Aluminous spinels in lamproites: Occurrence and probable significance

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ABSTRACT

Aluminous spinels are a common accessory mineral in peralkaline lamproites. They may be found as inclusions in leucite (Oscar plug, Australia) or in phlogopites (Leucite Hills, Wyoming; Murcia and Almeria provinces, Spain). On the basis of a comparison with assemblages obtained from high-temperature experiments performed on biotites and an analogy with what is found in biotite-bearing xenoliths included in volcanics, the spinels are interpreted as decomposed xenocrystic micas.

INTRODUCTION

Aluminous spinels included in leucite crystals, or more frequently found within clusters of leucite crystals, have recently been described in Australian lamproites (Jaques and Foley, 1985). Their occurrence has been interpreted as being due to low-pressure exsolution of Al (as well as Fe and Mg) that had been incorporated at high P and T within the leucite structure. Similar spinels exist in other lamproites, usually in close spatial association with phlogopite, e.g., in the Leucite Hills (Kuehner et al., 1981) where the spinels were interpreted as xenocrysts and in the Spanish lamproites where they were considered as decomposition products of xenocrystic biotites (Ventu-relli et al., 1984). The various interpretations pertaining to the existence of similar Al-rich phases in rocks whose general chemical characteristic is a strong Al undersaturation prompted us to reconsider the occurrence of these spinels and their possible significance.

Aluminous spinel is an unexpected phase appearing in lavas whose Al undersaturation puts a strong imprint on the composition of all coexisting phases (Wagner and Velde, 1986). Lamproites do contain Cr-rich spinels, usually included within olivine, but they contain 50-60% Cr$_2$O$_3$ and less than 2% Al$_2$O$_3$ (Wagner and Velde, 1986). Accidental spinels, derived from disaggregated peridotite inclusions, are also found in lamproites, but these typical xenocrysts are easily distinguished from authigenic phases on textural (anhedral shape, large size, visible oxidation rim) as well as compositional grounds (Fig. 3, in Jaques and Foley, 1985). Aluminous spinels on the other hand were described initially by Lacroix (1893) and, more recently, by Maury (1976) and Grapes (1986) as high-temperature decomposition products of accidentally introduced biotites in lavas. The assemblages corresponding to these decomposition reactions, which classically include magnetite and feldspar as well as spinel, have been experimentally reproduced using natural biotite by Brearley (1986, 1987). The goal of our project is a comprehensive study of these aluminous spinels in lamproites, as well as the phases with which they appear to be associated, in order to draw an unambiguous conclusion as to their origin.

Aluminous spinels in lamproites examined in the present study occur either as small (20 μm) or large (150 μm) pale blue to pale mauve crystals always surrounded by phlogopite (Velde, 1969). Illustrations of typical occurrences are given in Figure 1. Two classical lamproite localities, southern Spain and the Leucite Hills of Wyoming, will be considered successively.

SPANISH LAMPROITES

Blue aluminous spinel, associated with phlogopite, is extremely widespread in the Spanish lamproites. The specimens considered below were collected in the Murcia (Fortuna, specimens 13A3, 13A6, 13A9; Barqueros, specimen 73A8) and Almeria (Vera, specimen 12A17) provinces. Aluminous spinels are found in glassy fortunites and verites, lavas composed of fresh and/or altered olivines, phlogopite, plus occasional pyroxenes, diopside, or orthopyroxene (Fuster et al., 1967). Three specimens from Fortuna, one from Barqueros, and one from Vera were considered. The glassy verite from Vera contains a few clinopyroxene and phlogopite crystals. The less glassy Fortuna specimens contain groundmass alkali feldspar. The Barqueros lava contains potassic richterite as a late crystallizing phase.

Lamproites contain mantle-derived inclusions and isolated xenocrysts: olivine, Cr-rich spinels, orthopyroxene, rutile, pseudobrookite, and ilmenite (Wagner and Velde, 1986). The Vera specimen also shows isolated biotite crystals usually surrounded by a phlogopitic rim: these biotites, partly decomposed, include transparent greenish Al-rich ferrous spinels (Fig. 1a) and will be described below.

The Fortuna lamproites frequently show globular associations of aluminous spinels, phlogopite, and alkali
TABLE 1. Analyses of aluminous spinels

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
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<th>10</th>
<th>11</th>
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<tbody>
<tr>
<td>Al₂O₃</td>
<td>61.02</td>
<td>63.32</td>
<td>64.09</td>
<td>60.83</td>
<td>62.00</td>
<td>62.23</td>
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<td>61.00</td>
<td>60.98</td>
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<tr>
<td>MgO</td>
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<td>16.34</td>
<td>18.22</td>
<td>15.38</td>
<td>17.77</td>
<td>18.40</td>
<td>16.22</td>
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<td>17.29</td>
<td>23.84</td>
<td>24.15</td>
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<tr>
<td>MnO</td>
<td>0.38</td>
<td>0.40</td>
<td>0.38</td>
<td>0.14</td>
<td>0.14</td>
<td>0.18</td>
<td>0.10</td>
<td>0.17</td>
<td>0.19</td>
<td>0.14</td>
<td>0.13</td>
<td>0.12</td>
<td>0.16</td>
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<tr>
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<td>0.04</td>
<td>0.06</td>
<td>0.04</td>
<td>0.14</td>
<td>0.24</td>
<td>0.44</td>
<td>0.10</td>
<td>0.19</td>
<td>0.15</td>
<td>0.47</td>
<td>0.87</td>
<td>0.07</td>
<td>0.05</td>
</tr>
<tr>
<td>Cr₂O₅</td>
<td>0.04</td>
<td>0.12</td>
<td>0.04</td>
<td>0.03</td>
<td>0.30</td>
<td>0.32</td>
<td>0.43</td>
<td>0.45</td>
<td>0.30</td>
<td>0.49</td>
<td>0.32</td>
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<td>0.11</td>
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<tr>
<td>ZnO</td>
<td>0.06</td>
<td>0.03</td>
<td>0.25</td>
<td>0.30</td>
<td>0.35</td>
<td>0.06</td>
<td>0.19</td>
<td>0.23</td>
<td>0.21</td>
<td>0.34</td>
<td>0.41</td>
<td>0.38</td>
<td>0.36</td>
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<tr>
<td>VO</td>
<td>0.10</td>
<td>0.07</td>
<td>0.20</td>
<td>0.21</td>
<td>0.23</td>
<td>0.14</td>
<td>0.12</td>
<td>0.12</td>
<td>0.33</td>
<td>0.23</td>
<td>0.04</td>
<td>0.02</td>
<td>0.04</td>
</tr>
<tr>
<td>Total</td>
<td>100.50</td>
<td>100.06</td>
<td>100.56</td>
<td>100.45</td>
<td>101.03</td>
<td>99.97</td>
<td>99.98</td>
<td>99.10</td>
<td>100.37</td>
<td>100.43</td>
<td>99.78</td>
<td>97.32</td>
<td>97.22</td>
</tr>
</tbody>
</table>

Note: Analyses in weight percent. Columns are 1—2—12A17, Vera; 3—5—73A8, Barqueros; 6—8—13A3, Fortuna; 9—11—13A9, Fortuna; 12—13—W2A1, Leucite Hills. Molecular percent was calculated from the atomic ratios of cations.

* FeO = total Fe as FeO.
** Fe²⁺ calculated assuming three cations.
† (Mn,Ni,Zn)FeO₄.

feldspar. The outside zone of these associations is exclusively composed of phlogopite, whereas the center is formed either by a cluster of the three phases or by poikilitic feldspar that includes the two other phases. Similar associations can occasionally be found in the Vera lamproites (Fig. 1d).

LEUCITE HILLS LAMPROITES

Only in the wyomingite type (Carmichael, 1967) were aluminous spinels found as tiny crystals surrounded by phlogopite. The rocks contain abundant leucite, with phlogopite and olivine phenocrysts. Priderite is a common groundmass accessory mineral, and the rock also contains diopside microphenocrysts. Glass (usually altered) may be present.

MINERALOGY

Experimental conditions

Spinel and micas were analyzed with a Camebax electron microprobe. Standards were natural minerals and metal oxides. The analysis conditions for spinels were 20 kV, 30 nA, and 20 s for major elements and 200 nA and 90 s for minor elements. Phlogopites were analyzed with 15 kV, 10 nA, and 20 s for the major elements and 40 nA and 90 s for minor elements.

Composition of aluminous spinels

The aluminous spinels found belong to the MgAl₂O₄—FeAl₂O₄ composition series, and Mg/Fe varies between 3 and 1, values that place them within the pleonaste range (Table 1). Their composition does not vary within a single specimen and, in particular, is not a function of the size of the crystals (Fig. 2). In the Spanish lamproites, FeO contents of spinels range between 16 and 20%, with a maximum of 32.55% for spinels included in the Vera biotite. In the Leucite Hills, the FeO content does not exceed 12%. Ti contents are low, usually between 0.1 and 0.5% TiO₂. Spinels contain very little Cr, commonly below 0.5% Cr₂O₅ and exceptionally up to 1%. There is no optical difference between the low- and high-Cr varieties, nor is there any apparent variation in other major ele-
Fig. 2. Composition of aluminous spinels included in phlogopite in terms of \(\frac{Al(Al + Cr + Fe^{3+})}{Mg/(Mg + Fe^{2+})}\). Data from this study: open triangles, 12A17; solid triangles, 73A8; open circles, 13A3; solid circles, 13A9; solid squares, W2A1. Insert graph: data from Jaques and Foley (1985): (a) aluminous spinels included in leucite; (b) coexisting groundmass Cr-rich spinels.

The Ni content of the aluminous spinels from the Spanish lamproites from Fortuna and those from the Leucite Hills (0.2 to 0.4%), whereas the spinels from Vera only show 0.03-0.08% NiO. The Spanish spinels are Zn-rich (0.2 to 0.6% ZnO), but only 0.12% is present in those from the Leucite Hills. V remains below the electron-microprobe detection limit in the Leucite Hills samples, whereas the Spanish lamproite spinels may contain from 0.12 to 0.40% V₂O₅.

In Figure 2, it appears that the compositional domain of these aluminous spinels in terms of \(\frac{Al(Al + Cr + Fe^{3+})}{Mg/(Mg + Fe^{2+})}\) is identical to the range shown by the aluminous spinels found in leucite in the Australian lamproites (Jaques and Foley, 1985).

### Composition of phlogopites

In all cases, compositional differences were found between phlogopites surrounding the aluminous spinels and contemporaneous, independent phlogopites found in the lava but not associated with aluminous spinels (Table 2).

<table>
<thead>
<tr>
<th>Mica Type</th>
<th>Composition of Aluminous Spinels</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phlogopite</td>
<td>1.27  0.43  0.35  0.30  0.15  0.15  0.30  0.15  0.15  0.30  0.15  0.15  0.30  0.15  0.15</td>
</tr>
<tr>
<td>Contemporaneous</td>
<td>2.27  0.54  0.32  0.21  0.10  0.10  0.21  0.10  0.10  0.21  0.10</td>
</tr>
</tbody>
</table>

#### Leucite Hills lamproite (W2A1)

Phlogopites associated with spinels are richer in Al and Fe than the independent micas: 13-15% rather than 12% Al₂O₃, and 4% FeO.

### Table 2. Analyses of micas

<table>
<thead>
<tr>
<th>Mica Type</th>
<th>Composition of Aluminous Spinels</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phlogopite</td>
<td>1.27  0.43  0.35  0.30  0.15  0.15  0.30  0.15  0.15  0.30  0.15  0.15  0.30  0.15  0.15</td>
</tr>
<tr>
<td>Contemporaneous</td>
<td>2.27  0.54  0.32  0.21  0.10  0.10  0.21  0.10  0.10  0.21  0.10</td>
</tr>
</tbody>
</table>

Note: Analyses (except for NiO) in weight percent. Columns are 1-2-12A17, Vera; 3-4-73A8, Barqueros; 5-7-13A3, 8-10-13A6, Fortuna; 11-14-13A9, Fortuna; 15-16—W2A1, Leucite Hills. FeO: total Fe as FeO. OH: calculated value. n.a.: not analyzed.

* Micas associated with aluminous spinels.
** Micas found within clusters; other analyses: authigenic micas.
rather than 2.3–2.5% FeO. The rims of the spinel-associated phlogopites, however, have a composition identical to that of the independent phlogopites. Considering minor elements, the independent micas contain more Cr and Ni than do the micas surrounding spinels. Ba contents are similar in both cases.

**Barqueros lamproite (73A8).** Little compositional difference exists between the phlogopites that surround the aluminous spinels and the other independent phlogopites from the lava. The only element indicating some difference is Al; the phlogopites around the spinels contain 13–15% Al₂O₃, whereas the others show only 11–12% Al₂O₃. Ba and Cr contents are identical in both groups of phlogopites. As in the Leucite Hills, the micas that crystallized around the spinels show lower Ni contents (0.02–0.07% instead of 0.15% NiO).

**Vera lamproite (12A17).** Compared with the authigenic micas, the phlogopites found around aluminous spinels are richer in Al (20% Al₂O₃ vs. 12–13%), Fe (8–10% FeO vs. 4–6%), and Ba (0.9–1.5% BaO vs. 0.1–0.4%), but poorer in Cr (0.05–0.32% Cr₂O₃ vs. 0.7–1.5%). Ni contents are variable (0.03–0.11% NiO) in both types of micas. They sometimes show a rim, with a skeletal structure and a composition identical to that of the groundmass micas.

**Table 2.** Analyses of partly decomposed biotite (1) and included spinel (2) from 12A17 (Vera)

<table>
<thead>
<tr>
<th>Cations per formula unit</th>
<th>1</th>
<th>2</th>
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<tbody>
<tr>
<td>SiO₂</td>
<td>35.01</td>
<td>35.01</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>18.37</td>
<td>18.37</td>
</tr>
<tr>
<td>FeO*</td>
<td>16.67</td>
<td>16.67</td>
</tr>
<tr>
<td>MgO</td>
<td>9.97</td>
<td>9.97</td>
</tr>
<tr>
<td>MnO</td>
<td>0.07</td>
<td>0.07</td>
</tr>
<tr>
<td>TiO₂</td>
<td>5.39</td>
<td>5.39</td>
</tr>
<tr>
<td>CaO</td>
<td>0.06</td>
<td>0.06</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.21</td>
<td>0.21</td>
</tr>
<tr>
<td>K₂O</td>
<td>9.54</td>
<td>9.54</td>
</tr>
<tr>
<td>BaO</td>
<td>0.69</td>
<td>0.69</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>F</td>
<td>1.10</td>
<td>1.10</td>
</tr>
<tr>
<td>Cl</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>O = F, Cl</td>
<td>0.47</td>
<td>0.47</td>
</tr>
</tbody>
</table>

Note: Analyses (except for NiO) in weight percent.

Partly decomposed biotite is rich in Al (18.37% Al₂O₃), Fe, and Ti (5.39% TiO₂) and contains no Cr (Table 3).

**Fortuna lamproites 13A6.** Micas surrounding aluminous spinels are richer in Al (16–18% Al₂O₃ vs. 12–13%) and Ba (0.6–0.7% BaO vs. 0.3%); they show lower Ti and Cr contents (2.5–2.7% TiO₂ vs. 3–4%; 0.03–0.45% Cr₂O₃ vs. 0.6–0.8%). In a small cluster of phlogopite, aluminous spinel, and feldspar, small late-crystallizing micas are Al and Ba rich (19% and 2.9%, respectively) and are also richer in Ti (4.3% TiO₂) and Fe (6.4% FeO).

**Fortuna lamproite 13A3.** Micas found around the aluminous spinels are Al rich (16–18% Al₂O₃ vs. 12–13%) and Cr poor (0.2–0.7% Cr₂O₃ vs. 0.7–1.3%) compared to the independent micas, which all show signs of instability. There is no apparent difference between the micas associated with spinels and the independent micas with respect to Fe (3–6% FeO) and Ti (2–4% TiO₂). Micas from a small spinel and K-feldspar cluster are rich in Ba (1.6–1.9% BaO vs. 0.10–0.58%) and slightly above other phlogopites with respect to Ti (4.3% TiO₂), in this respect resembling those found in similar textures in specimens 13A6 described above. The independent micas are enriched in Ni (0.12% NiO) relative to those associated with aluminous spinels.

**Fortuna lamproite 13A9.** A careful study of micas in this specimen shows great complexities. Micas associated with aluminous spinels have variable amounts of Fe and Ba, but are always richer in both elements than early-crystallizing phlogopite phenocrysts from the same specimen and are similar to late-crystallizing micas. Compared to the latter, however, micas associated with aluminous spinels all show high Cr contents (0.8–1.2% Cr₂O₃), and this character serves to distinguish them. Their high Fe and Ti contents are in contrast with the early-crystallizing phlogopites that have similar Cr contents.
Fig. 3. The distribution of Si and Al\textsuperscript{III} (Tt = total) in phlogopites: solid circles (core) and solid triangles (rim), micas associated with aluminous spinels; open triangles, authigenic micas; solid stars, micas found within clusters.

Again, the phlogopites associated with aluminous spinels are depleted in Ni (0.21 vs. 0.05-0.13% NiO).

**Summary.** Micas found around the aluminous spinels can be distinguished from the independent micas on various chemical grounds: Micas with aluminous spinels have higher Fe (Vera and the Leucite Hills), sometimes lower Si, and always higher Al contents (Fig. 3). Usually phlogopites associated with spinels have lower Cr and Ni but higher Ba contents (Fig. 4). A biotite, partly decomposed in a specimen from Vera, shows the mechanism by which aluminous spinels may be formed; in all other cases, it is impossible, save for the presence of the spinels, to texture.

**Discussion**

The aluminous spinels found in the lamproites cannot be interpreted as early precipitates from the lamproite magma because early-precipitating spinels, usually found included in olivines, are a Cr-rich and Al-poor variety. Aluminous spinels are rarely found isolated in the groundmass of some uncommon glassy lamproites (Vila et al., 1974, where the crystals were mistakenly identified as pseudobrookite). No evidence supports the hypothesis that two spinels would precipitate, one Al rich and now included in phlogopites, the other Al poor and now found in olivines. As a consequence, three possibilities remain to be explored: (1) aluminous spinels are exsolved phases (Jaques and Foley, 1985); (2) aluminous spinels are xenocrysts; (3) aluminous spinels represent products of the destabilization of some aluminous pre-existing phase.

The first hypothesis cannot be accepted: present knowledge on the mineralogy of peralkaline lavas, and potassic peralkaline lavas in particular, indicates (Wagner and Velde, 1986) that in Al-deficient liquids, peraluminous minerals are only rarely found (e.g., oligoclase crystals in an aenigmatite-bearing Hawaiian trachyte, Velde, 1978). A second argument is illustrated in Figure 1: the volume percentage of spinel found in these spinel-phlogopite associations is usually large and cannot represent simple exsolution. Last, its apparent proportion is variable from one site to the other even within a single specimen. These arguments would rule out this hypothesis.

The second possibility is unlikely. As shown by Jaques and Foley (1985, Fig. 3, reproduced as an insert in Fig. 2) xenocrystic spinels found in lamproites originated in mantle peridotites and have a different composition. In an extensive search of the literature, we have found a few spinels with compositions similar to those determined in the biotites. They were reported by Binns et al. (1970) in alkali basalts, and their origin remains unclear.
The third possibility to be considered is that the spinels are the products of a reaction between a xenocrystic phase accidently included in the magma at high temperature. We favor this explanation and think that their origin as biotite decomposition products can be proved.

**Evidence for breakdown of biotite to aluminous spinel**

Lacroix (1893, p. 575) described green-yellow spinel octahedra in altered biotites from metamorphic inclusions in volcanic rocks. Maury (1976) described the same associations within similar rocks. Yoder and Eugster (1954) in a study of phlogopite stability limits and Eugster and Wones (1962) in an investigation of biotite described the appearance of spinel as a high-temperature decomposition product of triotahedral micas. Recently, Grapes (1986) made a careful examination of the decomposition products of crustal inclusions found within West Eifel trachyte pyroclastics, among which aluminous spinel is abundant. Even more pertinent to our discussion is the comparison between the breakdown of natural biotite in a xenolith of pelitic gneiss from a dolerite, Isle of Mull (Scotland), and a kinetic experimental study by Brearley (1987). Brearley has shown that at 800°C, biotite decomposes to give a hercynite-type spinel associated with magnetite and alkali feldspar. At temperatures above 825°C, the association is similar, but alkali feldspar is replaced by glass. From Brearley’s kinetic study, it can be shown that spinel is the first phase to form. In all experimental runs reported, spinels are always more Fe-rich than the reacting biotite. Further, the effect of temperature is significant; the spinels that coexist with biotite are clearly more Mg-rich at 850°C than at 800°C, even though experiments have not reached equilibrium. The position of the point representing the spinel from the biotite found in the Vera lamproite (Fig. 5 and Table 3) near the 850°C line gives a rough estimate of the temperature at which the breakdown of the biotite probably occurred.

Because the composition of the biotite changes with the production of spinel, the composition of the biotite coexisting with the spinels does not reflect that of the original biotite. However, it should be pointed out that, unlike in Brearley’s experiments, no other Fe-Mg phases are produced in this reaction. Thus, if we assume that the aluminous spinels are derived from completely reacted mica and that the reaction was isochemical for these two elements, we may assume that the Mg/Fe ratio of the biotite is the same as that of the precursor mica. According to the Mg/Fe ratios in the spinels examined here, the original micas might have been Mg-rich phlogopites.

A final point must be considered: leucite is present around the spinels in the leucite lamproite from Oscar plug (Australia), whereas the spinels are always surrounded by biotite in the Spanish lamproites. In the Spanish examples described, the decomposition of biotite takes place in a liquid and might produce observable feldspar or feldspar may be incorporated in the liquid. Similarly, magnetite may or may not be present. The partial fusion of biotite in the liquid leads to the precipitation around the spinels of the phase with which the magma is saturated (McBirney, 1979). In the case of the Oscar plug lamproite, the magma was saturated with leucite. In all the cases examined in the present study, the magma was saturated with phlogopite.

**Conclusion**

Our conclusion is that aluminous spinels represent decomposition products of pre-existing trioctahedral micas incorporated within the lamproite magma. We have been able to show that in one case at least, aluminous spinels coexisted with a still-identifiable biotite. Since no other phases rich in Fe and Mg are produced in the breakdown of mica and assuming that the reaction is complete and isochemical, the Mg/Fe ratio of the spinel may reflect that of the original mica. If this hypothesis is correct, the micas incorporated in the lamproites studied were, for the most part, Mg-rich phlogopite, a composition compatible with a mantle origin. According to the Mg/Fe ratio of the spinels, the most magnesium phlogopites were found in the Leucite Hills magma. Such important relics, implying a nonnegligible amount of xenocrystic material, should be taken into account when trace-element budgets of lamproites are established.

**Acknowledgments**

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