Dawson's Oldoinyo Lengai calciocarbonatite: a magmatic sövite or an extremely altered natrocarbonatite?

J. GITTINS
Department of Geology, University of Toronto, Toronto, Ontario, Canada M5S 3B1
AND
R. E. HARMER*
Department of Geology, University of Pretoria, Pretoria 0002, South Africa

Abstract

In 1962 Dawson described a calcite carbonatite (specimen BD83) from the Tanzanian volcano Oldoinyo Lengai as a sövite, thus implying that at an earlier stage in its evolution this volcano had crystallized magmatic calciocarbonatites as well as the highly alkalic natrocarbonatite lava that has been erupted in more recent times. This proposition is difficult to reconcile with the currently fashionable hypothesis whereby the natrocarbonatite lava separated immiscibly from a type of nephelinite magma, most recently thought to be a wollastonite nephelinite. In 1993 Dawson sought to discredit the magmatic origin of this sövite specimen by arguing that it was derived from natrocarbonatite lava through extreme alteration (calcitization) in which process the original nyerereite was replaced by calcite in near-perfect pseudomorphs. We suggest that the arguments advanced in support of this concept are unconvincing and that the specimen is exactly what it was originally described as, namely a magmatic sövite in which the calcite crystallized from a magma rather than having replaced nyerereite. We do not seek to discredit the liquid immiscibility hypothesis but do believe that whatever process is responsible for the Oldoinyo Lengai natrocarbonatites and silicate rocks must also allow for the crystallization of calciocarbonatite.

KEYWORDS: sövite, altered natrocarbonatite, Oldoinyo Lengai, calcitization, Tanzania.

Introduction

WHEN is a sövite not a sövite? What may seem like a rather semantic question, especially when it concerns a small rock specimen (catalogue number BD83) from a remote Tanzanian volcano, assumes greater significance when the particular rock figures in discussions of the origin and differentiation of alkali-rich carbonatite magma.

Dawson (1993) seeks to discredit as a magmatic sövite the specimen of calcitic carbonatite (calciocarbonatite in the classification of Woolley and Kemp, 1989) that he described (Dawson 1962, 1989) from the volcano Oldoinyo Lengai. He now argues that it crystallized as a natrocarbonatite and has been converted to a calcitic rock through a process of calcitization. The distinction is important. If it is a magmatic sövite it indicates the presence, at some earlier stage in the evolution of the volcano, of calcitic carbonatite and at least keeps open the possibility that natrocarbonatite magma may be a fractionation product of commoner carbonatite magma rather than having been generated by liquid immiscibility. If, however, the mineralogy and chemical composition of the rock result from alteration, the fractional crystallization argument loses a valuable court exhibit in the continuing debate about the origins of natrocarbonatite magma and it is likely that natrocarbonatite magma might have been developed far more commonly than is often considered. It seems worthwhile, therefore, to examine Dawson's case for re-evaluation of the origins of this possible sövite.

* Present address: Council for Geoscience, Private Bag X112, Pretoria 0001, South Africa

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Dawson (1993) describes several features of the rock. Some of these are used in support of an origin by alteration, but for the remainder it is not clear what use is being made of them. We, therefore, list them and then examine them in sequence. These features are: (*a*) textural similarity between the supposed sövite and natrocarbonatite; (*b*) stable isotope chemistry; (*c*) chemical composition of the calcite that makes up most of the rock; (*d*) similarity of a spinel to those that occur in natrocarbonatite; (*e*) the presence of two types of apatite that are different from those of plutonic or hypabyssal sövites; (*f*) similarity of the *REE* concentrations and chondrite-normalised *REE* distribution patterns to those of natrocarbonatites; and (*g*) the presence of a Ba- and Mn-rich mineral thought to be similar to romanechite.

Discussion

Texture of the rock

The texture is well illustrated in Fig. 1 of Dawson (1993) and is identical to the type of Oldoinyo Lengai natrocarbonatite which is entirely quenched liquid and lacks phenocrysts of nyerereite and gregoryite. It is also very fine-grained, with calcite laths commonly between 0.5 and 1 mm long, as befits a largely quenched liquid. If this rock occurred in any other geological setting than a natrocarbonatite volcano its texture would have attracted little attention. It would have been interpreted without question as consisting of calcite that had crystallized from a calcitic magma during rapid quench cooling. However, Deans and Roberts (1984) sought to explain calcitic lavas from other volcanos as the result of 'calcitization' of the mineral nyerereite because of the perceived impossibility of calcite crystallizing from a carbonatite magma at atmospheric pressure and, ever since, it has been fashionable to ascribe such an origin to all lavas that contain calcite in interlocking needles or blades.

Gittins and Jago (1991) discussed the proposed alteration of natrocarbonatite to produce calcitic lavas and rejected the concept. They showed that crystallization of calcite at atmospheric pressure from a carbonatite magma is readily achieved if the magma has a moderate fluorine content and, hence, that replacement hypotheses are unnecessary. None of the arguments advanced in that paper has been addressed by Dawson in his 1993 discussion.

Dawson *et al.* (1987) sought to define the course of calcitization by reference to a partly altered natrocarbonatite (specimen number GA47), and the 1993 paper adds a description of another specimen from the same locality. Both consist of nyerereite laths, partly replaced by pirssonite, and a matrix of pirssonite and calcite in roughly equal amounts. It was Dawson's suggestion that this rock represents an intermediate stage in the calcitization of a natrocarbonatite. There can be no doubt that the rock is a natrocarbonatite in which nyerereite is partly replaced by pirssonite, but is it reasonable to extrapolate from it to a calcitic lava in which lath-shaped calcites are to be seen as perfect pseudomorphs? In our opinion this is an unjustifiably large interpretative leap.

First of all, as Gittins and Jago (1991) emphasized, any conversion of nyerereite into calcite involves a 57% reduction in molar volume and the production of a hexagonal unit cell from an orthorhombic cell. Although nyerereite crystallizes from the lava in the high nyerereite (hexagonal) form it inverts on cooling to an intermediate hexagonal form and finally (at 292°C) to the stable orthorhombic form (McKie and Frankis, 1977; Chang *et al.*, 1996). Calcitization, if it were to occur, must then have operated on the orthorhombic form. The idea of generating perfect pseudomorphs while reducing molar volume by more than one half and changing crystal systems strains credulity. Furthermore, the sequence proposed by Dawson involves three steps that seem to us unlikely:

(i) hydration of nyerereite to pirssonite

$$Na_2Ca(CO_3)_2 + 2H_2O = Na_2Ca(CO_3)_2{\cdot}2H_2O$$

(ii) leaching of Na from pirssonite, and (iii) subsequent dehydration of pirssonite to calcite.

$$Na_2Ca(CO_3)_2{\cdot}2H_2O = CaCO_3 + NaHCO_3 + NaOH + H_2O$$

Any such process must produce a rock of substantially smaller volume than the original natrocarbonatite unless Ca is introduced concurrently with the removal of Na. Yet, the ground water generated by these processes must be charged with sodium bicarbonate and sodium hydroxide and will rapidly approach saturation in these components as it percolates through the lavas. It is difficult to imagine such a solution carrying appreciable amounts of Ca, even if there were an obvious source for the Ca, and even more difficult to imagine why such a solution might be induced to precipitate the Ca at the same time as it dissolves even more Na.

Dawson points out (p.93) that the calcite laths and needles "are not single crystals but consist of numerous tiny coalescing calcite grains". The implication is that, consequently, they can not have crystallized from a magma. However, Gittins and Jago (1991) addressed this point and said that the temperature of the carbonatite magma was probably high enough for the calcite in such lavas to have crystallized as Calcite IV or V and subsequently inverted to Calcite I, an inversion that is likely to generate a mosaic texture of the type seen in this rock.

The similarity in appearance of BD 83 to natrocarbonatite can not be disputed, but at the same time it has to be accepted that the texture of the supposed sövite is a perfectly valid quench texture in its own right for which there is ample petrographic precedent elsewhere. It is also interesting that no claim has ever been made for the perfect pseudomorphing of gregoryite by calcite, yet if calcitization of one Na-K-Ca carbonate (nyerereite) can occur why can it not equally affect another (gregoryite), which is as abundant as nyerereite in most natrocarbonatites. We conclude that textural similarity is not a convincing proof of the alteration hypothesis for the origin of the supposed sövite BD 83.

Stable isotope chemistry

Specimen BD 83 is shown to have "considerably heavier oxygen" (Dawson, 1993, Fig. 6) than is present in fresh natrocarbonatite. This " ... is indicative of equilibration with meteoric water". We do not disagree, but the C and O isotope composition has no relevance to the debate about the origin of the rock. It merely shows that it has exchanged its oxygen, a feature that it shares with most igneous rocks in the earth's crust.

Chemical composition of calcite

Calcite in the disputed rock has high contents of Sr and Ba. In the lath-shaped crystal aggregates it has up to 0.56 wt.% SrO and 0.10 wt.% BaO, while the inter-granular calcite, between the laths, has up to 1.63 wt.% SrO and 1.44 wt.% BaO. The significance of these compositions is not discussed further but we presume that it is considered to be an argument in favour of a calcitization origin for the supposed sövite.

Perhaps Dawson intended to infer that the high Sr content is inherited from nyerereite, which commonly contains up to 2.5 wt.% SrO. Yet, surely, the Sr and Ba content is at least equally an argument for an original magmatic origin. The common course of element fractionation in carbonatite magmas is toward enrichment in both Sr and Ba, to the extent that veins of strontianite and barite are not uncommon in late-stage carbonatites. It is especially significant that the interstitial calcite in BD 83 (and the identical rock BD 4162) has a far higher Sr and Ba content than the lath-shaped calcite, for this is exactly the order that would be expected if the calcite laths crystallized from the magma as early phenocrysts to be followed by inter-granular crystallization of the final stage, highly evolved liquid. Indeed, Deans and Roberts (1984), in describing lavas that were considered to have laths of magmatic calcite as single crystals, as well as laths of granular calcite that were thought to represent calcitized nyerereite, drew attention to the higher Sr content of the single-crystal calcites and argued that this was further evidence of their direct magmatic origin.

The chemical composition of calcite in the supposed sövite seems to favour crystallization from a magma rather than alteration of nyerereite.

Nature of the spinel minerals

Spinel in the supposed sövite is said to belong to "a distinctive set of spinels that occur in all the Oldoinyo Lengai carbonatite lava" and that "other members of this set are found in pristine natrocarbonatite". It is further said that this set of spinels contrasts "with other Oldoinyo Lengai spinels". The argument advanced by Dawson is essentially as follows: all natrocarbonatites contain a distinctive spinel; specimen BD 83 contains a similar spinel; *ergo* BD 83 was derived from a natrocarbonatite. There are difficulties with this argument. If the calciocarbonatite (BD 83) is a sövite of direct magmatic origin it would be comagmatic with the natrocarbonatites, and might be expected to contain spinels that were part of a suite of compositions. After all, there is considerable compositional variation within the "distinctive set of spinels" presented in Table 3 (p. 96). The greatest compositional contrast in the spinels of Table 3 is with those from the ijolites and nepheline syenites. It is an interesting point that, if the natrocarbonatites are derived from nephelinite magma, as required by the currently popular liquid immiscibility hypothesis, one might expect a less extreme contrast. Yet, if the carbonatite lavas are not derived directly from the magma that produced the silicate rocks, there is no reason why their spinels should not have different compositions. We find the spinel argument unconvincing.

Apatite compositions

It is stated (pp. 97–98) that the supposed sövite contains two types of apatite that are morphologically and chemically distinct from each other and "from high-alkali fluor-apatite that occurs in pristine natrocarbonatite". The fact that these apatites are different from those of natrocarbonatite seems a poor argument for the supposed sövite having a natrocarbonatite precursor. Indeed, it seems to be a reasonable argument for the precise opposite.

While discussing apatite it is also instructive to examine Dawson's Fig. 3 which is a pair of backscattered electron images of the matrix of the supposed sövite. The images show calcite laths and apatite grains in a form that is a very convincing example of a quench texture in which co-crystallized calcite and apatite are in a micrographic to dendritic relationship. For the calcite in this texture to be derived from nyerereite the natrocarbonatite magma would have had to crystallize a nyerereite–apatite mixture with some fluorite. This is unlikely. Jago (1991) has shown that, although some apatite crystallizes from such magmas it is in very small amount and not likely to be part of the late, evolved carbonatite liquid.

In short it is difficult to be persuaded that the nature of the apatites supports a calcitization hypothesis.

Similarity of the REE concentrations and the chondrite-normalised REE distribution patterns to those of natrocarbonatites

These similarities do not appear to have any great significance. *REE* concentrations and distribution patterns with such extreme La/Yb ratios are not uncommon in carbonatites. Oldoinyo Lengai is a most unusual carbonatite; indeed, it is probably unique. If the rock that we are discussing is a magmatic sövite from an earlier stage in the evolution of such a carbonatite complex its *REE* characteristics might or might not be identical to the natrocarbonatites. Similarity between this rock and natrocarbonatite does not require it to be derived from natrocarbonatite. It also seems highly unlikely that a weathering process could leach more than one half of a natrocarbonatite lava (as Na), introduce a corresponding amount (as Ca) and still preserve the original *REE* distribution and content.

The presence of possible romanechite

A brief account of material that is dominantly Mn and Ba concludes that it is most probably romanechite. Since the only other known occurrence of romanechite in a carbonatite appears to be due to weathering, Dawson concludes that the presence of romanechite in the supposed sövite (BD 83) is evidence that the rock has been weathered. We have no quarrel with this conclusion, but it is difficult to be convinced that it in any way supports the calcitization hypothesis. The supposed sövite may well be partly weathered given its position in the volcanic edifice; indeed, it would be surprising if it were not, but no conclusions beyond that seem to be justified. The possibility of its being partly weathered in no way precludes a magmatic origin, and it is certainly no argument in support of calcitization.

Fluorine content of the supposed sövite

Although Dawson does not discuss the fluorine content of the supposed sövite (BD 83) we believe that it is important. Natrocarbonatites are noted for their very high F contents, up to 4.5 wt.% in the most completely aphyric (phenocryst-free) types, and it is to the phenocryst-free type that Dawson makes the closest comparison in his paper. Yet the supposed sövite contains only *0.18 wt.%* F. One might expect the argument to be that fluorine has been leached during the calcitization process but this is not feasible. Almost all of the fluorine in natrocarbonatite is contained in fluorite, a mineral that is almost insoluble in ground waters. The very low fluorine content of specimen BD 83 requires explanation if an alteration (calcitization) origin is to be accepted.

Conclusions

Dawson's paper presents some interesting data but they do not convince us that the calcite, which comprises most of the rock, has a non-magmatic origin. Indeed, some of the data seem to make a case for the opposite of what Dawson is trying to prove. The paper once again raises the question of whether some calcitic carbonatite lavas are the result of calcitization of former natrocarbonatite. We detect a shift in the position of the "calcitizers" in that at least some of the calcite-bearing lavas "... undoubtedly represent extrusion and consolidation of primary calcium carbonate liquid" (Dawson, 1993, p. 98), but it seems to us that Gittins and Jago (1991) set down some carefully drawn arguments against the whole concept of converting nyerereite-bearing lavas into calcitic ones in which the calcite perfectly pseudomorphs the allegedly pre-existing nyerereite. None of these points has even been referred to by Dawson (1993) and we do suggest that they deserve a reasoned answer.

Some of Dawson's arguments have the quality of circular reasoning. Thus, on p. 99 under the heading "New features compatible with alteration of the protolith.", feature number 4 states "The bulk chemistry of BD 83 represents the culmination in a 'trend' towards Ca enrichment, and Na, K, Cl and S depletion, already noted in partly altered natrocarbonatite". A 'trend' exists only if one accepts that the sövite is indeed the ultimate product of a replacement process observed in some natrocarbonatites. Specimen BD 83 and natrocarbonatites are two extremes. We contend that no data have been presented to link them genetically, and so to say that BD 83 represents the culmination of a 'trend' is to allow the conclusion to drive the reasoning rather than for reasoning to lead to a conclusion.

We do not disagree that an interesting argument exists for the natrocarbonatite lavas of Oldoinyo Lengai having been derived by liquid immiscibility. However, we suggest that there remains a strong case for the calcitocarbonatite BD 83 being exactly what it was originally described as by Dawson (1962), namely a sövite. Until more convincing arguments are advanced we believe that the possibility of Oldoinyo Lengai exhibiting evidence of the presence of sövites at some earlier stage in its history remains a viable one. To the conclusion that "...sövite must be deleted from the list of rock-types known to exist at Oldoinyo Lengai" we must demur for the time being.

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