

The occurrence and significance of xenocrysts of apatite, ilmenite, and Na-Fe-Ti oxide in ultrapotassic lavas from the Leucite Hills, Wyoming

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Abstract

The occurrence and mineral chemistry of apatite, magnesian ilmenite, and an Na-Fe-Ti oxide in lavas from the Leucite Hills are reported. Magnesian ilmenite and apatite occur as xenocrysts and as crystals in amphibole-mica-pyroxenite xenoliths. Na-Fe-Ti oxide and also rutile occur as inclusions in ilmenite. The latter mineral contains up to 7.2% MgO and shows evidence of oxidation by, and reaction with, the host magma. The apatite differs from that which occurs as phenocrysts and microphenocrysts inasmuch as REE were not detected. The occurrence of these minerals, which are important repositories for REE and High Field Strength elements, together with phlogopite in the upper mantle source regions of ultrapotassic lavas, is important and may explain some unusual aspects of the geochemistry of such lavas (low K/Rb, P_2O_5/Ce , Ti/Zr, high Ti/V, Zr/Nb). The source regions must be grossly heterogeneous and a two-component model is suggested for the source. This model is similar to that suggested for the source regions of other alkaline magmas and is capable of explaining the unusual Nd-Sr isotopic characteristics of the Leucite Hills lavas.

KEYWORDS: xenocrysts, apatite, ilmenite, ultrapotassic lavas, Leucite Hills, Wyoming.

Introduction

DESPITE recent intensive research, the origin of K-rich mafic-ultramafic magmas remains an enigma. Most workers agree that the ultimate source of these magmas lies in the upper mantle, and it is widely assumed that the upper mantle source regions must be highly enriched in certain incompatible elements such as K, Rb, Ba, Sr, REE, P, Zr, etc. However, experimental (Barton and Hamilton, 1979, 1982; Nicholls and Whitford, 1983) and geochemical (Kay and Gast, 1973; Vollmer *et al.*, 1984) studies have failed to resolve the precise nature of the source material. Barton and Hamilton (1979) concluded that silica-poor lamproites, such as madupite, could be derived from mica-pyroxenite, a conclusion broadly consistent with results presented by Nicholls and Whitford (*op. cit.*), though these workers favoured magma evolution via a process akin to zone-refining. On the other hand, Barton and Hamilton (1982) demonstrated that silica-rich magmas such as wyomingite and orendite may have equilibrated

with harzburgite or phlogopite-harzburgite, and a similar conclusion was reached by Arima and Edgar (1983) on the basis of an experimental study of a wolgidite from the West Kimberley area, Australia.

Barton and Van Bergen (1981) studied xenoliths and xenocryst/megacryst assemblages in the lavas of the Leucite Hills, Wyoming, and concluded that the upper mantle beneath this region was grossly heterogeneous and included both harzburgites (i.e. refractory material from which a melt had previously been extracted) and relatively iron-rich pyroxenites with accessory amphibole, phlogopite, apatite, and Fe-Ti oxide. These data suggest that the source regions of K-rich mafic-ultramafic magmas may be more complex than previously assumed. Upper mantle sources such as phlogopite-lherzolite, phlogopite-garnet lherzolite, and richterite-phlogopite harzburgite (e.g. Aoki, 1974, 1975) cannot yield magmas with the observed P_2O_5/Ce ratios or LREE enrichment factors.

Additional petrographic and mineral chemical studies have been carried out on the xenocrystal

assemblages in the Leucite Hills lavas and the results are reported here. Specifically, data for oxide phases and apatite are presented and their significance discussed. The results provide strong support for the concept of a heterogeneous source region and indicate that further studies are both warranted and essential. K-rich mafic-ultramafic lavas are unique in that they display the highest concentrations of K and associated elements of all terrestrial magmas and hence an understanding of the genesis of these lavas is fundamental for evaluation of models of mantle evolution and geodynamics.

Brief description of locality and previous work

The Leucite Hills comprise 22 exposures of lamproite in S.W. Wyoming near Rock Springs. The extreme composition of these lavas has been known since the work of Cross (1897); the lavas are ultrapotassic with up to 12 wt. % K_2O , $K_2O/Na_2O \gg 2:1$, and $(K_2O + Na_2O)/Al_2O_3 > 1$. Two broad groups of lava are distinguished on the basis of chemistry, mineralogy, and petrography (Cross, 1897; Carmichael, 1967): orendite-wyomingite (silica-saturated lavas), and madupite which is silica-undersaturated. The latter is unique inasmuch as it contains phenocrysts and microphenocrysts of magnetite and perovskite. The two groups of lava show restricted ranges of composition and phenocryst/microphenocryst phases likewise show limited compositional variation (Carmichael, op. cit.; Barton, 1979; Kuehner *et al.*, 1981).

In terms of $Mg/(Mg + \Sigma Fe)$ ratios, the Leucite Hills magmas could represent primary melts which have not been modified by fractionation during ascent. Ni contents of the wyomingites and orendites (> 240 ppm; Kuehner *et al.*, 1981; Vollmer *et al.*, 1984) are consistent with derivation from an olivine-bearing source rock, whereas Ni contents of the madupites (160–192 ppm; Kuehner *et al.*, 1981; Vollmer *et al.*, 1984; unpubl. data) suggest that olivine may have been absent from the source rock (cf. Barton and Hamilton, 1979). These lavas are thus suitable for use as probes of upper mantle composition.

Analyses of xenocrysts

All analyses were performed with an electron microprobe using both EDS and WDS techniques and appropriate standards. Operating conditions were: 15 kV accelerating potential, 3–16 nA sample current, and 40–100 seconds counting time. On-line data reduction included corrections for dead-time, background, atomic number, absorption, and fluorescence.

Fe–Ti oxide occurs as resorbed or rounded anhedral grains less than 1 mm in diameter in the lava and in clinopyroxene-rich xenoliths. Analysis indicates that the Fe–Ti oxide is Mg-rich ilmenite (Table 1), with MgO contents up to 7.2 wt. %. Similar, Mg-rich ilmenites have been reported from kimberlites and from lherzolite xenoliths in kimberlite (Dawson, 1980; Bector and Boyd, 1981). Apart from MnO, which is present in concentrations up to 1.25 wt. %, the concentrations of oxides other than FeO, Fe_2O_3 , TiO_2 , and MgO are low. Cr_2O_3 , for example, is present in amounts below 0.34 wt. %. Fe_2O_3 contents were calculated by assuming that the cation sum is 2 on a 3 oxygen basis, and average 8.5 wt. % (or about 21 % of the total iron), which is comparable to values reported for ilmenites in kimberlites from South Africa and in lherzolite xenoliths (Dawson, 1980). However, a few ilmenites have high Fe_2O_3 , with values ranging up to 29 wt. % (about 58 % of the total iron). This is possibly due to oxidation by the enclosing magma as, in a few grains, the amount of Fe_2O_3 was found to increase towards the margins of the xenocrysts. The Leucite Hills magmas were probably highly oxidized, judging from the high Fe_2O_3/FeO ratios (Carmichael, 1967; Kuehner *et al.*, 1981).

Many of the xenocrysts analysed were found to be extremely heterogeneous with respect to TiO_2 , MgO, and FeO(total) contents. In many cases, TiO_2 increases and FeO(total) and MgO decrease towards the margin of the crystals; in some cases TiO_2 contents are highest in the cores. The precise causes of these compositional variations are difficult to ascertain as the megacrysts occur in relatively few samples and only a small number of crystals have been encountered in thin section. Nevertheless, two factors may be responsible. The first is reaction with the host magma (? preferential removal of Fe_2O_3 and MgO) and, as noted above, there is evidence for oxidation of ilmenite near the margins of the grains. However, the high TiO_2 contents of the cores of some grains are unlikely to result from oxidation and are due to the presence of minute inclusions of rutile. Needle-like inclusions were observed using back-scattered electron-imaging techniques, but could not be analysed. Na was below detection levels ($\ll 0.1$ wt. %) in the cores of the ilmenites thus far described so it is unlikely that the high TiO_2 contents reflect the presence of inclusions of the Na–Fe–Ti oxide described below.

Routine analysis of a complex intergrowth of oxide phases in a single megacryst indicate that one of the intergrowth phases has a highly unusual composition inasmuch as Na_2O contents up to 8.8 wt. % were detected. It should be emphasized that the size of the Na–Fe–Ti oxides phase ($> 10 \mu m$) allowed several spot analyses to be made and that

Table 1. Microprobe analyses of magnesian ilmenite, Na-Fe-Ti oxide, priderite and apatite.

	Ilm Core	Ilm Margin	Na-Fe-Ti Oxide Inc. in Ilm	Priderite Groundmass	Apatite Megacryst
SiO ₂	0.28	0.36	0.22	1.25	-
TiO ₂	50.47	41.86	73.11	70.08	-
Al ₂ O ₃	0.18	-	-	0.22	-
Fe ₂ O ₃ *	8.61	29.06	-	-	-
FeO*	32.59	21.14	16.12 ⁽¹⁾	11.46 ⁽¹⁾	-
MnO	1.21	1.33	0.40	0.19	-
MgO	6.68	7.18	1.98	0.98	-
CaO	-	-	-	0.12	55.28
Na ₂ O	-	0.59	8.38	0.44	-
K ₂ O	-	-	-	4.78	-
BaO	-	-	-	8.80	-
P ₂ O ₅	-	-	-	-	41.91
Total	100.67	101.51	100.21	98.32	97.19
Number of cations on the basis of					
	0=3	0=3	0=16	0=16	0=25
Si	0.007	0.009	0.025	0.158	-
Ti	0.913	0.746	6.226	6.663	-
Al	0.005	-	-	0.033	-
Fe ³⁺	0.156	0.518	1.354	-	-
Fe ²⁺	0.655	0.419	0.172	1.212	-
Mn	0.025	0.027	0.039	0.021	-
Mg	0.239	0.254	0.335	0.185	-
Ca	-	-	-	0.016	10.010
Na	-	0.027	1.841	0.108	-
K	-	-	-	0.770	-
Ba	-	-	-	0.436	-
P	-	-	-	-	5.995
Total	2.00	2.00	9.99	9.60	16.005

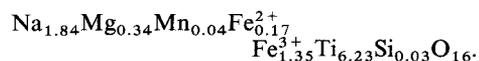
*Calculated assuming perfect analysis and ideal stoichiometry.

⁽¹⁾Total iron as FeO.

this phase is completely enclosed by ilmenite, so that the high Na₂O contents are probably not due to contamination. Furthermore, Al₂O₃, CaO, and K₂O were not detected, whereas SiO₂ was detected in minor quantities only (< 0.23 wt. %) which also suggests that the high Na₂O contents do not result from contamination. Indeed, no other phase (including interstitial glass) in the Leucite Hills lavas contains as much Na₂O (Carmichael, 1967; Barton, unpubl. data) which appears to conclusively rule out the possibility of contamination. In thin section, the Na-bearing oxide is opaque and it would therefore be easily overlooked during routine optical examination.

A representative analysis is reported in Table 1. MnO is present in low amounts (< 0.42 wt. %) along with MgO (< 2.0 wt. %) and SiO₂. The similarity to priderite (A₂B₈O₁₆ where A = K, Ba and B = Fe, Ti) is striking; an analysis of priderite from the Leucite Hills (in a Wyomingite lava from Hatcher Mesa) is presented for comparison. The latter is similar to analyses reported by Carmichael (1967). The oxide phase in the ilmenite megacryst differs inasmuch as BaO was not detected. However, the formula corresponds closely to that of priderite especially if some of the iron is in the ferric state. The amount of Fe³⁺ has been calculated by

assuming that Σ cations = 10 on a 16 oxygen basis and the corrected formula is:



It seems probable that the reported mineral is Na-priderite, although X-ray data are required to confirm this and it is unfortunate that the mineral is so intimately intergrown with ilmenite. Nevertheless it is noteworthy that ceramic engineers have synthesized alkali-bearing titanium oxides (E. Hellstrom, pers. comm., 1984) and it is probably inevitable that some of these will be found in nature.

Analysis of apatite megacrysts (Table 1) were undertaken primarily for comparison with the apatite phenocrysts in the host lavas. The latter are characterized by high REE contents (Carmichael, 1967; Kuehner *et al.*, 1981) with total REE generally > 1.0 wt. %. It seems likely that the REE substitute mainly for Ca in the apatite structure. REE were not detected in the megacrysts which have compositions close to that of pure apatite. The calculated formula is especially important as it indicates minor replacement of Ca or P by other elements. The analytical data are thus significant inasmuch as they confirm the conclusion, based upon detailed petrographic studies (Barton and

Van Bergen, 1981), that the apatite megacrysts are indeed xenocrysts.

Discussion

The results presented in this study extend the work of Barton and Van Bergen (1981) and document the complexity of the megacryst assemblages in the Leucite Hills lavas. The occurrence of magnesian ilmenite is particularly interesting as this is one of the most characteristic minerals of kimberlite (Dawson, 1980). Magnesian ilmenite may crystallize from the host kimberlitic magma or may be xenocrystal (Dawson, *op. cit.*), and evidence presented by Barton and Van Bergen (1981) indicates that the latter origin is most likely correct in the case of the Leucite Hills lamproites. The ilmenite in the latter is clearly derived from ultramafic xenoliths which are disaggregated during ascent. This mineral is relatively rare in peridotite xenoliths in kimberlite (Dawson, *op. cit.*), and has not been discovered in harzburgite xenoliths in the Leucite Hills lavas. The origin of the ilmenite xenocrysts thus lies in the disaggregation of pyroxenite xenoliths which contain, in addition to salitic pyroxene, amphibole (now represented by pseudomorphs), and apatite. The presence of mica-rich xenoliths in the xenocryst-bearing lavas was reported by Barton and Van Bergen (1981) but these workers could not determine the relationship (if any) between the mica-rich and the pyroxene-rich xenoliths. More recent work has established that eastonite-rich phlogopite xenocrysts and xenoliths (up to 50% eastonite component) in the Leucite Hills lavas are inevitably associated with relatively Fe-rich spinel (unpubl. data), and there is some evidence that the latter mineral originates by disaggregation of pyroxenite xenoliths. It seems likely that the eastonitic phlogopite develops as a result of reaction between spinel and the host magma. However, some megacrysts of relatively Al-poor phlogopite are undoubtedly derived from pyroxenite xenoliths.

The mineralogy of the pyroxenite xenoliths is to some extent similar to that of the MARID suite of xenoliths in kimberlite (Dawson and Smith, 1977). However, minerals of the MARID suite (especially phlogopite and pyroxene) are more magnesian than those in the pyroxenite xenoliths. Wass (1980) has described similar assemblages, also characterized by relatively iron-rich minerals, from xenoliths in alkaline magmas erupted in New South Wales, and it appears possible that pyroxenites are common in the source regions of many alkaline magmas and that the constituent minerals display a relatively wide range of composition—especially in terms of $Mg/(Mg + \Sigma Fe^{2+})$. Barton and Van

Bergen (1981) and Wass (1980) have argued that these xenoliths are derived from the upper mantle, as proposed also by Dawson and Smith (1977) for the MARID suite. The origin of these anomalous rocks remains to be resolved, but probably reflects either pervasive regional metasomatism (*cf.* Lloyd and Bailey, 1975) or crystallization of magmas in the upper mantle (*cf.* Irving, 1980). Wyllie and Sekine (1982) have recently shown that mica-pyroxenites could result from interaction of subducted oceanic crust and the overlying mantle. Menzies and Wass (1983) have argued that formation of pyroxenites may be accomplished by CO_2 -rich fluid, and it may thus be significant that carbonates have been discovered in the pyroxenite xenoliths from the Leucite Hills (unpublished observations).

It is premature to draw definite conclusions from the results presented in the present study or those presented by Barton and Van Bergen (1981), as the inventory of the xenocrystal and xenolithic assemblages is not yet complete. However, it is pertinent to briefly examine the possible importance of these assemblages for the genesis of ultrapotassic magmas. Some geochemical data relevant to this discussion are presented in Table 2.

As noted previously, the high $Mg/(Mg + \Sigma Fe^{2+})$ and Ni contents of the wyomingites and orendites from the Leucite Hills are consistent with derivation from an olivine-bearing, relatively refractory upper mantle source. The Cr contents (350–400 ppm) suggest that Cr-diopside and Cr-spinel were not major components of this source or were consumed prior to magma segregation. These data are consistent with derivation from a harzburgitic source such as that recorded by xenoliths in the magmas. Chromite is a minor component of the harzburgites (Barton and Van Bergen, 1981) and, moreover, the presence of phlogopite in the harzburgites provides a ready explanation for the low K/Rb ratios of the host magmas. K/Rb ratios of wyomingites and orendites fall in the range 324–364 (Barton, unpubl. data; Vollmer *et al.*, 1984) whereas K/Rb ratios for many alkaline lavas are ~ 400 (Clague and Frey, 1982). It is known that phlogopite, if it is residual after the melting event, may strongly influence the K/Rb ratios of primary magmas (Beswick, 1976). High-pressure experimental studies (Barton and Hamilton, 1978, 1979, 1982) demonstrate that olivine, orthopyroxene, and phlogopite may be residual phases after the genesis of ultrapotassic magmas, and the experimental work of Arima and Edgar (1983) confirms this. It is worth recording that xenocrysts of olivine, orthopyroxene, Cr-spinel, and phlogopite, which are compositionally identical to the minerals in the harzburgite xenoliths of the Leucite Hills lavas are

Table 2. Some relevant geochemical data for lavas from the Leucite Hills

	Wyomingite/ Orendite	Madupite	Chondrites	(1) Required amount of melting			
				Source 1 Wyomingite/ orendite	Madupite	Source 2 Wyomingite/ orendite	Madupite
K/Rb	324-364	273-324	386-432				
P ₂ O ₅ /Ce	50, 8-68	26-28	77-81				
Zr/Nb	25	10	10-16				
Ti/Zr	16	16	98-139				
Ti/V	242	174-383	8.8-10				
K ₂ O				1.2	1.9	0.8	1.3
P ₂ O ₅				3.7	3.4	2.4	2.4
Rb				0.4	0.6	1.5	2.2
Zr				1.2	1.3	0.7	0.7

Notes: ⁽¹⁾ From Clague and Frey (1982). K/Rb and P₂O₅/Ce ratios for alkaline lavas from Hawaii. Source 1 is model mantle reported by Frey et al. (1978); Source 2 is model mantle reported by Ridley and Dawson (1975). Data for Leucite Hills lavas from Vollmer et al., (1984) and Barton (unpublished analyses). Required amount of melting calculated assuming equilibrium and D₁⁰ = 0 (as is appropriate for highly incompatible elements).

present in the K-rich lavas of S.E. Spain (Barton, work in progress).

The CaO contents of wyomingites and orendites (2.8-7.3%) are inconsistent with derivation from a harzburgite source which implies that pyroxenites (probably occurring as veins) are a significant component of the source region and that the relatively low Cr content of the salitic pyroxenes (Barton and Van Bergen, 1981) accounts for the low Cr-contents of the host magmas. The occurrence of apatite in the xenoliths is also important since this mineral is a known repository for REE and the Leucite Hills lavas are strongly enriched in REE relative to chondrites (Kay and Gast, 1973). In wyomingites and orendites, P₂O₅/Ce ratios range from 50 to 68 (Vollmer et al., 1984; unpubl. data) in contrast to other alkaline lavas with P₂O₅/Ce ratios of 77 to 81 (e.g. Clague and Frey, 1982). However, it is not clear from current studies whether apatite can be a residual phase after magma genesis. The experimental results of Green and Watson (1982) suggest that P₂O₅ contents of wyomingites and orendites (1.35-2.14 wt.%; Barton and Van Bergen, 1981) are too low for them to equilibrate with residual apatite, but apatite is stable in supra-solidus experiments on the Leucite Hills lavas (Barton and Hamilton, 1979, 1982). However, the stability of this phase will depend upon the a_{H₂O} and a_F and it is thus possible that apatite is a liquidus phase under some conditions.

The other minor phases described in this paper may play a role in determining the geochemistry of the K-rich (and, perhaps other) alkaline magmas. The very high Ti/V and Zr/Nb ratios combined with the low Ti/Zr ratios (Table 2) relative to other alkaline lavas and chondrites (Wass, 1980; Clague and Frey, 1982) can be interpreted to indicate that oxides were present in the source regions and that at least some of the High Field Strength (HFS) elements were retained in these phases during magma genesis. A similar conclusion, i.e. that the

HFS elements do not show the same degree of compatibility during melting in the mantle, was reached by Clague and Frey (1982). In the case of the Leucite Hills lavas, it may be suggested that Ti, V, and Nb were retained in mineral phases relative to Zr (see Table 2). The latter element shows about the same degree of incompatibility in the Leucite Hills magmas (assuming the mantle sources identified in Table 2) as K, Ti and V may be retained in ilmenite although more work is required to establish the behaviour of V and, in particular, to determine the effect of f_{O₂} on distribution coefficients. Ti and Nb may also be retained in rutile (n.b. Boctor and Boyd, 1981, report up to 1.22 wt. % Nb₂O₅ in rutiles), and it should be emphasized that this phase, in contrast to ilmenite, occurs near the liquidus of K-rich lavas (Arima and Edgar, 1983). Other oxide phases, not yet identified in the xenoliths, could also be important repositories for HFS and other elements. Likely candidates are perovskite, an important host for Nb and LREE, and priderite. It is thus of interest that a Na-Fe-Ti oxide similar to priderite occurs in xenoliths in the Leucite Hills lavas.

Whether or not any of the minerals reported in this study do form refractory phases during magma genesis, their occurrence as xenocrysts and in xenoliths is important inasmuch as it provides strong evidence that the source regions were grossly heterogeneous. The available data suggest a two-component source, one component composed of relatively refractory harzburgite which contains phlogopite, and the other component composed of apatite-ilmenite pyroxenites which contain phlogopite and amphibole. As noted previously, apatite-pyroxenite/amphibole xenoliths form an important component of the source regions of alkaline magmas erupted in the Southern Highlands Province, Australia (Wass, 1980), and Menzies and Wass (1983) have argued that the type of enrichment events recorded by this type of

xenolith produces a unique isotopic signature—a near vertical trend on a $^{143}\text{Nd}/^{144}\text{Nd}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$ diagram. The isotopic compositions of the Leucite Hills lavas define just such a trend (Vollmer *et al.*, 1984). In interpreting their results, Vollmer *et al.* (op. cit.) favoured a model involving two independent LIL element enrichments of the source region and it is encouraging that the xenocryst/xenolith data currently available can be interpreted in precisely the same way.

Experimental (Barton and Hamilton, 1979) and isotopic (Vollmer *et al.*, 1984) studies prove that madupite cannot be derived from exactly the same source as orendite and wyomingite. However, these lavas share many geochemical characteristics, including high $\text{Mg}/(\text{Mg} + \Sigma\text{Fe}^{2+})$, high Cr, low K/Rb, low $\text{P}_2\text{O}_5/\text{Ce}$, low Ti/Zr, and high Ti/V ratios; Zr/Nb ratios are approximately chondritic in madupites suggesting that these elements behave coherently in these lavas. These data imply that at least some phases (?ilmenite, phlogopite, clinopyroxene, and apatite) were present in both source regions, but the relatively low Ni contents of madupites (160–190 ppm) indicate that olivine was almost certainly not one of these phases (see also Nicholls and Whitford, 1983). It is speculated that madupites are predominantly produced from the mica-pyroxenite component of the source region and further that a reaction relation existed between these silica-undersaturated magmas and residual harzburgite. This model is consistent with the generally higher Cr contents of madupites (500–600 ppm) compared with orendites and wyomingites and with the intimate association of madupites and wyomingite/orendite at some localities (Barton, 1975; Ogden, 1979). To date, however, no xenocrysts or xenoliths have been found in madupite, so this hypothesis awaits further testing.

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