocks as “carbonatite” leads to inappropriate genetic speculation. Hence in this work, all of the REE–F-rich carbonate rocks described above are termed carbothermal residua.

Anatectic “pseudocarbonatites”

The Haliburton–Bancroft area of Ontario, southwestern Quebec and parts of northern New York State are characterized by the presence of actinide-rich phlogopite – apatite – fluorite – calcite ore deposits. These occur as conformable tabular bodies and discordant veins within metamorphic rocks of the Grenville Province. The deposits are enriched in elements that are not found in Grenville marble, and their parental fluids appear to have interacted with the marbles to

form calc-silicate skarns. Heinrich (1966) and Shaw *et al.* (1963a, b) have noted that the rocks are generally coarse-grained to pegmatitic, abundant where marbles occur, and most common in rocks of the highest metamorphic grade. Their mineralogical character is diverse and highly variable over short distances. Petrographic gradations occur between these rocks and amphibolite, marble, gneiss and granite. The principal minerals are pink calcite, diopside, amphibole, phlogopite, fluorite, quartz, apatite, and allanite, with lesser zircon, uraninite, and betafite. Notably absent are REE–Sr fluorocarbonates and Ba–Ca–Mg carbonates.

The rocks have been designated as carbonatites by Heinrich (1966) and Lentz (1998, 1999b), regarded as skarns by Shaw *et al.* (1963a, b), and not even included in the carbonatite pantheon by Gittins (1966) and Woolley (1987). It is important to note that nepheline syenite gneisses and pegmatites of uncertain origin, as well as nepheline syenites with an indisputably igneous texture, also occur in this part of the Grenville Province. The region has abundant metasedimentary marble and figured prominently in Day’s and Shand’s limestone syntexis hypothesis for the genesis of nepheline-bearing rocks. In a recent variant of this process, Lentz (1998, 1999b) has proposed that the “carbonatites” result from intrusion-related pneumatolytic melting of limestone. In this scenario, intrusion of H₂O-rich alkaline magmas generates skarns and causes melting of limestone under H₂O-saturated conditions rather than simple decarbonation. Carbonatites are produced by volatile fluxing and emplaced in the upper crust distally to the magma causing the melting. Thus, a direct spatial association between the alkaline rocks and the carbonatites cannot be observed. The hypothesis is supported by experimental data on the melting relationships of calcite in the presence of H₂O, and the contemporaneous association with high-grade metamorphic rocks is inescapable. However, melting during anatexis without connection to alkaline magmas is just as feasible, with the H₂O and other volatiles, especially fluorine, required for pneumatolysis derived by the metamorphic dehydration of diverse hydrous silicates or from the granite and syenite pegmatites, which also formed the U-bearing pegmatites. Discussion of the details of the genesis of these rocks is beyond the scope of this work.

The mineralogy and geological association of these calcite-rich rocks is so different to that of carbonatites of indisputable igneous origin that they should not even be considered as *bona fide* carbonatites. The rocks are better termed mobilized calc-silicate skarns or anatectic calc-silicate veins. They are clearly anatectic melts of crustal rocks and are totally unrelated to mantle-derived alkaline magmas.

Summary

On the basis of the above, there seem to be four varieties of carbonatite that could be considered as

primary magmatic “carbonatite” *sensu stricto*, namely the carbonatites of the nephelinite and melilitite clans, those of the aillikite–carbonatite association, and natrocarbonatite. As discussed below, even these carbonatites might have a multiplicity of origins. Calcite kimberlites could conceivably be considered as *bona fide* carbonatites given that they are ultimately formed from mantle-derived magmas. However, Mitchell (1979, 1986, 1996) has argued strongly against this terminology, as it leads to unwarranted genetic speculation as to relationships between kimberlite and other unrelated magma-types.

The “carbonatites” associated with potassic and sodic plutonic rocks, and occurrences of the Ravelli–Lemhi type that are rich in minerals of barium, strontium and rare-earth elements appear in all instances to represent low-temperature and low-pressure carbothermal, fractional-crystallization-related derivatives of diverse, and commonly unidentified, parental magmas. Similar rocks are also produced by the differentiation of “carbonatite-forming melts” associated with nephelinite- and melilitite-clan magmas. As a consequence of the thermodynamic stability-fields of Sr, Ba and REE carbonates, barite and fluorite, it is not surprising that there should be mineralogical convergence between rocks derived by extensive fractional crystallization of magmas of diverse genetic lineage at low temperature and pressure. Thus, carbothermal residua associated with Kangankunde and Tundulu carbonatites are not genetically equivalent to similar rocks found at Mountain Pass, as they have different antecedents. Hence, all members of this group are best termed “carbothermal residua” and not “carbonatites”, in order to emphasize these genetic differences. This approach leads to a more “long-winded” descriptive terminology, but at least it has the merit of being genetically exact, *e.g.*, calcite carbothermal veins derived from peralkaline sodic agpaite syenite (Khibina), bastnäsite–barite carbothermal veins derived from a nephelinite-clan magma (Tundulu), quartz–parisite carbothermal veins derived from an unknown magma (Snowbird). Thus, it is a contention of this work that a genetic distinction should be made between carbothermal deposits of diverse genesis. Note that if usage of the term “carbonatite” is restricted to carbonate-bearing (*variety unspecified*) rocks that are proven to have formed *only* from the low-temperature residua of nephelinite- and melilitite-clan magmas, then these derivatives might still be termed “carbonatite”, *e.g.*, bastnäsite–barite carbonatite. Although such a designation recognizes the continuum between primary magmatic carbonate and carbohydrothermal residua, its use can still lead to inappropriate petrogenetic comparisons with genetically different but modally similar carbothermal vein deposits.

The “pseudo-carbonatites” formed by anatectic melting of crustal rocks are considered *not* to be “carbonatites” either *sensu lato* or *sensu stricto*, and their designation as such engenders only misleading

speculation as to their origins and relationships to *bona fide* carbonatite. They should be referred to as anatectic skarn and vein deposits.

Although there is no claim that the above classification is definitive, as it is a work in progress, it is clear that the use of a single term for carbonate-rich rocks of diverse genesis is undesirable.

PETROGENESIS: HENS OR EGGS? NEITHER OR BOTH?

Heinrich (1966) summarized the various petrogenetic schemes advocated to explain the relationships between carbonatites and associated silicate rocks in terms of the familiar “hens-versus-eggs” conundrum. These hypotheses suggest that either the initial magma is silicate-rich (hens) and carbonatites are differentiates (eggs), or the initial magma is carbonatite (hens) and reacts with country rocks to produce silicate rocks (eggs). In the latter case, the ijolite series are regarded as rheomorphic ultrafenites. Surprisingly, some 40 years later, petrogenetic schemes for carbonatite origins have not substantially changed (see Gittins & Harmer 2003). Currently, as a result of experimental work on synthetic carbonatites and carbonated upper mantle, coupled with isotopic studies, there is a better understanding of the potential sources and depths of origins of diverse magma-types, but the fundamental dichotomy regarding the status of carbonatite remains. The introduction of liquid immiscibility as a genetic process does not change these general concepts, as it is really only a modification of the silicate parent hypothesis. One hindrance to all genetic studies is a limited understanding of the character of any of the carbonatite-forming magmas.

In the plutonic environment, most carbonatites definitely do not represent quenched liquids; rather, they are mobilized crystal cumulates formed from one or several batches of a particular magma-type. In the extrusive environment, the only lavas known are the unique natrocarbonatites of Oldoinyo Lengai, and one very small flow of calcite carbonatite from the Kalyango crater, Fort Portal, Uganda (Barker & Nixon 1989). Unfortunately, the latter is, for the most part, altered and contains crustal xenoliths, and is not associated with any silicate lavas. Most extrusive calcite carbonatites (Fort Portal, Kaiserstuhl in Germany) are pyroclastic rocks (Barker 1989, Keller 1989) consisting of lapilli whose petrographic character is influenced by crystal sorting prior to, and during, eruption. There are no indisputable examples of rapidly quenched calcite or dolomite carbonatitic liquids that might represent either primary or derivative magmas. Hence, petrologists have no real idea of the mineralogical and compositional character of any carbonatite-forming magma except that it must contain some Si, Mg, Fe, P and alkalis (Twyman & Gittins 1987) to account for the common presence of mafic silicates, oxides, and phosphates. Consequently, current petrogenetic schemes are rife

with unconstrained speculation, as the composition of the magmas discussed must necessarily be inferred. Thus, the question remains, are carbonatites always residua formed by the fractional crystallization of less-evolved carbonated silicate magmas or *vice versa*, and what other possibilities exist?

Figures 1–3 suggest some petrogenetic schemes for the generation of diverse carbonatitic magmas and accompanying silicate rocks of the nephelinite clan. Similar schemes can be devised for carbonatites associated with the melilitite clan. Magma composition is considered in terms of SiO_2 and CaO , as the content of these elements reflects whether or not the liquids are essentially carbonatite, silicate carbonatite or silicate. Three scenarios for the production of carbonatite melts are considered: fractional melting, batch melting, and liquid immiscibility together with fractional crystallization. Which path is taken will undoubtedly have a profound influence on the composition and mineralogy of the carbonatites produced.

In the following, it is assumed, for the purposes of discussion only, that all carbonatites and associated silicate magmas are produced by partial melting of carbonated lherzolite source-rocks located within the asthenospheric upper mantle (Wyllie & Huang 1976, Brey 1978, Canil & Scarfe 1990, Dalton & Presnall 1998), with the formation of different primary magmas depending upon the extent of partial melting and the depth at which it occurs (see below). According to isotopic studies (Bell & Tilton 2001), there are undoubtedly lithospheric contributions to these magmas. To illustrate the general concepts of carbonatite formation,

the initial discussion is restricted to magmas of the nephelinite clan.

Fractional and batch melting with fractional crystallization

If carbonatites and associated silicate rocks represent a primary partial-melting sequence (with or without reaction with mantle material), the activity of silica in the melt can be expected to increase as partial melting progresses and the initial carbonate-rich melt becomes enriched in silica (Fig. 1). Hence, one can envisage that during fractional melting of a carbonated mantle-source, with increasing degree of melting, there could be extraction of a carbonatite liquid followed by a separate silicate liquid such as a nephelinitic magma. Importantly, the latter, if also containing carbonate, could in turn differentiate into a residual carbonatite and ultimately carbothermal residua. Thus, two varieties of carbonatite, one a primary melt (*carbonatite-1*) and the other a residual melt (*carbonatite-2*) can be formed. Thus, intrusion of carbonatite can be followed by nephelinite-clan rocks with subsequent emplacement of late-stage REE-enriched carbonatite or carbothermal rocks (or both), as seen at Chilwa, Tundulu and Kangankunde (Garson 1965).

A second possibility might involve termination of partial melting at an early stage. In this case, carbonatites (*carbonatite-1*) and silicate carbonatites (*silicate carbonatite-1*) might be emplaced with no associated nephelinite-clan rocks. Note that silicate carbonatite-1 could also differentiate to REE–calcite-rich residua.

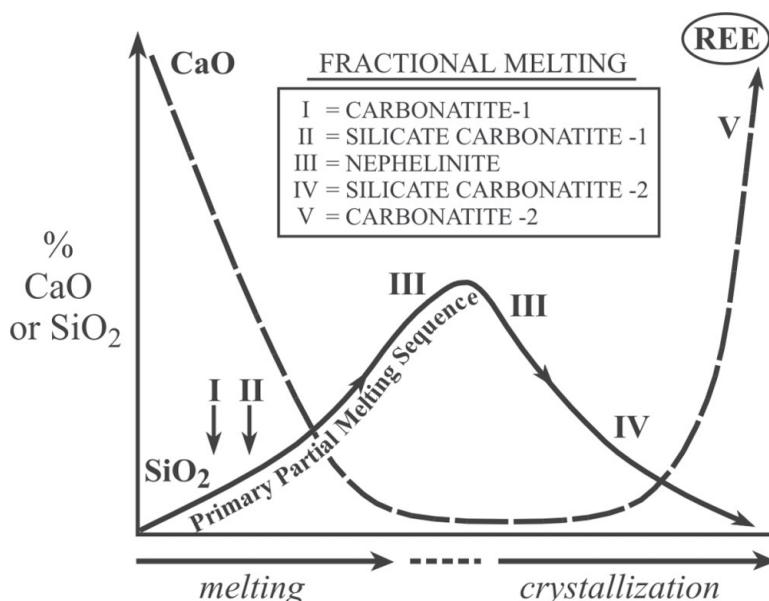


FIG. 1. Fractional melting and crystallization model for the genesis of carbonatites.

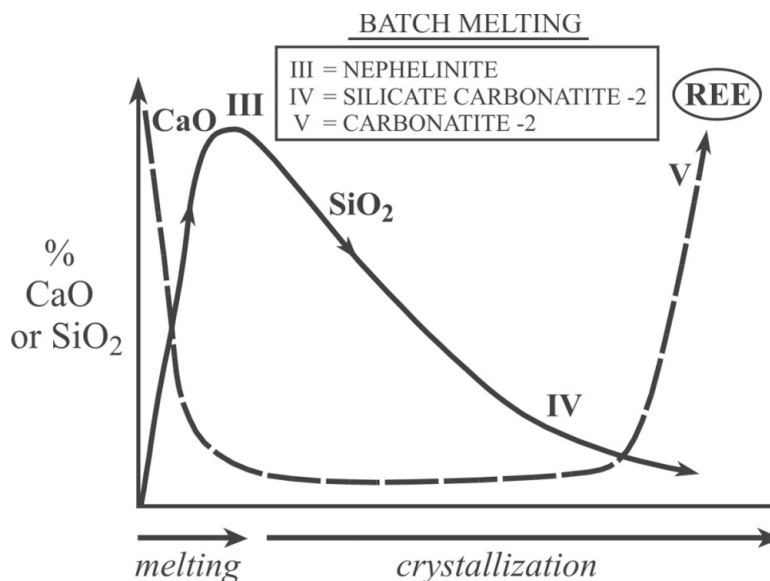
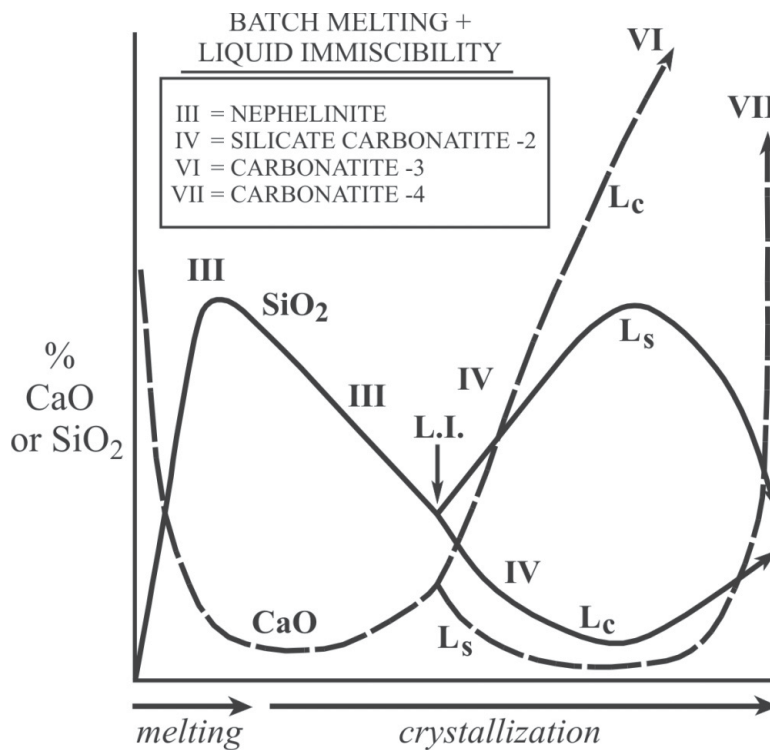


FIG. 2. Batch melting and fractional crystallization model for the genesis of carbonatites.

FIG. 3. Batch melting, liquid immiscibility (L.I.) and fractional crystallization models for the genesis of carbonatites. L_s represents the silicate-rich liquid, and L_c , the carbonate-rich liquid.

Such a scenario might explain the Sisimiut–Sarfartoq–Maniitsoq regional dike swarm of olivine phlogopite dolomite carbonatite and associated differentiated carbonatite at Sarfartoq.

A further possibility is that if all carbonate is extracted from the source during an initial period of melting, then later batches of magma extracted from the same source might contain insufficient carbonate to generate carbonatites as differentiates. In this case, nephelinite volcanism could occur with or without contemporaneous precursor carbonatite, but without any associated late-stage carbonatites.

In batch melting, which is rapid and more extensive, any initial carbonate-rich melts are directly incorporated into the silicate melt (Fig. 2). In this case, carbonatites (*carbonatite-2*) could only be formed by differentiation of this magma, and carbonatites would be emplaced subsequent to nephelinite-clan rocks, *e.g.*, in the Fen complex.

Batch melting, liquid immiscibility and fractional crystallization

A further complicating factor in carbonatite genesis is that there is the possibility of a carbonated silicate magma undergoing liquid immiscibility (Lee & Wyllie 1998, Kjarsgaard & Hamilton 1989). Carbonatitic magmas might represent one of the immiscible fractions that will coexist with a silicate fraction which, by definition, also must be “carbonated”. The latter also would have the potential to differentiate by fractional crystallization to a carbonate-rich melt (Fig. 3). In this case, two further distinct varieties of “carbonatite” (*carbonatite-3* and 4) could be formed from the same parental magma. These would differ in their composition and mineralogy from each other (and also from *carbonatites-1* and 2), because of initial compositional differences imposed by differing partition-coefficients for many elements between the silicate and carbonate fractions. Using synthetic carbonated nephelinite and melilitite, Brooker (1998) has demonstrated that immiscibility can take place in the upper parts of the lithospheric mantle or the crust, as the size of the two-liquid field is strongly dependent upon changes in CO₂ pressure.

The above points of discussion suggest that at least *four* distinct varieties of carbonatite could be produced from magmas of the nephelinite clan. If a similar number can be formed from melilitite-clan magmas then, in genetic terms, at least *eight* types of carbonatite are possible! This can be further increased to *ten*, if calcite kimberlites are regarded as “carbonatites” (*sensu lato*) and natrocarbonatites are included in the group. Each of these carbonatites should possess distinct mineralogical, compositional and isotopic characteristics, and in principle, it should be possible to distinguish among the various types. Unfortunately, discussion of such characterization is well beyond the scope of this work.

Relationships among carbonatites derived from different petrological clans

Although discussion of the genesis of the kimberlite, melilitite and nephelinite clans is also beyond the scope of this work, these magmas are commonly considered to be derived from different depths in the asthenospheric upper mantle with or without lithospheric contributions to their composition (Canil & Scarfe 1990, Brey 1978, Mitchell 1995, 1996). On the basis of published experimental work on the partial melting of potential carbonated sources in the mantle, it is unlikely that there is a melting continuum to produce these different magmas at a single depth, although it is possible that an uprising “diapir” of mantle-material might melt over a wide range in temperature and pressure, and generate melts of diverse composition. Currently, kimberlites are considered to form at depths greater than 150 km (pressures > 5 GPa), melilitites at 120–80 km (4 – 2.7 GPa) and nephelinites at shallower depths (<80 km; < 2.7 GPa). Thus, carbonatites that originate as primary partial melts could be derived over a wide range of temperatures and pressures. Those that result from crystal–liquid differentiation or liquid immiscibility (or both) are probably formed at relatively low pressures (< 1 GPa), with some examples undoubtedly forming under near-surface conditions (<0.1 GPa). Some possible spatial relationships with respect to the upper mantle sources of these magmas are illustrated in Figure 4. Note that similar conclusions with respect to the relative depth of origin of carbonatites associated with kimberlitic, melilititic and nephelinitic magmas have been reached by Woolley (2003, Fig. 1), although he also includes phonolites, trachytes and alkali basalts as other silicate rocks with which carbonatites are associated. Although melilitite- and nephelinite-clan magmas are regarded as distinct magma-types, it should be realized that within each clan, there can be variation in the composition of the primary melts depending upon the extent and depth of partial melting (Brey 1978, Mitchell 1996). Consequently, some of the earlier-forming magmas of the nephelinite clan produced by limited partial melting of their source rocks can be melilitite-bearing, *e.g.*, at Napak. However, these are not melilitites and are typically minor components of a less strongly undersaturated, more voluminous nephelinite suite, formed by more extensive partial melting.

CONCLUSIONS

Carbonatites (*sensu lato*), when considered in terms of mineralogical-genetic classifications, can be divided into a group of *bona fide* primary carbonatites formed from diverse predominantly asthenospheric mantle-derived magmas, *i.e.*, carbonatites associated with the melilitite, nephelinite, aillikite and kimberlite clans, with the last best being termed calcite kimberlites. Carbonatites associated with the melilitite and neph-

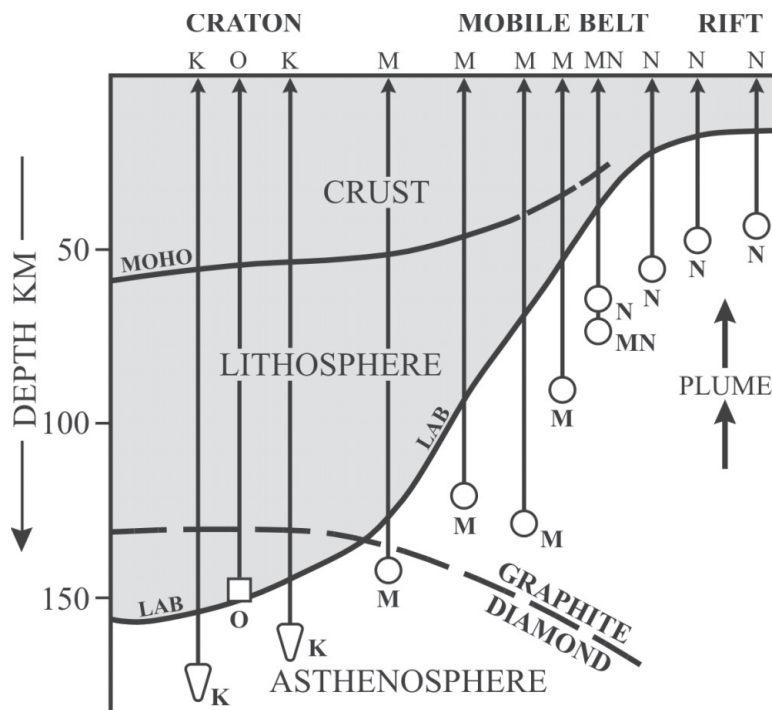


FIG. 4. Relative depths of origin of diverse mantle-derived undersaturated melilitite and nephelinite magmas in a plume-related extensional tectonic setting. Kimberlites and orangeites, which occur only in stable cratonic regions, are not considered to result from plume-related magmas. K: kimberlite, M: melilitite, MN: melilitite nephelinite, N: nephelinite, O: orangeite, LAB: lithosphere–asthenosphere boundary.

elinite clans have diverse origins and can be formed by partial melting, fractional crystallization and liquid immiscibility.

A second group of carbonate-rich rocks associated with potassic and sodic peralkaline magmas, perhaps derived from predominantly metasomatized lithospheric mantle, together with REE-carbonate-rich rocks of indeterminate genesis, are best termed carbothermal residua rather than carbonatite (*sensu stricto*). There can be mineralogical convergence between these rocks and low-pressure REE-rich derivatives of *bona fide* primary carbonatites.

A third group of carbonate-rich rocks formed by pneumatolytic melting of crustal rocks should not even be considered as carbonatites (*sensu lato or stricto*). The origin and genetic relationships of the Oldoinyo Lengai natrocarbonatite cannot be unambiguously determined, and they are regarded as a distinct variety of primary carbonatite (*sensu stricto*).

In summary, this work confirms the conclusions of Bell *et al.* (1998) and Gittins & Harmer (2003) in recognizing the possibility that many genetically

distinct varieties of carbonatite (*sensu stricto*) can exist. Novel contributions of this work are redefinitions of carbonatites and associated silicate–carbonate rocks, together with the recognition of a division between primary carbonatites and carbothermal residua. To paraphrase H.H. Read, there must be **carbonatites** and **carbonatites** and carbonatites, in addition to a wide variety of “pseudocarbonatites”!

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