

THE PROBLEM OF Na-Li SUBSTITUTION IN PRIMARY Li-AI PHOSPHATES: NEW DATA ON LACROIXITE, A RELATIVELY WIDESPREAD MINERAL

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ABSTRACT

Wet-chemical analyses and X-ray-diffraction patterns of minerals of the amblygonite-montebbrasite series occurring in the Gatumba pegmatite field, Rwanda, show that an impurity is detectable on the powder pattern if Na₂O exceeds 0.2 wt.%. In a sample from Buranga with 2.05 wt.% Na₂O, this impurity was identified as lacroixite. Electron-microprobe and wet-chemical analyses of two lacroixite samples from Rusororo and one from Buranga showed them to contain Li and OH in substitution for Na and F, respectively. The two lacroixite samples from Rusororo have the following idealized formulas: (Na_{0.61}Li_{0.39})AlPO₄(F_{0.59}OH_{0.41}) and (Na_{0.86}Li_{0.14})AlPO₄(F_{0.81}OH_{0.19}). The formula of lacroixite from Buranga is (Na_{0.78}Li_{0.22})AlPO₄(F_{0.65}OH_{0.35}). The unit-cell parameters of these three samples show that *b* decreases with increasing Li contents. The petrographic texture of the Buranga lacroixite suggests that this mineral is either a Na-metasomatic product or an exsolved phase in its montebbrasite host, whereas the decreasing Li contents of the Rusororo lacroixite samples can be tentatively related to the lowering of temperature. An X-ray-diffraction examination of the so-called "Na-rich amblygonite" from Hebron showed it to be a mixture of amblygonite and lacroixite.

Keywords: lacroixite, amblygonite-montebbrasite series, Na-Li substitution, composition, X-ray data, Gatumba pegmatite field, Rwanda.

SOMMAIRE

Des analyses chimiques par voie humide et des clichés de diffraction des rayons X, effectués sur des minéraux de la série amblygonite - montebbrasite provenant du champ pegmatitique de Gatumba, Rwanda, montrent qu'une impureté apparaît sur les diffractogrammes de poudre dès que Na₂O atteint 0.2% en poids. Grâce à un échantillon de Buranga avec 2.05% Na₂O, cette impureté a été identifiée comme étant lacroixite. Les résultats des analyses à la microsonde électronique et par voie humide montrent que les deux échantillons de lacroixite de Rusororo et celui de Buranga contiennent Li et OH, qui se substituent au Na et au F, respectivement. Les formules idéalisées des deux échantillons de Rusororo sont (Na_{0.61}Li_{0.39})AlPO₄(F_{0.59}OH_{0.41}) et (Na_{0.86}Li_{0.14})AlPO₄(F_{0.81}OH_{0.19}). Pour l'échantillon de Buranga, la formule est (Na_{0.78}Li_{0.22})AlPO₄(F_{0.65}OH_{0.35}). Le paramètre cristallographique *b* de ces trois échantillons décroît quand la teneur en Li augmente. Tandis que les teneurs décroissantes en Li des exemples de Rusororo pourraient éventuellement être

reliées à l'abaissement de la température, la texture pétrographique de la lacroixite de Buranga montre que le minéral est soit un produit de la métasomatose sodique, soit une phase d'exsolution dans les cristaux de montebbrasite. Un examen par diffraction X d'un échantillon d'"amblygonite riche en Na" de Hebron révèle un mélange d'amblygonite et de lacroixite.

Mots-clés: lacroixite, série amblygonite-montebbrasite, substitution Na-Li, composition, données de diffraction X, champ pegmatitique de Gatumba, Rwanda.

INTRODUCTION

Although amblygonite was first encountered at the Buranga pegmatite, Rwanda, in the nineteen-forties by R. de Dycker (Buttgenbach 1947), and although minerals of the LiAlPO₄(F,OH) series were extensively mined before the nineteen-sixties in several pegmatites near Buranga, in the Gatumba field, these minerals are poorly known, and the relatively few data on their mineralogical properties are dispersed in the literature (Polinard 1950, Thoreau & Bastien 1954, von Knorring 1970, Dubois *et al.* 1972, Fransolet & Abraham 1983).

The initial purpose of this study was to chemically determine the fluorine contents of minerals in the amblygonite-montebbrasite series, collected from several phosphate-rich lithium-bearing pegmatites of the Gatumba field. The interpretation of about twenty chemical analyses, complemented by a systematic X-ray investigation to check the reliability of methods proposed to determine values of the F/OH ratio (Cerná *et al.* 1973, Kallio 1978), subsequently led to the identification of lacroixite from the Rusororo pegmatite and the Buranga mine.

Recently, Lahti & Pajunen (1985) reviewed the historical background of lacroixite NaAlPO₄F, published new data for this mineral, and determined its crystal structure. The present study of three lacroixite samples from the Gatumba pegmatites provides evidence of Li and OH substitution for Na and F, respectively, and warrants further discussions of the chemical composition and crystallographic properties of lacroixite, as well as the problem of Na-for-Li substitution in the amblygonite-montebbrasite series.

ANALYTICAL METHODS

All minerals reported here were identified by X-ray diffraction, using Debye-Scherrer cameras and a diffractometer equipped with a Fe target and a monochromator. The d values of the lacroixite samples, corrected with a $\text{Pb}(\text{NO}_3)_2$ internal standard (a 7.8568 Å) and recorded with a scanning speed of $\frac{1}{2}^\circ$ 2θ /min, were used to refine the unit-cell parameters with the program of Appleman & Evans (1973) and the structural data of Lahti & Pajunen (1985). The space group $C2/c$, determined by the latter authors, was checked by the Weissenberg technique on the material from the Rusororo pegmatite.

The wet-chemical analyses were performed using atomic absorption for Al, Fe, Mn, Mg, Ca, Na, K and Li, colorimetry for P, direct titration with a specific electrode for F, and the Penfield method for H_2O . Lacroixite was dissolved in concentrated HCl heated at 190°C in a steel bomb lined with teflon.

The electron-microprobe analyses were done with CAMEBAX equipment using the ZAF correction procedure of Henoc & Tong (1977) and the following standards: synthetic berlinite (P and Al), Fe_3O_4 (Fe), metallic Mn, MgO (Mg), albite (Na), wolastonite (Ca), and topaz (F).

MINERALOGICAL DATA

Lacroixite from the Rusororo pegmatite

Near the quarry of Rusororo, briefly described by Bertossa (1960) and cited as the "Amblygonite mine of Rongi" by Varlamoff (1961, 1973), now deserted and overgrown since the end of the mining activities, some boulders of montebrasite displaying complex replacement textures are still visible. Around the cores of montebrasite is an alteration rim, up to 30 cm wide, that consists mainly of an intimate mixture of abundant berlinite and of faintly bluish lacroixite. The individual grains of lacroixite (up to 10 mm) display a vitreous to pearly luster on the cleavages (as in montebrasite). Many of the alteration rims are interrupted by irregular masses of bluish white lacroixite in close association with deep blue scorzalite, easily recognized in the field. Both lacroixite and scorzalite occur as irregular veinlets, pods, or diffuse specks. Berlinite is seldom present in this second association.

Thin sections containing the two types of association show that the bluish color of lacroixite is caused by minute inclusions of scorzalite (10 to 50 μm) that look like exsolution products. The lacroixite grains are generally turbid and are locally stuffed with other undetermined minute inclusions. In the first association, however, berlinite occurs as a mosaic, and lacroixite has a poikiloblastic-like texture and appears rather transparent at the contact with the

embedded globular grains (200 μm to 1 mm) of berlinite.

The mean of several microprobe analyses of the lacroixite (Ru-I) containing embedded berlinite is given in Table 1. On the basis of 1 $(\text{PO}_4)^{3-}$ per formula unit (p.f.u.), the main cation numbers show a distinct nonstoichiometric ratio, in contrast with the results of Lahti & Pajunen (1985). Attempts at purification did not succeed in complete separation of berlinite from lacroixite in this assemblage, and several wet-chemical analyses indeed gave an excess of Al_2O_3 and P_2O_5 . These analyses, however, showed Li and H_2O to be present in the mixtures. These results allowed me to complete the microprobe analytical data by calculating the Li_2O and H_2O contents to render stoichiometric the formula for lacroixite Ru-I, as indicated in Table 1. The simplified formula of this mineral is $(\text{Na}_{0.61}\text{Li}_{0.39})\text{AlPO}_4(\text{F}_{0.59}\text{OH}_{0.41})$.

Lacroixite Ru-II, which occurs in the scorzalite-rich veinlets of the second type of association and cross-cuts the association rich in berlinite, was purified by hand-picking under a binocular microscope. All minute inclusions of scorzalite could not be eliminated, but X-ray examination showed that impurities other than scorzalite are absent. The results of a wet-chemical analysis of lacroixite Ru-II are given in Table 2. Because Mg was not detected either in lacroixite Ru-I (Table 1) or the Buranga lacroixite (see below), the MgO content of 0.29 wt.% is assigned to scorzalite. The composition of the scorzalite inclusions was determined by microprobe analysis; the composition of lacroixite Ru-II was deduced (Table 2) to be $(\text{Na}_{0.86}\text{Li}_{0.14})\text{AlPO}_4(\text{F}_{0.81}\text{OH}_{0.19})$.

TABLE 1. MICROPROBE DATA ON LACROIXITE FROM RWANDA

	Rusororo Ru-I (n=19)		Buranga Bu. 5.21 (n=7)	
	1	2	1	2
P_2O_5	44.23	1.000	43.08	1.000
Al_2O_3	32.27	1.016	32.18	1.040
FeO	1.56	0.035	0.61	0.014
MnO	0.37	0.008	0.39	0.009
CaO	0.12	0.003	-	-
Na_2O	11.27	0.584	14.25	0.758
Li_2O^*	[3.45]	0.370	[1.99]	0.219
		1.000		1.000
H_2O^*	[2.30]	0.410	[1.90]	0.347
F	6.99	0.590	7.52	0.652
		1.000		1.000
Total	102.56		101.92	
O = F	- 2.94		- 3.16	
Total	99.62		98.76	
Na/(Na+Li)		0.612		0.776

1. Analytical results (Analyst: Prof. K. Abraham)

n = number of point analyses

* = calculated wt. % for stoichiometry

2. Cation number on the basis of 1 (PO_4) per formula unit.

TABLE 2. RESULTS OF A WET-CHEMICAL ANALYSIS OF LACROIXITE FROM RUSORORO

	1	2	3	4	
P ₂ O ₅	43.24	3.93	42.98	1.000	1.000
Al ₂ O ₃	31.62	2.82	31.49	1.020	1.020
FeO	2.99	1.47	1.66	0.038	
MnO	0.37	-	0.40	0.009	
MgO	0.20	0.29	-	-	
CaO	0.35	-	0.38	0.011	1.002
Na ₂ O	13.88	-	15.18	0.809	
K ₂ O	0.02	-	0.02	0.001	
Li ₂ O	1.11	-	1.21	0.134	
H ₂ O ⁺	1.51	0.50	1.10	0.202	
H ₂ O ⁻	0.40	-	-	-	1.036
F	8.78	-	9.60	0.834	
Total	104.56		104.02		
O = F -	3.69		4.03		
Total	100.89		99.99		

1. Analysis of lacroixite Ru-II (Analyst: J.-M. Speetjens).
2. Impurity of scorzalite calculated on the basis of the electron microprobe analysis: 42.39% P₂O₅; 31.74% Al₂O₃; 16.02% FeO; 0.13% MnO; 3.07% MgO (Analyst: Prof. K. Abraham).
3. Analysis "1" recalculated to 100%.
4. Cation numbers on the basis of 1(PO₄)³⁻ per formula unit.

This wet-chemical analysis provides an important argument to justify the addition of appropriate amounts of Li₂O and of water to complete the partial microprobe analyses of lacroixite. Moreover, if it is assumed that Li is derived from a LiAlPO₄(F,OH) mineral, then the bulk-chemical analysis (Table 2) indicates about 13 wt.% of such a mineral in the mixture. Such an amount should be easily detected by X-ray-diffraction methods, but no ambygonite was observed in the present case.

TABLE 3. X-RAY POWDER-DIFFRACTION DATA FOR LACROIXITE FROM RUSORORO

I	d _{obs.}	hkl	d _{calc.}	I	d _{obs.}	hkl	d _{calc.}
45	4.723	110	4.720	5	1.769	202	1.770
40	4.624	111	4.620	10	1.708	204	1.708
15	3.402	021	3.402	5	1.702	042	1.701
25	3.246	111	3.246	<5	1.672	331	1.671
100	3.149	112	3.149	10	1.657	114	1.656
10	3.100	002	3.101				
85	2.897	200	2.897	5	1.647	242	1.647
10	2.805	202	2.805	10	1.623	222	1.623
15	2.520	221	2.520	20	1.573	330	1.573
35	2.464	022	2.466	<5	1.550	004	1.550
				<5	1.538	333	1.540
10	2.360	220	2.360				
10	2.198	113	2.199				
25	2.152	131	2.153	<5	1.492	422	1.493
5	2.125	132	2.124			243	1.490
10	2.054	311	2.053	<5	1.470	152	1.469
				5	1.449	400	1.448
5	2.034	040	2.035	10	1.435	134	1.435
5	2.028	312	2.028				
<5	1.986	221	1.986				
10	1.927	223	1.927				
10	1.879	310	1.879				

Sample Ru-I; diffractometer; monochromatized Fe radiation; lead nitrate as internal standard; intensities estimated visually.

TABLE 4. UNIT-CELL DIMENSIONS OF LACROIXITE

	1	2	3	4
a(Å)	6.414	6.415(1)	6.420(2)	6.422(1)
b(Å)	8.207	8.190(2)	8.181(6)	8.139(2)
c(Å)	6.885	6.870(2)	6.883(5)	6.875(1)
β	115°28'	115°29'(1')	115°31'(3')	115°34'(1')
V(Å ³)	327.2	325.8(1)	326.3(2)	324.2(1)
Na/(Na+Li)	1.000	0.858	0.776	0.612
F/(F+OH)	1.000	0.805	0.653	0.590

1. Greifenstein (Lahti & Pajunen 1985). 2. Rusororo (Ru-II); 27 d values were used in the calculation. 3. Buranga (Ru.5.21); 6 d values were used (see Fig. 1). 4. Rusororo (Ru-I); 34 d values were used (see Table 3).

Although the X-ray powder-diffraction data obtained from lacroixite Ru-I are in good agreement with the results of Lahti & Pajunen (1985), a new list of indexed diffraction peaks for lacroixite rich in Li and OH seems in order because several *d* values have significantly shifted as a result of a noticeable shortening of the *b* parameter (Tables 3, 4). For example, 040 and $\bar{2}42$ occur as additional peaks, and the indexing of a few other lines has been slightly modified (e.g., indices for 2.054 and 1.657 Å).

Lacroixite from the Buranga pegmatite

A pale greyish blue sample of montebasite from the Buranga pegmatite, replaced by pure white montebasite, gave 2.05 wt.% Na₂O (Table 5), rather similar to the Na content quoted for Hebron ambygonite (Černá *et al.* 1973). However, the X-ray pattern of the Buranga sample showed 6 readily discernible diffraction-peaks attributable to lacroixite (Fig. 1). These *d* values were used to calculate the unit-cell dimensions of this lacroixite (Table 4), whose occurrence can be added to the long list of phosphate minerals known for this famous pegmatite.

In thin sections the greyish blue montebasite looks turbid or cloudy from place to place, particularly along some directions corresponding roughly to the main cleavages. Observation at high magnification clearly shows minute grains (up to 10 × 75 μm) with a lower birefringence than that of the host material, and with uniform optical orientation (Fig. 2). As is evident in Figure 3, the backscattered-electron image as well as the mapping of Na in such a "turbid zone" provide evidence for the existence of minute inclusions of a Na-rich mineral within a Na-free host.

The analytical results obtained with the electron microprobe corroborate the presence of lacroixite within montebasite (Tables 1, 5). The interpretation of the microprobe analysis of lacroixite (Table 1) again suggests the presence of additional Li and OH to maintain stoichiometry. On the basis of 1(PO₄)³⁻ p.f.u., the idealized composition for this Buranga lacroixite is (Na_{0.78}Li_{0.22})AlPO₄(F_{0.65}OH_{0.35}).

A similar heterogeneous distribution of Na within

montebrasite associated with triplite and griphite was described in the Buranga pegmatite (Fransolet & Abraham 1983). However, the presence of lacroixite could not be verified.

DISCUSSION

The new data on lacroixite from two pegmatites in the Gatumba field invite several comments and expand our knowledge of this mineral.

From infrared spectral results, Lahti & Pajunen (1985) considered that only a very limited substitution of OH for F in lacroixite may be possible. The wet-chemical and electron-microprobe analytical data in the present study indicate that Li and OH may occur in major quantities in lacroixite. In addition, marked variations of $\text{Na}/(\text{Na} + \text{Li})$ and $\text{F}/(\text{F} + \text{OH})$ have been noted (Table 1), even from one association to another in the Rusororo pegmatite. Although the available data are still too scarce, it seems that these variations affect the unit-cell parameters of lacroixite. To test this possibility further, the unit-cell dimensions given in Table 4 have been plotted against $\text{Na}/(\text{Na} + \text{Li})$ (Fig. 4). Whereas the a and c parameters seem relatively unaffected by the substitution of Li for Na, b decreases significantly as Li increases, possibly because of the smaller ionic radius of Li compared to Na. This conclusion, based on only four data-points, admittedly is tenuous, especially as F-for-OH substitution affects the unit-cell values in a similar way (Fig. 4).

The wet-chemical and the microprobe analyses both indicate a persistent presence of Fe, Mn and Ca, in contrast with the chemical data for lacroixite from Greifenstein (Lahti & Pajunen 1985). As the octahedral sites populated by Al are either filled completely, or Al is in excess (possibly because of analytical error), it is reasonable to accept that Ca, Mn and also Fe occupy the larger alkali positions. Such a hypothesis implies a coupled substitution to maintain the charge balance of the chemical formula, *i.e.*, $\text{Na}^+ + (\text{F}, \text{OH})^- \rightarrow \text{R}^{2+} + \text{O}^{2-}$. The anion involved in this coupled substitution occupies the non-tetrahedral oxygen position of the titanite structure. Therefore, the water contents calculated for stoichiometry (Table 1) are a little too high. In the case of lacroixite Ru-I, the presence of 0.046 divalent cations in the alkali positions implies 0.046 oxygen with 0.590 F and 0.364 OH p.f.u. From this OH content, one deduces 2.04 wt.% H_2O . Following the same argument, the lacroixite from Buranga contains 1.78 wt.% H_2O . The analytical error associated with a determination of H_2O by the Penfield method is relatively high, however, so that this argument cannot be corroborated by the results of the wet-chemical analysis (Table 2).

The fact that lacroixite is characterized by a titanite-type structure (Lahti & Pajunen 1985), which

is rather flexible in accepting various coupled substitutions, could explain why lacroixite contains the highest weight-percentages of FeO of any Al-rich phosphate minerals occurring in the two pegmatites (except, of course, the minerals of the scorzalite - lazulite and the childrenite - eosphorite series). Whereas 1.56% FeO was detected in lacroixite Ru-I (Table 1), the FeO contents determined previously for these Al phosphates have never exceeded 1 wt.%, *e.g.*, 0.18% as an upper limit in montebrasite, 0.27% in trolleite, 0.08% in brazilianite (wet-chemical procedure), 0.26% in augelite, and 0.85% in bertosaite (electron microprobe) (unpublished results).

From a genetic point of view, the two different values of the $\text{Na}/(\text{Na} + \text{Li})$ ratio reported for lacroixite from Rusororo are puzzling. Lacroixite Ru-I associated with berlinite replaced montebrasite, and was replaced in turn by another association charac-

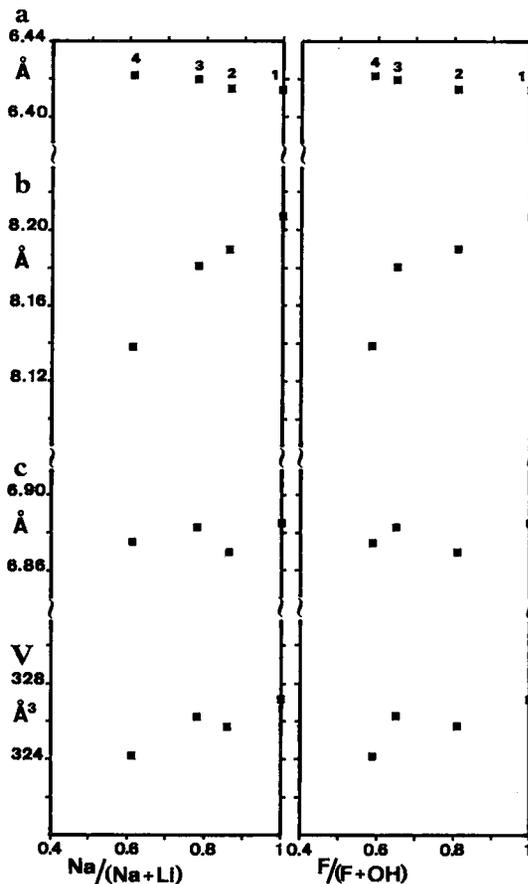


FIG. 4. Unit-cell dimensions of lacroixite plotted against the $\text{Na}/(\text{Na} + \text{Li})$ and $\text{F}/(\text{F} + \text{OH})$ ratios (1, 2, 3, 4 refer to Table 4).

terized by lacroixite Ru-II, with abundant scorzilitite and a small amount of berlinite. This observation suggests that the decreasing Li contents perhaps relate to a lowering of the temperature. However, more data are required to ascertain whether Li contents may serve as a geothermometer for lacroixite formation; interestingly, Li-free lacroixite from Greifenstein occurs in druses of a lithium-bearing granite, associated with other phosphate minerals that are typical of a low-temperature hydrothermal environment (Lahti & Pajunen 1985).

The Buranga montebrasite - lacroixite association raises another problem, critical for both the crystal chemistry and genesis of the $\text{LiAlPO}_4(\text{F},\text{OH})$ minerals. The petrographic texture exhibited by the lacroixite grains within montebrasite suggests an analogy with chessboard albite, or exsolution (Figs. 2, 3).

In the first case, lacroixite would be taken to represent a product of Na metasomatism that affected the amblygonite - montebrasite masses, as reported by Moore (1973). This evolutionary stage of Al-rich phosphate associations could be related to the same event of Na metasomatism that is known to transform the $\text{Li}(\text{Fe},\text{Mn})\text{PO}_4$ mother-phases into alluaudite in Fe-Mn phosphate associations (Moore 1971, Fransolet *et al.* 1985, 1986).

The second alternative is that lacroixite and montebrasite are the exsolution products of a pre-existing homogeneous $(\text{Li},\text{Na})\text{AlPO}_4(\text{OH},\text{F})$ phase, thereby requiring a miscibility gap in the binary Li-Na montebrasite solid-solution series. If natromontebrasite ("fremontite") described by Schaller (1911, 1914) and Na-bearing amblygonite from Hebron (Černá *et al.* 1973) are actually homogeneous, a closure of the miscibility gap must be inferred to exist at higher temperatures. All the amblygonite - montebrasite samples initially studied here, however, were found to contain lacroixite impurities if the bulk samples contain more than 0.20 wt. % Na_2O . Consequently, a sample of the Na-bearing amblygonite from Hebron was re-examined. The wet-chemical results from a fragment of this sample (U.S.N.M. 62576) accompany those obtained from the Buranga material (Table 5). The X-ray powder pattern (Fig. 1) clearly demonstrates that the mineral from Hebron contains lacroixite in amounts roughly similar to those observed in the Buranga material.

Several occurrences of lacroixite in close association with amblygonite from various localities in Czechoslovakia are known (F. Čech, written comm. 1987), and lacroixite thus may be much more widespread than was considered previously. The possibility of a miscibility gap in the Na-Li montebrasite solid solution requires much additional study. A reinvestigation of the type natromontebrasite of Schaller (1911) is in progress to answer the important question of its validity as a mineral species.

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