

The Comparability of Carbonatitic Fluid Inclusions in Ijolites with Natrocarbonatite Lava

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ABSTRACT

The composition of carbonate, sulphate, halide and other phases trapped as fluid inclusions in apatite crystals in ijolite and urtite from sub-volcanic complexes in East Africa have been determined using the electron micro-probe. The bulk composition of these inclusions is alkali-rich and closely comparable to the natrocarbonatite lava from the active volcano of Oldoinyo Lengai in N. Tanzania. The inclusions are interpreted as having been originally immiscible alkali carbonate-rich droplets within a nephelinitic melt. The evidence suggests that magma of the composition of natrocarbonatite lava is that most likely for the parent magma during carbonatite petrogenesis.

INTRODUCTION

Igneous primary fluid inclusions trapped in an early liquidus phase mineral such as apatite, can provide information on the chemical composition of the host melt, which is not otherwise available due to such effects as alkali and volatile loss during crystallization.

The purpose of this contribution is to show that the composition of the material trapped as primary inclusions within apatites in some East African sub-volcanic ijolitic rocks is comparable to the whole rock chemistry of natrocarbonatite lava. The lava was first recorded by DAWSON (1962). The term natrocarbonatite, originally introduced by DU BOIS *et al.* (1963) for the lava and recommended by STRECKEISEN (1979), is used in preference

to the term lengaitite sometimes employed (BELL *et al.*, 1973). If the inclusions represent droplets of carbonatitic fluid or magma, then the similarity to natrocarbonatite indicates that the present bulk chemical composition of the intrusive carbonatites associated with ijolite is very different from that of the magma from which the carbonatites crystallized. This suggestion is put on a quantitative basis in this paper. The presence of carbonate droplets captured by apatites growing in a silicate melt is further interpreted as indicating immiscibility between carbonate and silicate melts.

THE INCLUSIONS

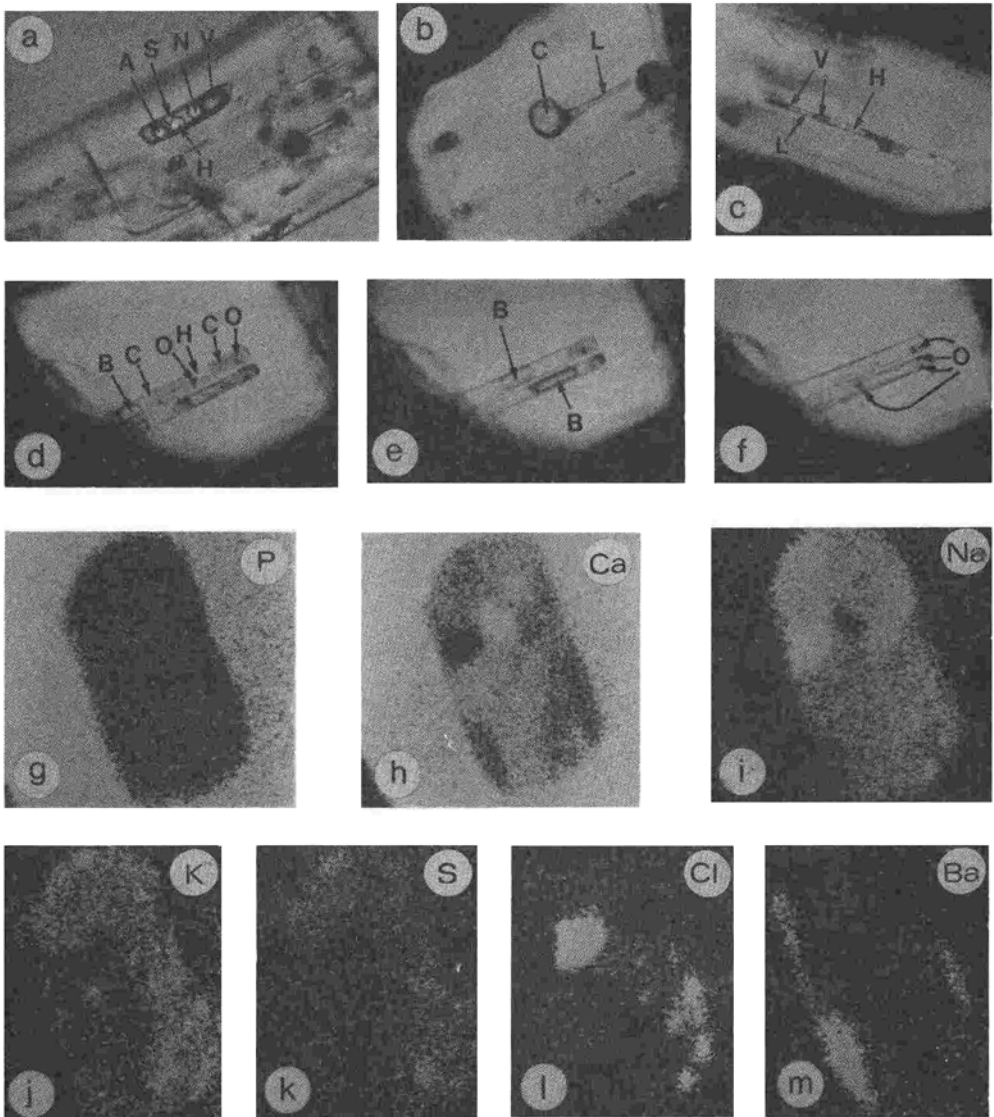
The inclusion-bearing apatite crystals were chosen from ijolites and urtites from East Africa which have been described in detail by LE BAS (1977). The ijolites form small intrusions in fenitized granitic basement and are also found as volcanic blocks ejected from Oldoinyo Lengai volcano. Their essential mineralogy is nepheline and aegirine-augite with variable amounts of melanite-garnet, apatite and sphene. Urtite intrudes the ijolite as dykes and bosses, and is composed mainly of nepheline, wollastonite, some apatite and often a little aegirine-augite. Textural evidence of apatite crystals enclosed in the silicate phases shows that apatite is the earliest phase to crystallize on the liquidus of both, apart from trace amounts of iron oxides and sulphides, but its crystallization history continues throughout most of the crystallization of the ijolites and urtites. Late apatite occurs

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interstitially. The presence of inclusions in apatites is mainly confined to the early and more prismatic apatite crystals.

Both primary and secondary inclusions are present in the apatites. Primary inclusions are derived from material trapped from the magma at the time of crystallization of the apatite; secondary inclusions

represent material subsequently introduced along fractures in the apatites during the later stages of crystallization of the host rock. The former are recognised (Fig. 1) by the frequent negative crystal habit and the high pressure phase assemblage (CO_2 is released under pressure when inclusions are opened), whereas



the secondary inclusions are usually associated with fractures, ironstaining and the inclusions are commonly almost empty apart from fluids.

The primary inclusions comprise gas, liquid and solid phases, and the compositions of the phases have been determined and reported by RANKIN and LE BAS (1973, 1974a, 1974b), LE BAS *et al.* (1977), and ASPDEN (1980, 1981). The details of the analytical methods are given in these papers, together with the corrections made for alkali-loss and decarbonation. The solid phases were analysed at 15 kV and 20nA specimen current using a 1 micron beam on a Cambridge Mark V two spectrometer Electron Micro-Probe Analyser (EMPA) housed in the Geology Department of Leicester University. The 10 sec counts (5 sec for Na and K) obtained were corrected for dead-time and ZAF using a modified MAGIC IV program. The following elements were analysed, two at a time in the order quoted, during the analysis of each solid phase in the

inclusions: Na, K, Ca, P, Ba, S, Cl, F, Si, Mg, Mn, Fe, Ti, Al, La, Ce. Probe analysis requires a polished surface on the material to be analysed, and this entails exposing the inclusion to the atmosphere. If the inclusion contained much gas or liquid, the solid contents were invariably disgorged during the polishing process. The remedy was to choose inclusions where necking had occurred whereby the fluids had separated themselves from the bulk of the solids (Fig. 1b) by a process of interaction, solution and re-precipitation between the fluids and the host apatite at the end and side walls of the inclusions. A basic assumption made in this contribution is that if a large enough number of samples are taken, the sum composition of the phases in the solid inclusions adequately represents the solid fraction and daughter products of the original fluid inclusion.

Any apatitic component of the trapped material would not be determined because any such component would, on

← FIG. 1 - (a) Photomicrograph in plane polarized light of apatite crystal from the Uyi ijolite (U 849) at Wasaki Peninsula near Homa Bay, W. Kenya (LE BAS, 1977). It is in immersion oil (R.L = 1.57). The apatite crystal which is 80 μm across contains several fluid inclusions with vapour (V), liquid and crystals (A - alkali sulphate, S - sylvite, H - halite, N - nahcolite).

(b) Photomicrograph in crossed polars of broken apatite crystal 60 μm across extracted from Usaki ijolite (U 488), W. Kenya. The large inclusion in the middle of the apatite crystal is composed of an aggregate of carbonate and sulphate crystals (C) with an opaque phase near the end (on the right) of the elongated and necked portion of the inclusion. This elongated portion is filled by liquid (L); no vapour remains.

(c) Photomicrograph in crossed polars of apatite crystal 50 μm wide in immersion oil, extracted from Uyi ijolite as (a). The central inclusion shows necking with the vapour and liquid phases restricted to the narrower portion of the negative crystal form of the inclusion. V - vapour, L - liquid, H - halite, the remaining crystals are mainly carbonates.

(d), (e) and (f) Photomicrographs taken in crossed polars of an apatite crystal from Usaki ijolite (U 1256), W. Kenya. The crystal is immersed in cold dilute (10%) HCl, and contains two inclusions each about 40 μm long. The grain has been crushed with a steel probe and the inclusion contents exposed to the acid. In (d) a bubble (B) of CO_2 can be seen in the mouth of each inclusion. The solids inside the inclusion include carbonates (C), halite (H) and opaques (O). (e) shows the reaction after a few minutes at a stage when the acid fills much of the inclusion having dissolved away most of the contents, accompanied by periodic expelling of the CO_2 produced. (f) shows the reaction gone to completion with all the inclusion materials dissolved away except for the opaque phases (O) which remain unattacked. The opaque phase (O) midway along the upper inclusion in (d) has been swept away by the expelled CO_2 .

(g) to (m) Series of X-ray back-scatter images of an inclusion in apatite from Usaki ijolite (U 366). The white areas indicate the presence of the elements being analyzed, and the intensity is proportional to the amount present. The inclusion is made mainly of alkali-bearing carbonates together with calcite, halite and some Ba carbonates at the edges (see Fig. 2a). The inclusion is 30 μm wide, and all the elemental images (g) to (m) are printed to the same scale to aid comparison. The black lines at the lower left corners of (g) and (h) mark the edge of the apatite crystal. The elements analyzed are (g) P, (h) Ca, (i) Na, (j) K, (k) S, (l) Cl, (m) Ba. No Al, F, Fe, Mg or Si were detected.

TRAPPED AND DAUGHTER MINERALS

Ca-Na-K Carbonates

Seven typical analyses including two co-existing pairs are given in Table 1. These carbonates divide into two main groups: the Na-rich and the Ca-rich, as seen in Fig. 3.

The carbonates in the ijolitic inclusions cluster in one or other of two elongate groups centred around $CC_{11} NC_{88} KC_3$ and $CC_{55} NC_{41} KC_4$, the former approximating to gregoryite and the latter to nyerereite, which minerals have recently been analysed by GITTINS and MCKIE (1980) and MCKIE and FRANKIS (1977). Some more sodic carbonates are apparently pure Na ones, such as nahcolite which has been previously described from these rocks (RANKIN and LE BAS, 1974). In the urtites, the more Ca-rich carbonates include a marked cluster around $CC_{74} NC_{19} KC_7$ with a scatter towards more sodic compositions approximating to shortite (see Fig. 3), and these co-exist with strongly Na-rich carbonates as shown by the tie-line 5-6.

Two significant features are apparent from Fig. 3. The first is the similarity of the pair of carbonate phases from the ijolitic inclusions to the two constituent carbonate phases of the natrocarbonatite from Oldoinyo Lengai, which suggests similar geochemical parentage. The second is that the tie-line for the co-existing pair 5-6 in Fig. 3 spans the cotectics and thermal divide. These boundaries were determined for the system at 1 kb (COOPER *et al.*, 1975), and GITTINS and MCKIE (1980) have shown they are appropriate to the conditions of formation of the natrocarbonatite lava. However, the daughter products plotted on Fig. 3 do not all fit these phase relations and it may be that they reflect higher pressure conditions, perhaps equivalent to deeper in the African lithosphere, but which pressure is now preserved trapped and encapsulated in the «fluid» inclusions. It seems unlikely that the few per cent SO_3 , Cl and F recorded in the natural systems would affect this.

TABLE 1 - EMP analyses of Ca-Na-K carbonates in multisolid inclusions, given in wt. %

Analyses 1, 2, 3, 4 from ijolite U 366; 5, 6, 7 from urtite U 79, all from Usaki complex, Homa Bay, W. Kenya. Analyses 1 and 2, and 5 and 6 are co-existing pairs. Analyses 3 and 7 are of carbonates similar to 2 and 6 respectively. Analysis 4 is a frequent and more Ca-rich carbonate phase in the ijolite. Further analyses are given in ASPDEN (1977).

	1	2	3	4	5	6	7
CaCO ₃	11.77	40.93	38.96	56.08	6.87	61.11	71.49
Na ₂ CO ₃	79.58	45.15	55.72	36.79	81.99	29.45	20.05
K ₂ CO ₃	4.60	6.22	2.49	5.10	3.04	9.40	7.42
Total	97.95	91.40	97.16	97.97	93.90	89.96	98.95

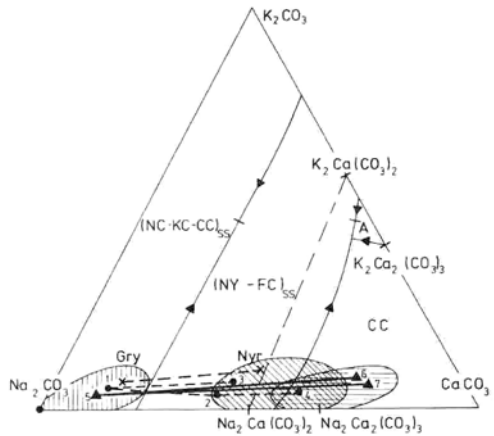


FIG. 3 - Wt. % compositions of Ca-Na-K carbonate pairs from ijolites (circles) and urtites (triangles) in the system $CaCO_3-Na_2CO_3-K_2CO_3$, after COOPER *et al.* (1975). The nos. 1-7 are analyses quoted in Table 1. The vertically ruled area is the field of Na-rich carbonates in the inclusions within both ijolites and urtites; the diagonally ruled area is the field of Ca-rich mixed carbonates in the ijolite inclusions and the horizontally ruled area that for the urtite inclusions. Crosses mark the compositions of the two main phases in natrocarbonatite: Gry - gregoryite, Ny - nyerereite (natural). The tie-lines of co-existing pairs of phases are indicated. The solid lines with arrows are cotectics and the dashed lines is the NY-FC tie-line. Phases are NC - Na_2CO_3 ; KC - K_2CO_3 ; CC - $CaCO_3$; NY - $Na_2Ca(CO_3)_2$ Nyerereite (synthetic); FC - $K_2Ca(CO_3)_2$ Fairchildite; A - $K_2Ca_2(CO_3)_3$. $Na_2Ca_2(CO_3)_3$ is shortite.

Ba-Ca Carbonates

Ba-rich carbonates are fairly common in the inclusions within apatites of both ijolites and urtites (Fig. 2), and appear as strongly birefringent rhombohedral crystals. Ba in natrocarbonatite averages 1 wt. % and it apparently lies mainly in the glassy groundmass. EMP analysis of the Ba carbonates in the inclusions suggests two compositional groups (Fig. 4), one corresponding to barytocalcite ($\text{BaCa}(\text{CO}_3)_2$), the other being intermediate between barytocalcite and witherite (BaCO_3).

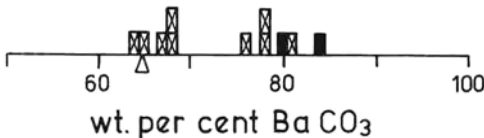


FIG. 4 - Percentage BaCO_3 in Ba-rich carbonates within inclusions, showing bimodal occurrence.

Crossed rectangles = ijolite host; Solid rectangles = urtite host; Triangle = barytocalcite.

Sulphates, Halides, Opaques and Silicates

Barium sulphate is rare, having been detected in only two inclusions, both in ijolite. EMP semi-quantitative analysis of a small crystal of baryte gave BaO 54.2 and SO_3 28.5 wt. %. A larger crystal of fluorite gave Ca 52.8, and one halite gave Na 38.2, Cl 54.9 wt. %. Sylvite has been identified only by back scatter imagery.

Sodium-potassium sulphates are common and constitute about 11% of the total solid bulk of the inclusions in the ijolites, but only 4% in the urtites. One EMP analysis of an alkali sulphate gave 43.1 wt. % Na_2O , 35.3% SO_3 and showed only a trace of K_2O , and no other elements were detected. Two other sulphates gave similar results. Despite low totals, all gave $\text{Na}_2\text{O}/\text{SO}_3$ ratios of 1.2 to 1.3 which is much too high for thenardite (Na_2SO_4) which has a ratio of 0.8. The only known mineral of appropriate

composition is burkeite ($\text{Na}_6(\text{CO}_3)(\text{SO}_4)_2$) where the ratio $\text{Na}_2\text{O}/\text{SO}_3$ is 1.2 and Na_2O is 47.7 wt. %. A sodium-rich member of the apthitalite group $\text{KNa}_3(\text{SO}_4)$ also appears to be present.

Other solid phases in the inclusions include some iron-rich opaques and EMP analysis of several grains indicated pyrrhotite, with Fe 59.90 and S 33.21 giving almost stoichiometric FeS . Some of the iron-rich opaques are not sulphur-bearing, nor contain any Ti, and are thought to be oxides, probably magnetite.

Inclusions of silicate material are not uncommon, usually about 5% of the whole. They occur as minute very weakly anisotropic specks, and are similar to those described from other inclusions in the same rocks (RANKIN and LE BAS, 1974b; LE BAS *et al.*, 1977).

MODAL PROPORTIONS AND BULK COMPOSITION OF THE PHASES

The approximate modal proportions of the minerals in the inclusions (Table 2) was deduced from estimating the area covered by the individual minerals in 36 inclusions of which 8 are illustrated in Fig. 2. These modes have been calculated from X-ray back scatter photographs as shown in Fig. 1, g-m. Photographs were taken for each element detected (Na, K, Ca, P, Ba, Sr, S, Cl, F, Si, Mg, Mn, Fe, Ti, Al, and sometimes La and Ce) for each inclusion. Each set of photographs was printed to the same scale so that those for each inclusion could be superimposed and the relative compositions of each of the phases qualitatively estimated. The elemental images were in most cases quite clear cut, and there was no doubt about the identification. Some areas on the photographs, however, did not appear to be composed of single phases but of intimate mixtures or perhaps mixed crystals. These could not be resolved beyond estimating, for instance, that they were mixed calcium and sodium and potassium carbonates or bicarbonates. Sylvite and halite were sometimes mixed, and the data in Table 4 has had to be estimated

TABLE 2 - Mean modal proportions (vol. %) of the mineralogical composition of inclusions in: (1) ijolite (13 samples), (2) urtite (15 samples), from East Africa.

	(1)	(2)
Ca-Na-K carbonates	57	79
CaCO ₃	5	2
Ba-Ca carbonate	7	6
Sr-rich carbonate	1	0
Alkali sulphate	11	4
Ba sulphate	tr	0
Halite	6	0.7
Sylvite	6	1.3
Fluorite	2	0
Silicates	4	7
Fe-rich opaques	1	tr

according to the relative intensities of K and Na.

Whilst the presence of sulphate and halide minerals could be determined directly from the presence of S, Cl or F, the occurrence of carbonate, bicarbonate and oxide minerals had to be deduced from the absence of other anionic groups. This, coupled with the optical properties and calculations based on the stoichiometry of minerals, usually permitted the phase to be adequately identified.

The figures obtained in Table 2 do not show any marked contrasts, allowing for the large errors inherent in this type of semi-quantitative determination, as found by NESBITT and KELLY (1977) in a similar calculation, except perhaps for the halide and sulphate minerals. The main difference lies in the average composition of the Ca-Na-K carbonate which is $CC_{40}NC_{54}KC_6$ in the ijolite (wt. percentages derived from the mean ijolite point in Fig. 3), and $CC_{52}NC_{42}KC_6$ in the urtite.

When the data of Table 2 are recalculated into weight percentage oxides (Table 3), a greater contrast between the ijolite and urtite inclusion data is seen. One

significant feature of Table 3 is the close chemical comparability between the ijolite fluid inclusion composition (column 3a) and that of the natrocarbonatite extruded from Oldoinyo Lengai volcano in 1960 (columns 1 and 2), whilst that for the urtite (column 4a) is slightly different.

The compositions calculated in Table 3 of the materials trapped in the apatite crystals in ijolite and in urtite have been estimated for two cases: first, trapped solids only (columns 3 and 4) and second, making allowance for the presence of about 10% saturated alkali chloride solution (Na:K = 1.6:1 by weight) in addition to the solids (columns 3a and 4a). The contribution by any vapour phase present (mainly CO₂) would be negligible. It may also be noticed in Table 3 that the compositions of these inclusions are quite different from those of the associated and perfectly normal carbonatite intrusions nearby (column 5). This reinforces the arguments made elsewhere (LE BAS, 1977, 1981) that the composition of carbonatitic magma is not the same as that of the solid carbonatitic plutonic product.

DISCUSSION

The EMP study of the material trapped as inclusions in ijolitic apatites supplies chemical data on the composition of the carbonatitic melts. The fact that the compositions closest to natrocarbonatite occur within apatites in the ijolites, combined with the fact that these apatites were the earliest main phase to crystallize in the ijolitic/nephelinitic magma, indicates that the carbonatitic droplets trapped by the apatites co-existed with the ijolitic/nephelinitic melt. They must therefore be immiscible droplets of carbonate in silicate melts, as has been shown experimentally for synthetic compositions (KOSTER VAN GROOS, 1975; VERWOERD, 1977; HAMILTON *et al.* 1979; FREESTONE and HAMILTON, 1980).

Having determined the approximate compositions of immiscible pair of liquids, *i.e.* the carbonate from Table 3 column 3a, and the silicate from average ijolite (given in Table 4, column 2), it is possible

TABLE 3 - Estimated compositions (wt. per cent) of carbonatites and of the liquid available for trapping by apatite in ijolitic and urtitic melts.

	1	2	3	3a	4	4a	5
SiO ₂	1.2	-	2.1	2.0	3.8	3.7	1.83
Al ₂ O ₃	1.7	0.2	1.0	1.0	1.9	1.8	1.33
FeO		0.3	0.9	0.9	-	-	2.88
MgO	1.9	0.4	-	-	-	-	0.71
CaO	18.3	14.5	19.8	19.1	25.8	25.0	50.15
BaO	1.0	1.1	4.5	4.4	4.0	3.9	0.28
SrO	0.9	1.3	0.5	0.5	-	-	-
Na ₂ O	29.5	32.2	26.1	26.3	20.6	20.9	-
K ₂ O	7.2	6.8	5.4	6.1	5.2	5.9	-
CO ₂	31.4	32.5	30.2	29.2	36.2	34.9	37.49
SO ₃	2.9	2.5	4.5	4.4	1.9	1.8	0.34
Cl	2.6	2.4	5.3	6.9	0.8	2.6	-
F	-	2.1	1.4	1.4	-	-	0.20
		102.1	101.9	102.2	100.2	100.5	
less O ≡		1.5	1.9	2.2	0.2	0.5	
(Cl,F,S)		100.6	100.0	100.0	100.0	100.0	

1. Mean composition of natrocarbonatite lava from DU BOIS *et al.* (1963). No fluorine could be detected in their samples although fluorite was observed, but the fluorine content in the samples collected by DAWSON average 2.3% (1962).

2. Mean composition of natrocarbonatite lava from GITTINS and MCKIE (1980). Analysis includes MnO 0.4, P₂O₅ 1.0, S 0.2, H₂O ± 4.2.

3. Anhydrous composition of mineral assemblage inside inclusions in ijolite as listed in Table 2 column 1. The Ca-Na-K carbonate is calculated as CC₄₀NC₅₄KC₆; Ba-Ca carbonate as 65% BaCO₃ (see Fig. 4); Sr-rich carbonate as SrCO₃; alkali sulphate as burkeite (Na₆(CO₃)(SO₄)₂); silicate as the mean anhydrous composition of silicate glass given in LE BAS *et al.* (1977); and Fe-rich opaques as magnetite plus pyrrhotite in the 1 : 1 proportion.

3a. As column 3 with addition of saturated solution of (Na,K)Cl where Na:K = 1.6:1 weight ratio, and assuming this solution occupies 10% of the volume of the fluid inclusion.

4. Anhydrous composition of mineral assemblage in inclusions in urtite as listed in Table 2 column 2. The Ca-Na-K carbonate is calculated as CC₅₂NC₄₂KC₆, and the other compositions are as given for column 3 of this Table.

4a. As column 4 with addition of (Na,K)Cl solution as in column 3a.

5. Typical sövite from Wasaki Peninsula, W. Kenya (LE BAS, 1977). The analysis includes TiO₂ 0.20, MnO 0.35, P₂O₅ 2.31, H₂O 1.26.

TABLE 4 - Compositions of the silicate magmas co-existing with the carbonate melts in Table 3.

1. Mean composition of nephelinitic lavas associated with the natrocarbonatite lavas at Oldoinyo Lengai (DAWSON, 1966, table III analyses 6-9).

2. Mean composition of E. African ijolite (LE BAS, 1977, Appendix 2, p. 307).

3. Composition of chilled margin of urtite intrusion, W. Kenya (LE BAS, 1977, Appendix 2, p. 308).

	1	2	3
SiO ₂	40.79	40.01	43.64
TiO ₂	2.71	2.38	0.00
Al ₂ O ₃	12.41	15.25	23.15
Fe ₂ O ₃	5.67	6.61	1.40
FeO	3.89	3.39	0.70
MnO	0.33	0.16	0.16
MgO	3.67	3.50	0.96
CaO	15.51	16.36	9.42
Na ₂ O	8.45	7.10	12.00
K ₂ O	3.35	3.10	5.30
P ₂ O ₅	1.24	0.88	0.08
CO ₂	0.76	0.56	0.57
H ₂ O ⁺	0.33	0.64	1.45
F	0.36	-	-

to plot a pair of points on the immiscibility solvus surface for the system ijolite-carbonatite (join Ij - IC on Fig. 5). A second tie-line (Ur - UC on Fig. 5) can be plotted if the carbonate material trapped by the apatites in urtite (Table 3, column 4a) is taken to represent the composition of the immiscible carbonate fraction which was exsolved from the urtitic magma. The analysis of a wollastonite-urtite (U 1111) from a chilled contact within the Usaki complex best represents the composition of the urtite magma (Table 4 column 3).

The third tie-line (Neph-NCb) represents the conjugate pair of immiscible liquids (one silicate, one carbonate) which are deduced to co-exist at Oldoinyo Lengai volcano in N. Tanzania. It is closely similar to the Ij - UC tie-line, and the data plot reasonably well on the solvus determined for the synthetic system at 1100°C and 3 kb (Fig. 5). This confirms that magma of the composition of natrocarbonatite is appropriate for that of natural carbonatite magma which can separate by liquid immiscibility from a nephelinitic or phonolitic parent magma.

Two new conclusions may be drawn:

1. The carbonate-rich primary inclusions trapped in apatite crystals from ijolitic and urtitic melts can provide a remarkably accurate clue to the composition of carbonatite melt co-existing with silicate melt.

2. The close similarity of these trapped carbonate-rich inclusions to the 1960 natrocarbonatite lava of Oldoinyo Lengai volcano indicates that the parent magma of the East African carbonatitic igneous rocks most likely had a chemical composition close to that of the natrocarbonatite lava.

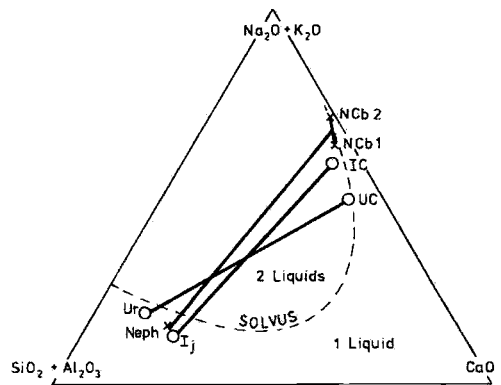


FIG. 5 - Tie-lines for estimated co-existing immiscible pairs plotted weight per cent on the synthetic system investigated by FREESTONE and HAMILTON (1980). The solvus indicated is that determined for 1100°C at 3 kb. Neph, Ij and Ur are analyses in Table 4; NcB 1, NcB 2, IC and UC are analyses in Table 3 columns 1, 2, 3a and 4a respectively.

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