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POTASSIAN ZIRCONIUM AND TITANIUM SILICATES AND STRONTIAN CERIAN PEROVSKITE IN LAMPROITES FROM THE LEUCITE HILLS, WYOMING

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ABSTRACT

The groundmass of lamproites from Middle Table Mountain, Leucite Hills, Wyoming, is characterized by the presence of potassium zirconium silicates and Sr–Ce-rich perovskite. Two intimately intergrown K–Zr silicates of differing Ti content are recognizable by cathodoluminescence techniques. Both are similar in composition to wadeite. Intergrown with these minerals is a phase having the composition of the Ti analog ($K_2TiSi_3O_9$) of wadeite ($K_2ZrSi_3O_9$) that is considered to be a new mineral. Groundmass poikilitic perovskite is unusually rich in Sr (4–7 wt.% SrO) and rare-earth elements (5–12 wt.% Ce_2O_3) relative to perovskite occurring in other lamproites and kimberlites. The formation of this unusual suite of accessory minerals in the groundmass is considered to reflect the extreme differentiation of this batch of lamproitic magma relative to other occurrences of lamproite in the Leucite Hills.

Keywords: perovskite, potassium zirconium silicate, potassium titanium silicate, wadeite, lamproite, kimberlite, Leucite Hills, Wyoming.

SOMMAIRE

La pâte des roches lamproïtiques de Middle Table Mountain, à Leucite Hills, au Wyoming, contient un assemblage de zirconosilicates de potassium et une pérovskite riche en Sr et Ce. Deux zirconosilicates de potassium en intercroissance, différant dans leur teneur en Ti, ont été décelés par cathodoluminescence. Les deux ont une composition semblable à celle de la wadéite. Leur est associée, en intercroissance, une phase qui serait l'analogue titanifère ($K_2TiSi_3O_9$) de la wadéite ($K_2ZrSi_3O_9$), et qui est considérée une nouvelle espèce minérale. Les grains poecilitiques de pérovskite dans la pâte sont anormalement enrichis en Sr (4–7% SrO, en poids) et en terres rares (5–12% en poids), en comparaison de la pérovskite typique des lamproïtes et des kimberlites. La formation de cette suite inusitée de minéraux accessoires dans la pâte résulterait d'une différenciation extrême d'une venue lamproïtique, comparé à la norme dans les lamproïtes de Leucite Hills.

(Traduit par la Rédaction)

Mots-clés: pérovskite, zirconosilicate de potassium, titanosilicate de potassium, wadéite, lamproïte, kimberlite, Leucite Hills, Wyoming.

INTRODUCTION

The Leucite Hills lamproite province has been described by Cross (1897), Ogden (1979), Carmichael (1967) and Mitchell & Bergman (1991). The majority of the lavas comprising this province are relatively unevolved leucite phlogopite and sanidine phlogopite lamproites. Recent mineralogical studies of the province by Mitchell (1989) and Mitchell & Bergman (1991) have shown that the rocks comprising Middle Table Mountain represent the most evolved lamproites of the province.

Middle Table Mountain is a small butte located 200 m to the south of North Table Mountain (Ogden 1979). The butte consists primarily of dense fine-grained columnar-jointed lamproite containing flow-aligned phenocrysts of phlogopite. The rocks were originally described as a new locality of madupite by Ogden (1979) and Ogden *et al.* (1977). Using the current nomenclature of lamproitic rocks (Scott Smith & Skinner 1984, Mitchell & Bergman 1991), the rocks are termed leucite phlogopite transitional madupitic lamproites, as they contain both phenocrystic and groundmass phlogopite of distinct composition. In the context of a detailed study of the compositional evolution of mica in the Leucite Hills lamproites, Mitchell (1989) discovered that the groundmass of the Middle Table Mountain lamproite contains the unusual assemblage of accessory minerals that are the subject of this paper.

PETROGRAPHY

The lamproite from Middle Table Mountain contains inclusion-free phlogopite phenocrysts, which are characteristically mantled by inclusion-bearing phlogopite. Phlogopite of similar character to that in the mantles also occurs as poikilitic plates in the groundmass. Microphenocrysts of leucite, diopside and apatite occur as chadacrysts in phlogopite and are enclosed in an optically unidentifiable, very fine-grained mesostasis, which may represent devitrified glass or altered sanidine.

The mesostasis contains patches, up to 200 μm in size, of colorless zirconium and titanium potassium silicates and mauve perovskite, which poikilitically enclose leucite, diopside and apatite. The potassian silicates are only readily identifiable by back-scattered electron imagery.

The trend of compositional evolution of the micas in the Leucite Hills province (Mitchell & Bergman 1991) indicates that the groundmass mica of the Middle Table Mountain lamproites is depleted in Al and enriched in Fe and Ti relative to mica phenocrysts. The groundmass is thus interpreted as representing an evolved lamproite magma. The Middle Table Mountain lamproites differ from the phlogopite madupitic lamproites occurring at Pilot Butte in that the latter contain chromite and microphenocrystic perovskite but lack phenocrystic phlogopite, poikilitic perovskite and potassian Zr- and Ti-silicates.

Table 1 provides new whole-rock compositional data for the Middle Table Mountain lamproites. The rocks are highly enriched in Sr, Ba and Zr. On the basis of their mineralogy and whole-rock composition (Table 1), the Middle Table Mountain lamproites are considered to be more evolved than those from Pilot Butte (Mitchell & Bergman 1991).

TABLE 1. COMPOSITION OF TRANSITIONAL MADUPITIC LAMPROITE FROM MIDDLE TABLE MOUNTAIN, LEUCITE HILLS, WYOMING

	1	2		1	2		1	2
SiO_2	47.8	46.4	Co	27	27	La	250	267
TiO_2	2.44	2.43	Cr	387	390	Ce	472	542
Al_2O_3	8.98	8.69	Ni	177	174	Nd	190	206
Fe_2O_3	5.50	5.42	Ga	22	20	Sm	227	23.6
MnO	0.12	0.12	Rb	203	209	Eu	4.66	5.42
MgO	9.04	8.51	Sr	2980	4680	Tb	0.99	1.11
CaO	8.32	8.96	Y	15	21	Yb	0.83	0.87
Na_2O	1.39	1.07	Zr	1430	1390	Sc	17	17
K_2O	7.44	8.06	Nb	89	92	Th	31	31
P_2O_5	1.57	1.74	Ba	7580	7930	Ta	4.6	4.5
LOI	5.47	5.85				Hf	41	42

LOI = Loss on ignition. Concentrations of major elements and Co, Cr, Ni, Ga, Rb, Sr, Y, Zr, Nb and Ba by X-ray-fluorescence spectrometry (X-Ray Assay Laboratories). Concentrations of rare-earth elements, Sc, Th, Ta and Hf by neutron activation analysis (Lakehead University) Oxides are in wt.%, trace elements in parts per million.

POTASSIUM ZIRCONIUM AND TITANIUM SILICATES

Back-scattered electron (BSE) images combined with cathodoluminescence (CL) images and energy-dispersion X-ray spectrometry (EDS) readily show the occurrence of intergrowths of zirconium and titanium potassium silicates in the groundmass of the lamproite. All phases show blue CL.

Two such intergrowths are illustrated in Figures 1 and 2. BSE images (Figs. 1a, 2a) show the presence of one phase of high average atomic number. EDS analysis shows this region to be a K–Zr silicate. Intergrown with this phase is a mineral of lower average atomic number, which is shown by EDS analysis to be a K–Ti silicate. Polychromatic CL-images of the same regions (Figs. 1b, 2b) show the presence of two distinct CL intensities within the Zr-bearing area. One part of the Zr-bearing

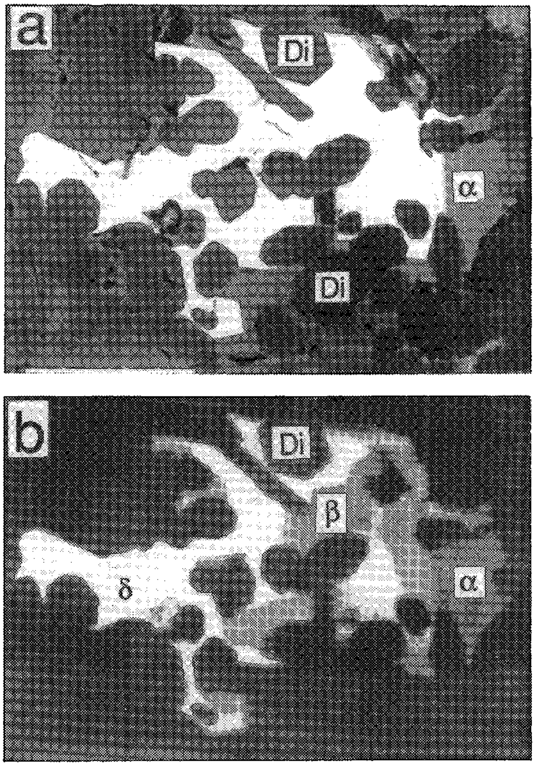

FIG. 1. Poikilitic intergrowths of potassium-bearing Zr- and Ti-silicates in the groundmass of Middle Table Mountain lamproite. (a) Back-scattered electron (BSE) image. (b) cathodoluminescence (CL) image. The area labeled α in (a) and (b) represents the potassian Ti-silicate. The K–Zr silicates cannot be distinguished in the BSE image but show two levels of CL brightness labelled β and δ. These domains probably represent two distinct minerals, one being wadeite (see text). Euhedral dark inclusions are diopside (Di), and

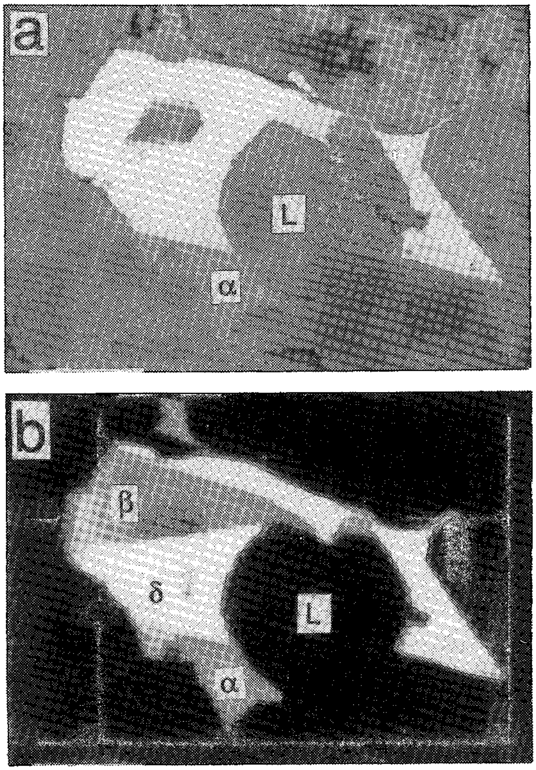

FIG. 2. (a) BSE image and (b) CL image of intergrown Zr (β and δ) and Ti (α) potassium silicates. Note the euhedral etch-pit in the dull CL Zr potassium silicate (β). L = leucite. Scale bar is 20 µm.

area exhibits a dull CL similar to that of the associated K–Ti silicate.

Wavelength-dispersion electron-microprobe analyses were obtained for the coexisting high-Zr phases using the CL intensity as a guide for the location of the points to be analyzed. In some cases, it was possible to analyze three coexisting minerals within a given poikilitic area. In other cases, only data for the Zr-bearing phase showing bright CL and the coexisting Ti-bearing phase could be obtained. Compositional data are given in Tables 2 and 3.

From Table 2, it is apparent that the K–Zr silicate showing bright CL always contains less Ti than the coexisting dull CL K–Zr silicate. The content of FeO is variable and does not correlate with CL intensity. Table 2 also shows that the stoichiometry of both Zr-bearing phases is close to $K_2(Zr,Ti)Si_3O_9$ and thus matches that of wadeite ($K_2ZrSi_3O_9$). Wadeite has previously been recognized from the Leucite Hills lamproites by Carmichael (1967).

It cannot be determined without X-ray-diffraction analysis whether or not both the dull- and bright-luminescing K–Zr silicates both consist of wadeite differing only in TiO_2 contents. However, the presence of kostylevite (monoclinic $K_2ZrSi_3O_9{\cdot}H_2O$; Khomyakov *et al.* 1983a) or umbite (orthorhombic $K_2ZrSi_3O_9{\cdot}H_2O$; Khomyakov at al. 1983b) cannot be ruled out because the electron-microprobe data are insufficiently accurate to permit the recognition by difference of the small amount of water present in these dimorphs. The small grain-sizes and the complex intergrowth of the phases preclude determination of structural water by ion microprobe or infrared absorption spectrometry. We have noted that the dull-CL phase commonly exhibits depressions having a crystallographic control on polished surfaces (Fig. 2a) and that it forms subhedral inclusions within the bright CL phase (Fig. 1b). Both of these observations suggest that it is a crystallographically distinct phase that may have formed prior to the bright-CL K–Zr silicate.

Table 3 shows that the coexisting high-Ti phase has the stoichiometry $K_2(Ti,Zr)Si_3O_9$. This does not correspond to any recognized mineral, although synthetic samples of this composition have been described (Choisnet *et al.* 1973, Shumyatskaya *et al.* 1973). The composition also is unlike that of shcherbakovite ($K_3Ti_2Si_4O_{14}$) or davenite ($K_2TiSi_6O_{12}$), minerals which also occur in lamproites. Accordingly, we suggest that this K–Ti silicate is a new mineral species, the Ti analogue of wadeite. Figure 1b shows that the mineral exhibits different levels of CL intensity. Dull-CL areas correspond to relatively Zr- and Fe-rich portions of the mineral (Table 3, columns 13 and 14). The coexistence of relatively pure examples of these two structurally similar cyclosilicates suggests that there is limited solid-solution between the Zr and Ti end-member compositions.

STRONTIAN CERIAN PEROVSKITE

Perovskite is a common poikilitic groundmass mineral (Fig. 3). This is an unusual habit, as in most alkaline igneous rocks, including the madupitic lamproite from Pilot Butte, perovskite occurs as euhedral crystals of cubo-octahedral habit.

Table 4 gives representative compositions of perovskite obtained by wavelength-dispersion electron-microprobe analysis. The determination of the concentrations of rare-earth elements (*REE*) followed closely the procedure of Exley (1980), with minor modifications of background positions following wavelength scans of the perovskite grains to be analyzed. A Cameca SX–50 electron microprobe at the University of Chicago was used for the indicated analyses and was operated at 25 kV, 100 nA, with a focused beam. The *REE* glass standards described by Drake & Weill (1972) were used, and count times were selected to obtain detection limits below 200 ppmw for the oxide.

The perovskite characteristically has high concentrations of Sr and the *REE*, and low Na and Nb contents.

TABLE 2. COMPOSITION OF COEXISTING ZIRCONIUM POTASSIUM SILICATES

Phase showing bright cathodoluminescence											
wt.%	1	2	3	4	5	6	7	8	9	10	11
SiO_2	45.6	46.5	45.5	45.4	45.5	45.1	45.7	45.7	45.1	44.8	45.5
ZrO_2	29.3	30.3	29.3	29.3	29.4	30.0	29.3	29.8	29.4	28.8	29.0
TiO_2	1.07	0.89	1.00	1.02	1.27	0.98	1.25	0.97	0.91	1.01	0.91
Al_2O_3	0.01	0.02	0.02	0.01	0.02	0.01	0.03	0.04	0.02	0.06	0.02
FeO	0.13	0.15	0.14	0.10	0.22	0.16	0.14	0.23	0.16	0.26	0.23
K_2O	23.8	22.0	23.7	23.9	23.3	23.8	23.2	23.6	23.6	23.3	23.6
Sum	99.91	99.86	99.66	99.73	99.71	100.05	99.62	100.34	99.19	98.23	99.26
Structural formulae based on 9 atoms of oxygen											
Si	3.001	3.030	3.005	2.999	2.998	2.979	3.005	2.996	2.996	2.991	3.014
Zr	0.942	0.964	0.942	0.943	0.943	0.965	0.940	0.955	0.951	0.938	0.936
Ti	0.053	0.043	0.050	0.051	0.063	0.048	0.061	0.048	0.045	0.051	0.045
Al	0.002	0.001	0.002	0.0	0.001	0.002	0 001	0.003	0.002	0.005	0.002
Fe	0.008	0.007	0.008	0.006	0.012	0.009	0.007	0.012	0.009	0.015	0.014
K	2.000	1.828	1.994	2.013	1.954	2.008	1.948	1.972	2.000	1.981	1.992
Coexisting phase showing dull cathodoluminescence											
wt.%											
SiO_2	46.0			45.5		45.5	46.6	45.2	45.6	45.8	44.3
ZrO_2	28.7			28.2		28.2	27.4	28.7	28.5	29.1	27.0
TiO_2	1.54			2.04		2.26	2.71	1.37	1.60	1.62	2.11
Al_2O_3	0.14			0.03		0.04	0.14	0.02	0.01	0.04	0.04
FeO	0.17			0.17		0.16	0.42	0.26	0.20	0.20	0.28
K_2O	23.4			22.8		22.9	22.2	23.9	23.5	23.0	23.4
Sum	99.95			98.74		99.06	99.47	99.45	99.41	99.76	97.13
Structural formulae based on 9 atoms of oxygen											
Si	3.010			3.005		2.997	3.019	2.994	3.003	3.003	2.992
Zr	0.915			0.909		0.906	0.865	0.925	0.917	0.928	0.888
Ti	0.075			0.102		0.112	0.133	0.068	0.079	0.079	0.107
Al	0.010			0.001		0.003	0.010	0.002	0.001	0.003	0.003
Fe	0.009			0.009		0.009	0.022	0.015	0.010	0.010	0.015
K	1.949			1.916		1.920	1.833	2.019	1.974	1.922	2.016

Zoning is not present. Other elements sought but not detected included Si, Dy, Er, Y, Pb, Cl and F. Perovskite compositions are plotted in Figure 4, together with those of other examples of Sr-bearing perovskite. The Leucite Hills perovskite-group mineral could be termed latrappite using the nomenclature proposed by Nickel & McAdam (1963). However, low concentrations of Nb and significant contents of REE and Sr (Fig. 4) suggest that it is best regarded as a member of a solid-solution series between perovskite ($CaTiO_3$), loparite [$(Na,Ce,Ca)(Ti,Nb)O_3$] and tausonite ($SrTiO_3$). Because of the inadequacies of the existing nomenclature and the imprecise definition of latrappite and loparite, we prefer to refer to the mineral as strontian cerian perovskite.

In order to establish the range of composition of this species in lamproites and kimberlites, grains of perovskite were also analyzed by energy-dispersion analysis. A Hitachi 570 scanning electron microscope (Lakehead University) was used with the Tracor Northern full ZAF correction routine SQ applied to results of the "standardless" analysis. Correction factors used in this program were modified by calibration with barium potassium titanate and rare-earth carbonate standards. A

TABLE 3. COMPOSITION OF POTASSIUM TITANIUM SILICATE

wt.%	2	3	4	5	12	13	14
SiO_2	51.3	50.4	49.8	50.3	50.4	51.0	51.5
ZrO_2	0.70	0.19	0.66	0.94	1.90	1.23	0.02
TiO_2	22.4	22.3	21.8	21.6	21.6	22.2	22.3
Al_2O_3	0.03	0.03	0.03	0.06	0.04	0.09	0.02
FeO	0.35	0.37	0.40	0.38	0.28	0.39	0.19
K_2O	25.1	25.9	26.3	25.8	25.8	24.9	25.9
Sum	99.88	98.10	100.02	99.08	98.99	99.81	99.93
Structural formulae based on 9 atoms of oxygen							
Si	3.010	2.995	2.985	3.002	2.985	3.000	3.024
Zr	0.019	0.006	0.020	0.027	0.055	0.036	0.0
Ti	0.991	0.998.	0.981	0.968	0.964	0.981	0.985
Al	0.003	0.001	0.003	0.004	0.003	0.006	0.001
Fe	0.016	0.018	0.020	0.019	0.013	0.019	0.009
K	1.878	1.963	2.009	1.963	1.949	1.868	1.942

Column numbers correspond with those in Table 2 and represent coexisting Zr–Ti silicates within one intergrowth.

TABLE 4. COMPOSITIONS OF STRONTIAN CERIAN PEROVSKITE

	1	2	3	4	5	6	7	8
CaO	21.3	21.4	22.5	23.3	21.1	21.9	21.5	22.1
TiO_2	51.1	50.7	50.2	50.4	50.1	50.8	51.0	51.2
Na_2O	2.94	3.01	2.94	2.89	3.01	2.89	2.89	2.86
Al_2O_3	0.02	0.01	0.02	0.02	0.02	0.02	0.01	0.2
FeO	0.57	0.49	0.49	0.41	0.72	0.40	0.42	0.53
BaO	0.28	0.28	0.32	0.33	0.26	0.33	0.32	0.28
La_2O_3	4.01	3.94	3.72	3.56	4.14	3.44	3.66	3.78
Ce_2O_3	8.60	8.41	7.62	7.50	9.26	7.41	7.64	8.12
Pr_2O_3	0.85	0.82	0.78	0.76	0.87	0.75	0.80	0.81
Nd_2O_3	2.58	2.62	2.53	2.52	2.61	2.56	2.56	2.51
Sm_2O_3	0.16	0.17	0.20	0.22	0.15	0.21	0.20	0.18
Gd_2O_3	0.05	0.03	0.05	0.07	0.03	0.05	0.05	0.04
SrO	6.21	6.48	7.02	7.19	5.53	7.05	6.96	6.45
ZrO_2	0.03	0.03	0.0	0.0	0.04	0.01	0.01	0.03
Nb_2O_5	0.81	0.93	1.09	1.19	0.69	1.20	1.13	0.90
Sum	99.51	99.32	99.48	100.36	98.53	99.02	99.15	99.81

All compositions determined by wavelength-dispersion electron-microprobe analysis. All compositions determined at the University of Chicago by Ian Steele. Oxides reported in weight %.

sample of perovskite of known composition was used to assess the accuracy of the technique (Table 5). Unfortunately, Na cannot be satisfactorily determined using this instrument by this method because of the poor counting statistics for this element. The electron-microprobe data (Table 4) show that perovskite from the Leucite Hills contains about 3 wt.% Na_2O. Consequently, the SQ compositional data (Table 5) for perovskite are corrected by this amount. This correction is not significant with respect to either the data given in Figure 5 or the conclusions drawn from it, as the correction is within the reproducibility of the analysis for SrO and Ce_2O_3. Corrections are not required for perovskite of low Na content from kimberlite. Compositions of perovskite from Middle Table Mountain determined by EDS (Table 5) are in satisfactory agreement with those determined by WDS (Table 4).

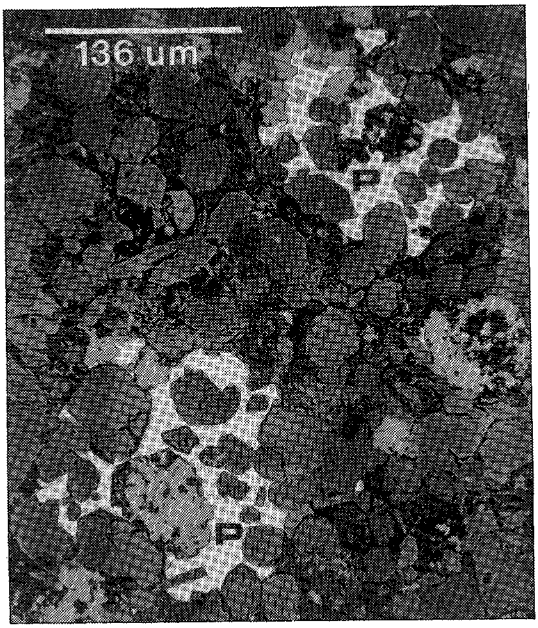

FIG. 3. Groundmass poikilitic perovskite (P). Back-scattered electron image.

Figure 5 shows that perovskite from Middle Table Mountain is richer in Sr (4 – 7 wt.% SrO) and Ce (5 – 12 wt.% Ce_2O_3) than that from Pilot Butte (2 – 4 wt.% SrO, 2.5 – 5 wt.% Ce_2O_3). This observation supports the contention that the Middle Table Mountain lamproites are more evolved than those from Pilot Butte. Figure 5 also shows that the perovskite from the Leucite Hills is characteristically richer in Sr and *REE* than perovskite from other lamproites. Perovskite from kimberlites, alnoites and perovskite pyroxenites are significantly poorer in Sr relative to perovskite in lamproite. Sr-rich perovskite is a rare mineral that has previously only been

TABLE 5. COMPOSITIONS OF STRONTIAN CERIAN PEROVSKITE FROM MIDDLE TABLE MOUNTAIN AND PILOT BUTTE LAMPROITES

	1	2	3	4	5	6	7	8
CaO	22.6	21.9	18.9	15.6	30.9	29.7	36.6	36.4
TiO_2	53.4	50.8	49.3	50.3	52.9	52.9	49.7	49.9
SrO	4.4	3.6	4.6	5.9	2.6	1.9	0.5	0.6
La_2O_3	2.5	4.1	5.9	6.1	2.3	3.3	2.8	2.4
Ce_2O_3	5.9	8.1	9.9	11.8	3.7	4.7	3.4	3.1
Pr_2O_3	n.d	1.6	1.5	0.8	n.d	n.d	n.d	0.2
Nd_2O_3	3.9	4.1	4.6	3.6	1.3	1.4	1.2	1.4
Nb_2O_5	1.6	0.9	1.2	1.0	0.6	0.5	2.1	2.0
Fe_2O_3	0.9	0.5	0.3	0.7	1.4	1.2	2.8	2.7

All compositions determined by energy-dispersion analysis. Samples 1-4 Middle Table Mountain, 5-6 Pilot Butte, 7-8 EDS and WDS analyses, respectively, of perovskite from the Walgidee Hills lamproite. n.d = not detected.

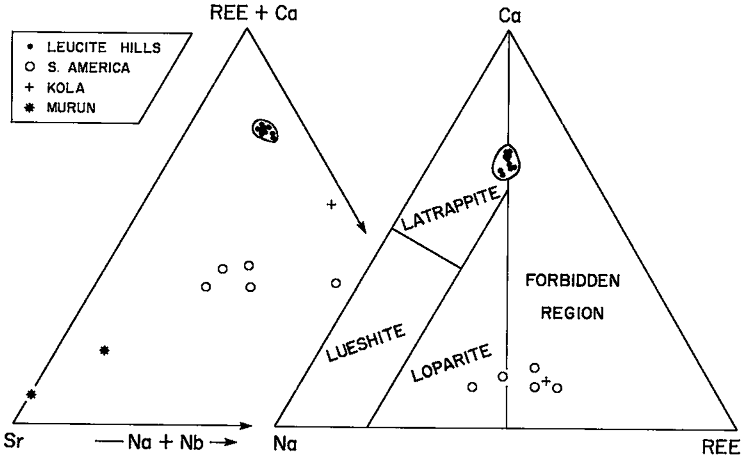

FIG. 4. Compositions of perovskite from Middle Table Mountain plotted in terms of atomic percentages in Ca–Na–*REE* and Sr–(*REE*+Ca)–(Na+Nb) plots. Data for perovskite from Sarambi, Chiriguelo and Salitre I fenites, South America, the Kola Peninsula, U.S.S.R., and Murun (U.S.S.R.) from Haggerty & Mariano (1983), Vlasov (1966) and Vorob'ev *et al.* (1984), respectively. Perovskite terminology is from Nickel & McAdam (1963).

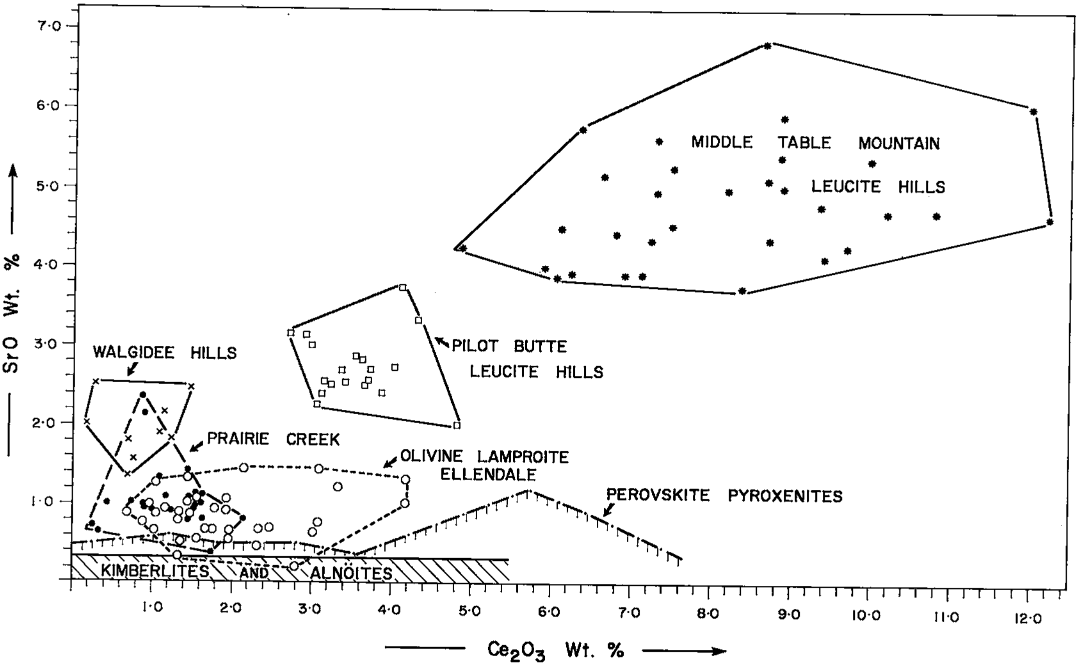

FIG. 5. Compositional variation, in terms of SrO *versus* Ce_2O_3, for perovskite from lamproite from the Leucite Hills (Middle Table Mountain, Pilot Butte), West Australia (Walgidee Hills, Ellendale) and Prairie Creek, Arkansas. These compositions contrast with those of perovskite from kimberlites, alnöites and perovskite pyroxenites. All data were obtained in this study.

described from fenites associated with carbonatite complexes (Fig. 4; Haggerty & Mariano 1983).

CONCLUSIONS

Concentration of Sr, Zr, K, Ti and Ba by fractionation leads to the formation of an unusual suite of groundmass minerals in lamproites. In the case of the Leucite Hills lamproites, the extreme fractionation of the magma has led to the formation of perovskite that is unusually enriched in Sr and Ba and to discrete potassian silicates of Zr and Ti.

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