THE ALTERATION PRODUCTS OF OLIVINE AND LEUCITE IN THE LEUCITE-LAMPROITES FROM THE WEST KIMBERLEY AREA, WESTERN AUSTRALIA

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Abstract

The leucite in the West Kimberley lamproites is largely replaced by a turbid substance. An *x*-ray, optical and chemical examination of such altered phenocrystal leucite indicates that the leucite has been replaced by an aggregate of orthoclase and an unidentified clay-like mineral. Olivine has been replaced by nontronite. The petrogenetical significance of the results is discussed.

INTRODUCTION

The leucite lamproites of the West Kimberley Area (Wade and Prider, 1940) are composed essentially of leucite associated with phlogopite, diopside, and magnophorite. Chemically their main features are a high potash content (in excess of alumina) and a silica content sufficient to have developed orthoclase, instead of leucite. Some unusual minerals from these rocks have been previously described (Prider, 1939). It was noted in this paper (p. 385) that the leucite is rarely fresh but replaced largely by a turbid clay-like mineral which, in view of the high potash content and silica saturated nature of the rocks, was thought to contain much more silica than leucite and also, in spite of its clayey appearance, still to contain considerable potash.

Insufficient material was available at that time to attempt any separation of the leucitic material, but a considerable amount of a lamproite from "P" Hill with leucite phenocrysts up to 1 mm. in diameter was obtained by one of us (R. T. P.) in 1939, and about half a gram of altered leucite was obtained by drilling the phenocrystal leucite with a dental drill. A partial chemical analysis and an optical and x-ray examination were made of this material and the results are described below. This same rock (from "P" Hill) contains a notable amount of olivine completely pseudomorphed by a fibrous, greenish mineral which was considered in the previous paper (Prider, 1939, p. 386) to be possibly nontronite. A sufficient amount of this material was obtained to enable an x-ray examination to be made. The powder representing the pseudomorphs after leucite may possibly contain a little of this greenish mineral as the altered olivines are often very close to the leucite phenocrysts, and in the same way the powder from the olivine pseudomorphs may be slightly contaminated by the leucitic alteration product.

The x-ray investigation of these materials was carried out by the pow-

der method of analysis. The technique employed and the general experimental procedure have been previously described (Shearer and Cole, 1940; Cole, 1941). In the examination of these samples it was found convenient to use Cu radiation filtered, so as to reduce the K_{β} component, by a 10μ thick nickel filter previously prepared by electro-deposition (Shearer and Cole, 1940).

The interplanar spacings were calculated directly from the Bragg equation on the basis of a constant camera radius of 2.798 cms. This was rendered possible by the fact that in this and previous work a camera of uniform radium was employed and it was shown (Cole, 1941) that the assumption of a uniform radius of 2.798 cms. for this camera yields results in which the total maximum percentage error for the strongest lines is 1.42 to 0.16 over the range 10Å to 1Å, on the assumption that (a) the radius along the film is uniform, and (b) d(2s) is 0.1 mm. in measurement of a line (where 2s is the separation of corresponding diffraction lines along the film in the equatorial plane).

In this work the materials examined contained no immediately recognizable standard substances from which calibrations of the films could be made, but it is believed that the assumption of a constant radius of 2.798 cms. yields results of the same order of accuracy as that obtained in previous investigations. As detailed below, calcite is a constituent of the leucite alteration product and the occurrence of a spotted line in the diffraction pattern of this material at 3.05Å, identified as the prominent 3.03Å calcite spacing, supports this contention.

THE ALTERATION PRODUCT OF OLIVINE

This material is greenish in colour, micaceous in habit, and completely replaces euhedral olivine (Wade and Prider, 1940, p. 67 and Fig. 7B). The optical properties of this mineral (Prider, 1939, p. 386) are: X = yellow green, Y = Z = bright green; absorption X < Y = Z. N_g=1.625, N_p= 1.595, N_g-N_p=0.030. The fibers are length slow. These characters indicate that the mineral is probably nontronite. Slices of the rock from "P" Hill were stained with malachite green without any preliminary acid treatment and the stain was strongly absorbed by this material (Fig. 1), thus supporting its determination as a clay mineral.

The x-ray powder photograph of this material gives the interplanar spacings as tabulated in Table 2, where the assigned origin of each line is also indicated. In preparing this and subsequent tables the principle was employed of assigning an origin to a line only when, on intensity considerations, the line was likely to be contributed to by the mineral in question. Data concerning the montmorillonite group of minerals were taken from Nagelschmidt (1938) and Kelley *et al.* (1939), and similar informa-

tion regarding the mica group of minerals was obtained from Nagelschmidt (1937). These publications include only data to which Miller indices related to a definite unit cell could be assigned.

Advantage was also taken of the fact that upon heating members of the montmorillonite group of clay minerals to 500° C. the characteristic 15Å basal spacing of air-dried material shrinks to 10Å. Members of the mica group are unaffected by such treatment. Heat treated material of the olivine alteration product showed this contraction and this further strengthens the conclusion arrived at from a consideration of the pattern as a whole that a clay mineral of the montmorillonite group is the predominant mineral in this sample.



Photo by H. J. Smith

FIG. 1. Photomicrograph of the lamproite from "P" Hill, West Kimberley. The field of view shows phenocrysts of altered leucite and olivine together with several smaller phenocrysts of diopside (upper left). The groundmass of the rock consists of a brownish turbid material in which are set many small leucite and rutile crystals. The section has been stained with malachite green and as a result the central turbid clayey portions of the leucite phenocrysts (lower left) appear blue. The stain has affected the nontronite pseudomorph after olivine (middle right) in such a manner that the outer margins appear purple and the darker coloured central parts green. Ordinary light, $\times 45$.

Optical evidence (as above) indicates that the mineral of the montmorillonite group is nontronite, and similar evidence suggests that the mica group mineral is phlogopite (the pseudomorphs after olivine are often rimmed with phlogopite (Wade and Prider, 1940, p. 67)). No residual olivine was seen under the microscope and in considering the origin of lines tabulated in Table 2 for the olivine alteration product, this constituent has not been considered. X-ray data yield very little evidence for the existence of any constituent other than those already considered.

The final x-ray conclusions regarding the constituents of the alteration product of the olivine are summarized in Table 4, in which all estimates of relative quantities are based on visual observation of line intensities.

THE ALTERATION PRODUCTS OF THE LEUCITE

(a) Microscopic examination and chemical analysis. The leucite in most of the lamproites from the West Kimberley area is altered, as has been noted, to a turbid clayey substance. In the rock from "P" Hill the leucite phenocrysts, in addition to the clayey alteration, are partly replaced by a weakly birefringent colourless mineral occurring in small rude spherules with their centers on the edges of the original leucite phenocrysts. This is the only rock from which a reasonably pure sample of altered leucite could be obtained by drilling the phenocrysts, and it was this material that was subjected to chemical and x-ray analysis. The usual alteration is however generally more turbid and contains very little of the colourless weakly birefringent material.

A description of the "P" Hill rock has been given previously (Wade and Prider, 1940, p. 70) and it was noted here that "the leucites are all perfectly shaped, clearer than usual and are partly replaced, especially on their outer margins, by a colourless, weakly birefringent spherulitic zeolite, with N=1.508." In the group of specimens from which the material at present under examination was obtained, the leucites appear to be similar to the description given above because the outer zones are replaced by a weakly birefringent material and the central parts by turbid clayey material (Fig. 1).

The x-ray analysis indicates the presence of feldspar and accordingly, a closer examination of the clear mineral was made. This material, quoted previously as having a refractive index of 1.508, is seen to consist of two substances: (1) a clear isotropic mineral with N=1.508, and (2) the weakly birefringent material with refractive index approximately 1.520 (orthoclase).

Staining tests were made on slices of this rock as follows:

(1) A section was stained with malachite green without any preliminary acid treatment, with the result that the central turbid parts of the leucite phenocrysts retained the stain (Fig. 1), behaving in this respect like a clay mineral.

(2) A section fumed with HCl and then stained with malachite green yielded the same result. A similar result was obtained by staining after the direct application of HCl to the rock surface. These tests indicate that the outer comparatively clear zone of the phenocrysts, which did

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not retain the stain, is composed neither of leucite nor of a zeolite.

(3) A section was fumed with HF and stained with sodium cobaltinitrite (after the method suggested by Keith, 1939, for staining potash feldspar) with the result that the clear portions retained the yellow stain, behaving in this respect like potash feldspar (in view of the absence of leucite or a potash bearing zeolite—see (2) above). It may be noted here that the groundmass of the rock also retained the yellow stain, indicative of the presence of potash.

In the sample which was drilled from the leucite phenocrysts and subsequently used for chemical and x-ray analyses, orthoclase (see above) and an isotropic clayey mineral with N = 1.508 (? cimolite) were the most abundant minerals identified. Mica (?), nontronite, diopside, and calcite were the only minor constituents recognized. Mica (?) occurs as occasional pale greenish rod-like forms scattered through the clayey mineral. These tiny rods have straight extinction, positive elongation, marked birefringence, and refractive index >1.581. Nontronite is rare and apparently has been derived from altered olivine which in places is closely associated with altered leucite. Diopside occurs as rare stout prisms. It has evidently come from inclusions in the original leucite phenocrysts or from the immediate vicinity of such phenocrysts. The more or less simultaneous crystallization of leucite and diopside in these rocks has been noted (Wade and Prider, 1940, p. 87). Calcite is in the form of tiny granules which effervesce readily in cold acid. None of this mineral could be detected in the thin slices and it seems probable than an occasional small amygdule (indistinguishable in hand specimen from a leucite phenocryst) has contributed this constituent.

SiO_2	52.63	
Al_2O_3	19.75	
Fe_2O_3	4.68	
TiO_{2}	1.54	
CaO	2.38	
MgO	3.40	
$^{1}\text{H}_{2}\text{O}+(+\text{CO}_{2})$	5.98	
H_2O-	2.70	
BaO	3.86	
${}^{2}K_{2}O(+Na_{2}O)$	3.08	
	·	
	100.00	

TABLE 1. CHEMICAL ANALYSIS OF THE ALTERATION PRODUCTS OF LEUCITE

¹ Loss on ignition of material dried at 110°C.

² By difference. It is considered, in view of the abundance of K₂O and absence of Na₂O from the analysis of a rock from "P" Hill (Skeats, 1926, p. 43), that this is entirely K₂O. *Analyst R. T. Prider.*

The chemical analysis of the alteration products of the leucite (containing the above constituents) is given in Table 1. As only 0.30 gm. of material was available, the CO_2 content could not be determined and the alkalies could only be estimated by difference.

(b) X-ray examination. The interplanar spacings of the leucite alteration products are tabulated in Table 2 (for the material both before and after heat treatment at 500° C.) together with the assigned origin of each line. In drawing up this table, data additional to that already quoted were necessary and these were obtained from Mehmel (1939), Norton (1939), and Hanawalt, Rinn and Frevel (1938). In considering the presence of calcite, reference was made to the data supplied by Nagelschmidt (1934).

From an examination of Table 2 it is apparent that the sample contains a small amount of two clay minerals, one belonging to the montmorillonite and the other to the mica group. This evidence is obtained from a consideration of the characteristic basal spacings of these two groups, viz. a 15Å spacing of montmorillonite which shrinks to 10Å on heating to 500° C., and a 10Å mica spacing which is unaffected by heat treatment.

The conclusion that potash feldspar is present, rather than leucite, is obtained from a consideration of the patterns characteristic of these two minerals. At the commencement of this work no data regarding leucite were known to the authors and a single crystal of leucite (from Albano, Italy) was therefore crushed and its powder diffraction pattern obtained. This pattern is given in Table 3 from which it can be seen that the strongest leucite lines occur at 5.33, 3.42, 3.24, 2.88, 2.80, 2.35 and 1.65Å. The strongest lines of orthoclase occur at 3.20, 2.87 and 1.778Å (Norton, 1939). The absence of a line at 5.33Å and the occurrence of only a weak line at 3.40Å in the diffraction pattern of the leucite alteration products has led to the above conclusion.

The presence of calcite is indicated by a spotted line at 3.05Å. This line does not appear after heat treatment, even though material which did not enter the container showed the presence of a small amount of carbonate when treated with HCl. Kelley *et al* (1941) have recently noted the tendency for calcite to decompose on heating to 500° C. in the presence of a clay mineral and the above observation supports this conclusion. It is noteworthy that the intensity of the lines assigned to feldspar generally increases after a heat treatment.

Chemical and microscopic evidence indicates a small amount of rutile, but as the strongest line of this material occurs at 3.24\AA (Kelley *et al* 1939) coincident with the strongest feldspar line, no conclusion, from the *x*-ray diffraction pattern, has been reached as regards its presence in the sample examined. There is very little evidence for the presence of any of the oxides or hydroxides of aluminum or iron. The significance of the possible presence of candle grease in the pattern has been referred to in a previous publication (Cole, 1941).

Alteration of Olivine		Alteration of Leucite						
Unheated				Unheated	Heated to 500°C			
Possible origin	Intensity	d/n	Possible origin	Intensity	d/n	Intensity	d/n	
Mo Mi MoMi (Mi) Mo MoMi MoMi MoMi MoMi Mo(Mi)	vs m vw s vw w vw s w d ? vw s w	$\begin{array}{c} 15.39^2\\ 9.92\\ 4.91\\ 4.46\\ 3.62\\ 3.29\\ 3.07\\ 2.57\\ 2.394\\ 2.172\\ 1.970\\ 1.671\\ 1.510\\ 1.301 \end{array}$	Mo Mo(Mi) U U U Mo(Mi) FCg FCg U F(Mi) F C MoF MoF (Mi) (F) F(Mi) U U (F)(Mi) (F) F U U (F)(Mi) (F) F U U (F) MoF(Mi) F U U (F) Mo F U U U U U U U U U U U U U U U U U U	w-m ? m m w w(d) m w vw vw vw w m m m m(d) vw ww vw vw vw vw vw vw vw vw	$\begin{array}{c} 14.87\\ 10.15\\ 8.05\\ 7.07\\ 6.30\\ 5.09\\ \\ \\ \{4.22\\ 3.97\\ 3.72\\ 3.53\\ 3.39\\ \{3.27\\ 3.05^1\\ 2.93\\ \{2.72\\ 2.52\\ \end{array}$	m m m w w w vw s s ms w s s ms w w vw vw vw vw vw vw vw vw vw vw vw vw	$\begin{array}{c} 9.92\\ 7.91\\ 7.07\\ 6.34\\ 5.09\\ \{4.50\\ \{4.24\\ \{3.96\\ 3.74\\ 3.57\\ 2.95\\ 2.72\\ 2.56\\ 2.381\\ \{2.199\\ 2.143\\ 2.088\\ 1.902\\ 1.840\\ 1.782\\ 1.745\\ 1.696\\ 1.658\\ 1.596\\ 1.578\\ \{1.522\\ 1.486\\ 1.421\\ 1.396\\ 1.287\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.170\\ 1.248\\ 1.205\\ 1.2$	

TABLE 2. INTERPLANAR SPACINGS OF THE ALTERATION PRODUCTS OF OLIVINE AND LEUCITE IN THE WEST KIMBERLEY LEUCITE LAMPROITES

vs=very strong; s=strong; ms=medium strong; m=medium; w=weak; vw=very weak; d=diffuse.

Mo=Montmorillonite; Mi=Mica; F=Feldspar; C=Calcite; Cg=Candle grease; U=Unidentified. ¹ A spotted line.

² This line shrinks to 10.0 Å after heating to 500°C.

As will be seen from Table 2 the authors have not been able to assign an origin to quite a number of lines in the pattern, in particular to those occurring at approximately 8.0, 7.07 and 6.30Å. In soil colloid work a

Leucite		Diopside				
Intensity	d/n	Intensity	d/n			
W	9.25	W	4.03			
VW	5.95	vw	3.65			
VS	5.33	W	3.28			
W	4.68	m	3.16			
VW	4.16	S	2.95			
w	3.77	w	2.89			
W	3.57	W	2.86			
VS	3.42		2.53			
VS	3.24	S	2.485			
VW	3.05	W	2.275			
S	2.88	W	2.187			
S	2,80	w(b)	2.108			
m	2.61		2.017			
VW	2.453	W	1.991			
S	2.352	VW	1.953			
VW	2.287	?	1.887			
m	2.118	VW	(1.843			
W	2.044	W	1.818			
VW	1.995	2	1.790			
W	1.905	W	1.738			
W	1.843	VW	1.658			
W	1.773	m	1.615			
m	1.714	VW	1.578			
S	1.652	vw	1.543			
W	1.615	W	1.517			
W	1.570	w	1.497			
W	1.520	vw	1.477			
m	1.469	m	1.414			
VW	1.431	W	1.402			
VW	1.401	W	1.322			
w	1.369	w	1.279			
VW	1.346	W	1.259			
W	1.314	W	1.244			
	∫1.296	vw	1.209			
W	1.285	VW	1.170			
vw	1.268	VW	1.144			
m	1.240	2	1.102			
VW	1.198	m	1.068			

TABLE 3. X-RAY DIFFRACTION PATTERNS OF LEUCITE (FROM ALBANO, ITALY) AND DIOPSIDE (FROM "P" HILL, WEST KIMBERLEY AREA, W.A.)

vs=very strong; s=strong; m=medium; w=weak; vw=very weak; b=broad.

7.0Å line normally indicates the presence of a member of the kaolinite group. However, this line is removed with the remainder of the kaolinite pattern when kaolinite bearing samples are heated to 500° C. The continued existence of the 7.07Å spacing in the leucite alteration products after heating clearly indicates the absence of kaolinite.

The only other minerals likely to be present and known to the authors to possess a 7Å spacing are the serpentines. Gruner (1937) quotes data for chrysotiles in which the strongest lines occur at 7.908, 7.118, 4.457, 3.588, 2.573, 2.421, 1.519 and 1.297Å. Most of these lines can be fitted into the pattern obtained from the leucite alteration products but the paragenesis of the serpentines and the chemical analysis seem to negative any possibility of the presence of any appreciable quantity of serpentine in this sample.

Occasional prisms of diopside were noted in the sample when examined microscopically. These do not appear to have produced any lines in the pattern. The pattern obtained from diopside separated from the "P" Hill rock is given in Table 3.

The composition of the alteration products of leucite as determined from the x-ray examination is given in Table 4.

(c) Discussion of results: In the light of the x-ray and microscopical examination of the leucite alteration products, the chemical analysis has been recast in terms of the minerals present and the result is shown in Table 5.

In this calculation it has been assumed that the K_2O and BaO are all contained in the feldspar and all the CaO is in calcite and diopside. In the absence of a knowledge of the CO₂ content, microscopic evidence suggesting that calcite is somewhat more abundant than diopside has been taken into account in determining the relative amounts of calcite and diopside.

The nontronite (= nontronite+Mg-beidellite) shown in the calculation as amounting to 21.6% is far in excess of nontronite which could be contributed by olivine and which was observed in the powder from the leucite, so it is probable that some iron and magnesia are present in the residue. The residue (which is apparently the material giving the unknown lines in the x-ray diffraction pattern) in the above calculation amounts to 40 per cent and consists essentially of silica, alumina, and an unknown amount of water (unknown in view of the fact that the water that should be assigned to nontronite and Mg-beidellite is not exactly known). This gives $Al_2O_3:SiO_2::1:3$ so that this unknown material (if it be a single mineral) may be $Al_2O_3\cdot 3SiO_2 \cdot xH_2O$ with the possibility of Mg and Fe entering its composition from the material calculated as nontronite (which as noted above is far in excess of nontronite observed under the microscope).

Petrogenetical Significance of the Investigation

It has been shown that in the pseudomorphs after leucite there are no remnants of the original leucite, but the pseudomorphs consist of a num-

TABLE 4. COMPOSITION	OF THE	ALTERATION	PRODUCTS OF	OLIVINE	AND LEUCITE IN THE
LAMPROITE	FROM "P	" HILL AS	Determined	BY X-RAY	ANALYSIS

Mineral	Olivine pseudomorphs	Leucite pseudomorphs
Montmorillonite group	Much	Little
Mica group	Little	Possible trace
Feldspar		Much
Calcite		Very little
Unknown		Much

ber of minerals including orthoclase, a montmorillonite group mineral (which may be montmorillonite derived from leucite or nontronite from olivine introduced into the powder while drilling the leucite phenocrysts,

	%	Mol. Props.	Rutile	Ortho- clase	Cel- sian	Cal- cite	Diop- side	Non- tronite	Mg- Beidel- lite	Residue
SiO ₂	52.63	877	-	198	52		20	116	100	391
Al ₂ O ₃	19.75	193	122	33	26		1	200	100	134
Fe ₂ O ₃	4.68	29	225	-	_	<u> - 200</u>		29	-	-
TiO ₂	1.54	19	19	-			-		-	-
CaO	2.38	43		-	-	33	10			
MgO	3.40	85			_		10		75	
$^{1}H_{2}O + (CO_{2} - 1.45)$	5,98	33			-	33	-		-	
$(H_2O + 4.53)$	3,90	252	-					29	25	198
$H_2O -$	2.70	-		- 200	1.000	-	-	2000	-	
BaO	3.86	26	_		26			-	-	
$K_{2}O$	3.08	33		33	2-	-	-		5 1	-
	100,00		1.54%	18.35%	8.20%	3,30%	2.16%	12.16%	9.45%	40.73%
								·		
	-		26.55% Hyalophane				21.61% Nontronite			

TABLE 5. CALCULATION OF THE MINERALS PRESENT IN THE PSEUDOMORPHS AFTER LEUCITE

 1 For the purpose of this calculation it has been assumed (see text) that the 5.98% loss on ignition consists of 1.45% CO2 and 4.53% H2O.

as indicated in the introduction, and an unknown mineral (or minerals). The orthoclase content of the pseudomorphs is probably higher than that of the powder examined as the orthoclase tends to be confined to the outer zones of the original leucite, and in boring the leucites there is a tendency to have the more central parts concentrated in the resultant powder.

The petrological interest lies in the fact that in this rock there has been

a considerable reaction of the early formed leucites with the siliceous residuum to form orthoclase, and this accounts to some extent for the silica saturated nature of these rocks and also gives further information regarding the temperature of final crystallization of these rocks which must have been at, or slightly lower than, the leucite-orthoclase reaction temperature.

The more general alteration of leucite is, however, to the turbid clayey material, which has been shown to be high in alumina and low in potash. This introduces further complications regarding the potash content of these rocks. The dominance of potash over alumina is a characteristic and most puzzling feature of the rocks of this suite (Wade and Prider, 1940, p. 74); if potash has been removed from the original leucite as indicated by this investigation, then the groundmass must contain considerably more potash than was previously thought.

The replacement of olivine by nontronite is unusual and does not appear to have been previously recorded.

The authors consider that these replacements (both of olivine and leucite) have been effected during a late magmatic stage and are not due to weathering. The facts (1) that the "clayey" alteration of leucite is so uniform throughout the suite, in otherwise perfectly fresh rocks, and (2) that the central parts of the original leucites are most affected while the outer orthoclase rim is unaltered, seem to point to late magmatic effects rather than weathering processes as being the dominant cause of the alteration.

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