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NATROMONTEBRASITE, A DISCREDITED MINERAL SPECIES

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Abstract

Two samples of "natromontebrasite", among which a fragment of the type material, were re-investigated by X-ray-diffraction techniques and chemically analyzed. The X-ray powder-diffraction patterns clearly show the presence of lacroixite, whereas additional weak peaks are tentatively attributed to wardite. Electron-microprobe measurements confirm that Na is heterogeneously distributed within the samples, and essentially located in domains of lacroixite, itself scattered in a mineral of the montebrasite–amblygonite solid-solution series. "Natromontebrasite", originally described as (Na,Li)$AlPO_4$(OH,F), must thus be considered as a mixture of an OH-rich amblygonite and lacroixite, with subordinate amounts of wardite. The discreditation of "natromontebrasite" has been approved by the IMA Commission on New Minerals and Mineral Names.

Keywords: "natromontebrasite", lacroixite, amblygonite, montebrasite, Canon City, Colorado.

Sommaire

Deux échantillons de "natromontebrasite", parmi lesquels un fragment du matériau type, ont été examinés à nouveau par diffraction des rayons X et par analyses chimiques. Les diffractogrammes de poudre montrent clairement la présence de lacroixite et d'autres raies, faiblement intenses, pourraient être attribuées à la wardite. Les analyses à la microsonde électronique prouvent que Na est distribué de façon hétérogène dans ces échantillons et qu'il est essentiellement localisé dans la lacroixite, elle-même dispersée dans un minéral de la série isomorphe montebrasite – amblygonite. La "natromontebrasite", initialement décrite comme (Na,Li)$AlPO_4$(OH,F), doit donc être considérée comme un mélange d'amblygonite riche en OH et de lacroixite, avec de la wardite accessoire. La discréditation de la "natromontebrasite" a été approuvée par la Commission des nouveaux Minéraux et Noms de Minéraux de l'IMA.

Mots-clés : "natromontebrasite", lacroixite, amblygonite, montebrasite, Canon City, Colorado.

Introduction

On the basis of results of a wet-chemical analysis, Waldemar T. Schaller described a new mineral species, a Na-rich member of the montebrasite – amblygonite series, occurring in the Eight Mile Park pegmatite, near Canon City, Fremont County, Colorado. As the analyzed mineral contains 11.23 wt.% Na_2O and 3.21 wt.% Li_2O, Schaller gave it the name "*natramblygonite*" (1911a, 1912) or "natronamblygonite" (1911b). Shortly afterward, Schaller (1913) measured crystals of this new mineral species and identified their forms.

In a paper commenting on this discovery, Gonnard (1913) pointed out that the name "natromontebrasite" would be better suited if the fluorine content (5.63 wt.% F) reported by W.T. Schaller in the chemical analysis of 1911 is taken into account. Finally, Schaller (1914, 1916) admitted both the possible confusion and the objections of etymological nature with the previously published names; consequently he proposed a new name, "*fremontite*".

The aim of this note is to show that the mineral from Canon City, initially investigated by Schaller (1911a), does contain lacroixite as the main impurity heterogeneously distributed in the host mineral. "Natromontebrasite" must be thus considered as being a mixture of a montebrasite–amblygonite-series mineral and lacroixite, with subordinate amounts of wardite. The discreditation has been approved unanimously by the IMA Commission on New Minerals and Mineral Names (IMA No. 05–E).

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Review of the Literature

In comparing a sample of amblygonite from Varuträsk, Sweden, with one of "*fremontite*" from Canon City, in which he qualitatively checked for the presence of sodium, Strunz (1939) did not note any difference, either in the positions of the diffraction lines on a powder diffractogram, or in the intensities of these peaks. Therefore, he concluded that the two mineral species are isomorphous.

Heinrich (1948) described the mineral association containing "*fremontite*" from the Eight Mile Park pegmatite. A few years after, Heinrich & Corey (1955) published results of a chemical analysis of a sample of montebrasite from this locality. Devoid of fluorine and with 6.23 wt.% Na_2O, this sample is characterized by a Na:Li ratio, 1:1.7, decidedly lower than 1.7:1 deduced from the data of Schaller (1911a, b). Polinard (1950) also reported chemical data for a sample of montebrasite from the Buranga pegmatite, Rwanda, with 6.7 wt.% Na_2O.

Considering the fluctuation of the chemical data available at that time and the absence of a systematic and accurate X-ray investigation of specimens of the isomorphous series montebrasite – amblygonite, Moss *et al*. (1969) proposed a variation of the crystallographic parameters of montebrasite – amblygonite minerals as a function of fluorine content. Although no specimen of true "natromontebrasite" was considered, one sample from Canon City was included with 3.2 wt.% Na_2O. No significant effect caused by Na was detected by these authors, who reported that the samples richest in F tend to have high Na contents, however. According to Moss *et al*. (1969), such a relationship could be correlated with the geological context and not with a crystallochemical constraint.

In their detailed study on the influence of fluorine contents on some physical properties of the montebrasite – amblygonite minerals, Černá *et al*. (1973) considered, among other cases, a sample of amblygonite from the Hebron (Maine) pegmatite with 10.17 wt.% F and 2.55 wt.% Na_2O. According to the results they obtained and in spite of the ionic radius of Na, the variations of the crystallographic parameters do not seem to be influenced by the presence of sodium. The point corresponding to this sample from Hebron, "empty circle AF–55" of Černá *et al*. (1973), does not deviate significantly from the curves published in their paper.

Fransolet (1989) has shown that X-ray diffraction using the powder method allows one to reveal the presence of lacroixite, $(Na,Li)AlPO_4(F,OH)$ as a distinct phase in samples of the montebrasite – amblygonite series collected in the Li-rich pegmatites from the Gatumba area, Rwanda, especially where more than 0.5 wt.% Na_2O is detected by the wet-chemical procedure. Moreover, the presence of lacroixite within the Hebron amblygonite clearly shows that Na practically does not occur in the crystal structure of the montebrasite – amblygonite minerals (Fransolet 1989). Consequently, Na cannot influence the unit-cell dimensions of the $LiAlPO_4(OH,F)$ minerals, as can easily be seen on the diagrams of Černá *et al*. (1973). More recently, Groat *et al*. (1990) revisited the structural aspects of the series montebrasite – amblygonite and refined the different members. They noted that Na could not substitute for Li.

The Type Material

Two "natromontebrasite" samples of Canon City were investigated by X-ray-diffraction techniques and chemically analyzed.

The first specimen is registered under the number U.S.N.M. 86843 with a special label "type material" (noted herein USNM 86843) in the mineralogical collection of the National Museum for Natural History in the Smithsonian Institution. This fragment has a triangular shape (2 × 2 cm) and is about 1 cm thick. It is a part of a montebrasite – amblygonite crystal with a narrow rim of a few albite grains and white mica flakes, visible in thin sections. This same specimen was also investigated by both wet-chemical procedure and electron-microprobe analysis (EMPA).

The second is a sample fragment of the Holden Collection (#121111–2) originating from the Mineralogical Museum of the Harvard University with the number 98079 (MMHU 98079).

The macroscopic aspect of the fragments received from these two institutions is completely consistent with the original description of Schaller (1911a) and the more recent one made by Heinrich (1948). In the fragments analyzed herein, "natromontebrasite" occurs as a whitish cleavable mass embedded in albite, pink tourmaline, and pinkish mica (named lepidolite in the description of W.T. Schaller).

Results of the Re-Investigation

The X-ray powder patterns were obtained on a Philips diffractometer with a Mn-filtered Fe radiation. The d values were corrected by using $Pb(NO_3)_2$ as an internal standard. These values were used for the calculation of the unit-cell parameters of the different minerals with an updated version of the LCLSQ program (Burnham 1991).

The interpretation of the pattern of the sample USNM–86843 clearly shows the presence of lacroixite (Table 1). Indeed, a comparison made with the powder pattern published by Fransolet (1989) reveals the diffraction peak d_{200} at about 2.90 Å, typical of the presence of lacroixite in an impure specimen of montebrasite – amblygonite. The calculation of the unit-cell dimensions for the $LiAl(PO_4)(OH,F)$ member allows the identification of all the other diffraction peaks in the powder pattern (Table 1) that do not match the peaks of the $LiAlPO_4(OH,F)$ compound. As a result of an indexing in agreement with the crystal data of Lahti & Pajunen (1985) and the data published later (Fransolet 1989), seven of the lines allow us to calculate the unit-cell dimensions of lacroixite. The other peaks were tentatively attributed to wardite. The X-ray powder pattern obtained for the second sample MMHU–98079 is virtually identical to the pattern for sample USNM–86843.

We report in Table 2 the unit-cell dimensions calculated for montebrasite and associated lacroixite in the two samples. One can notice that the data are reliable. Moreover, we also report in Table 2 the F contents calculated with the regression equations deduced by Černá *et al.* (1973) from variations of the crystallographic parameters, and the unit-cell volume of members of the $LiAlPO_4(OH,F)$ series as a function of their F content given in weight %. The mean value of the F contents indirectly and semiquantitatively obtained according to this procedure, *i.e.*, 5.7 wt.% F, is consistent with our identification of F-rich montebrasite.

A wet-chemical analysis of the type material, sample USNM 86843, also gave interesting results. Although the potentiometric determination of F is relatively more difficult than a measurement of the Na or Li contents with an atomic absorption spectrometer, the new chemical data (Table 3) gave values for the fluorine content similar to the data of Schaller (1911a), and values for Na_2O and Li_2O strongly divergent from the results of that author. Compared with the chemical data of Heinrich & Corey (1955) (Table 3, column 2) and the results of a partial chemical analysis given by Moss *et al.* (1969) (Table 3, column 3), the results obtained herein tend to show that Na and Li are not homogeneously distributed within the montebrasite – amblygonite samples from Canon City (Table 3). It also becomes evident that sample USNM 86843 is impure, as the P : Al : (Na+Li) : (F+OH) proportion, equal to 1 : 1 : 0.96 : 1.22, mainly shows a (F + OH) excess likely due to an excess of H_2O. This excess, already perceptible in the chemical data of Schaller (1911a), could provide partial support for the hypothesis of the presence of wardite, detected in this sample by X-ray diffraction.

TABLE 1. X-RAY POWDER DIFFRACTOGRAM OF "NATROMONTEBRASITE" FROM CANON CITY, SAMPLE USNM-86843

This work		Montebrasite			Lacroixite			Wardite	
I/Io	d_{obs} (Å)	I/Io*	*hkl*	d_{calc} (Å)	I/Io**	*hkl*	d_{calc} (Å)	I/Io	*d* (Å)
10	4.787	8	100	4.782				50	4.772
10	4.729				22	110	**4.732**	100	4.725
40	4.670	31	110	4.671					
50	4.653	72	001	4.646					
25	4.623	20	$0\bar{1}1$	4.619	30	$11\bar{1}$	4.627		
5	3.858	9	$\bar{1}\bar{1}1$	3.859					
					16	021	3.426		
15	3.345	5	101	3.346					
25	3.321	51	$\bar{1}01$	3.319					
25	3.260	46	$\bar{1}10$	3.259	15	111	3.256		
30	3.228	62	$\bar{1}21$	3.226					
30	3.191	46	011	3.189					
85	3.159	100	$02\bar{1}$	3.163					
		28	120	3.155	100	$11\bar{2}$	3.154		
5	3.110				15	002	**3.108**	55	3.116
5	3.086	4	020	3.085				75	3.086
5	2.995							75	2.999
100	2.963	90	$1\bar{1}1$	2.961					
10	2.919	7	111	2.921					
20	2.899				81	200	**2.895**		
5	2.804				20	$20\bar{2}$	**2.803**		
10	2.558	13	210	2.557				75	2.590
					10	$22\bar{1}$	2.525		
20	2.499	24	$01\bar{2}$	2.499					
10	2.477				35	022	**2.478**		
<5	2.447	9	$1\bar{1}\bar{1}$	2.447					
25	2.391	37	$21\bar{1}$	2.391					
<5	2.373	7	$13\bar{1}$	2.374	4	220	2.366		
5	2.335	6	220	2.335					
5	2.309	8	$02\bar{2}$	2.310					
10	2.286	11	$1\bar{2}1$	2.287					
<5	2.249	6	121	2.250					
5	2.195	11	130	2.195	10	$11\bar{3}$	2.203		
5	2.166				34	131	**2.167**		
5	2.132				10	$13\bar{2}$	2.136		
15	2.118	24	$20\bar{1}$	2.119				40	2.110
5	2.097	10	102	2.096				35	2.105
5	2.054	4	$23\bar{1}$	2.054	10	$31\bar{1}$	2.051		
					2	040	2.052		
<5	2.040	4	$13\bar{2}$	2.041					
					9	$31\bar{2}$	2.025		
<5	2.002	2	$2\bar{1}0$	2.001	3	221	1.991		
15	1.949	27	012	1.950	11	041	1.949		
10	1.929	7	$2\bar{1}1$	1.929	13	$\bar{2}23$	1.930		
15	1.896	16	$1\bar{2}2$	1.896					
					8	310	1.879		
5	1.792	13	$14\bar{1}$	1.792					
					7	202	1.773		
10	1.740	19	131	1.740					
					7	042	1.713		
<5	1.708				15	$20\bar{4}$	**1.708**		
<5	1.698	4	$24\bar{1}$	1.698					

* I/I_o values were obtained with the POWDERCELL program (Krauss & Nolze 1995) using the crystallographic data of Groat *et al.* (1990), but in the cell orientation of Baur (1959), however.
** I/I_o values were obtained with the POWDERCELL program (Krauss & Nolze 1995) using the crystallographic data of Lahti & Pajunen (1985).
For lacroixite, values in a bold-faced font were used for the unit-cell calculation. The *d* values of wardite were taken from the ICDD file 33–1202.

TABLE 2. UNIT-CELL DIMENSIONS FOR MONTEBRASITE AND THE ASSOCIATED LACROIXITE FROM CANON CITY, COLORADO

	Sample USNM 86843		Sample MMHU 98079	
	Mbs (n = 32)	Lcx (n = 7)	Mbs (n = 31)	Lcx (n = 7)
a (Å)	5.170 (1)	6.409 (3)	5.170 (2)	6.415 (4)
b (Å)	7.178 (1)	8.210 (5)	7.179 (2)	8.206 (7)
c (Å)	5.049 (1)	6.881 (2)	5.049 (2)	6.883 (2)
α (°)	113.05 (02)	-	113.03 (03)	-
β (°)	98.16 (02)	115.38 (4)	98.08 (04)	115.40 (6)
γ (°)	67.65 (02)	-	67.63 (03)	-
V (Å^3)	159.45 (4)	327.12 (20)	159.40 (8)	327.29 (29)
$\bar{F}$ %	5.76 (0.35)		5.63 (0.59)	

$\bar{F}$ %: Average value for the calculated fluorine contents following the regression equations proposed by Černa *et al.* (1973) for the variations of the unit-cell dimensions as a function of F wt.%. The average excludes the estimate based on b, which has a relatively poor correlation-coefficient.
(n): Number of d values used in the LCLSQ programme of Burnham (1991).
Symbols used: Lcx: lacroixite, Mbs: montebrasite.

TABLE 3. CHEMICAL DATA ON THE "NATROMONTEBRASITE" SPECIMENS FROM CANON CITY, COLORADO

	Wet-chemical analyses						Electron-microprobe analyses							
							Amblygonite				Lacroixite			
							USNM 86843		MMHU 98079		USNM 86843		MMHU 98079	
	1a	1b	2	3	4a	4b	5a (n = 7)	5b	6a (n = 5)	6b	7a (n = 3)	7b	8a (n = 2)	8b
P_2O_5 wt%	44.35	1.000	45.87		46.40	1.000	48.96	1.000	49.16	1.000	44.10	1.000	43.63	1.000
Al_2O_3	33.59	1.054	35.77		33.61	1.008	34.82	0.990	34.66	0.982	32.94	1.040	32.45	1.035
Fe_2O_3	-	-			0.06	0.001	-	-	-	-	-	-	-	-
FeO	-	-			-	-	0.03	0.001	0.04	0.001	0.02	0.000	0.00	0.000
MnO	-	-			0.01	0.000	0.01	0.000	0.03	0.001	0.07	0.001	0.00	0.000
MgO	-	-			0.03	0.001	0.00	0.000	0.00	0.000	0.04	0.001	0.00	0.000
CaO	-	-			0.67	0.018	0.03	0.001	0.00	0.000	0.02	0.000	0.00	0.000
Na_2O	11.23	0.580	6.23	3.2	3.44	0.170	0.05	0.003	0.19	0.009	15.60	0.810	15.64	0.821
Li_2O	3.21	0.344	5.02	6.9	7.74	0.792	[10.27]	0.996	[10.25]	0.991	[1.76]	0.190	[1.64]	0.179
K_2O	0.14	0.005	n.d.		-	-	0.02	0.000	0.00	0.000	0.00	0.000	0.00	0.000
F	5.63	0.474	tr.	5.2	5.20	0.419	6.97	0.532	6.40	0.486	10.28	0.870	10.33	0.885
H_2O	4.78	0.849	7.04		4.71	0.800	[2.75]	0.443	[2.90]	0.465	[1.44]	0.258	[1.22]	0.220
	102.93		99.93		101.87		103.92		103.63		106.26		104.91	
O ≡ F	- 2.37				- 2.18		- 2.93		- 2.69		- 4.33		- 4.35	
	100.56		99.93		99.69		100.99		100.94		101.93		100.56	

Columns: 1a. Chemical data published by Schaller (1911a); 1b. Schaller's analytical data calculated on the basis of 1 PO_4. 2. Chemical data published by Heinrich & Corey (1955). 3. Analytical data reported by Moss *et al.* (1969) for the specimen "S", BM 1912,116. 4a. Chemical composition of the specimen USNM 86843. Analyst: J.-M. Speetjens (this work); 4b. Speetjens's analytical data calculated on the basis of 1 PO_4. Note that n represents the number of point-analyses; data in brackets are calculated. 5a and 7a. Representative EPMA data for OH-rich amblygonite. Analyst: Philippe de Parseval. 6a and 8a. Representative EPMA data for lacroixite. Analyst: Philippe de Parseval. 5b, 6b, 7b, and 8b. Numbers of cations are calculated on the basis of 1 PO_4 per formula unit.

In addition, further electron-microprobe measurements were performed on both the material USNM 86843 and MMHU 98079 with a Cameca SX–50 instrument (Université Paul Sabatier, Toulouse) operating in the wavelength-dispersion mode, with an accelerating voltage of 15 kV and a beam current of 10 nA. The following standards were used: graftonite from Sidi-Bou-Othmane, Morocco (P), Al_2O_3 (Al), hematite (Fe), pyrophanite (Mn), periclase (Mg), wollastonite (Ca), albite (Na), sanidine (K), and fluorapatite from Durango (F).

These measurements clearly corroborate our inference that Na is heterogeneously distributed within the sample. This is particularly conspicuous from the mapping of Na (Fig. 1A). Moreover, it must be pointed out that the heteregeneous distribution of F coincides with the distribution of Na (Fig. 1B). The electron-microprobe analyses for lacroixite were made in these Na-rich zones in order to obtain higher values for the Na and F contents.

The analytical results reported in Table 3 lead to the following comments. Firstly, the role of the minor elements, mainly Fe, Mn, Mg, Ca, and Na, is virtually insignificant in the minerals of the $LiAlPO_4(OH,F)$ series, as already pointed out by Groat *et al.* (1990). Secondly, the F/(F + OH) value, equal to 0.546 et 0.511, obtained for the member of this series in the samples USNM 86843 et MMHU 98079, respectively, proves that it is amblygonite rather rich in OH.

Conclusions

Among his comments on the name "natramblygonite" originally proposed by Schaller (1911a), Gonnard (1913) drew attention to several compositions of amblygonite and montebrasite in the literature of that time, because of their non-negligible Na_2O contents. The new investigation proves that the samples of "natromontebrasite" from the Eight Mile pegmatite, Canon City, consists of a mixture of an OH-rich amblygonite

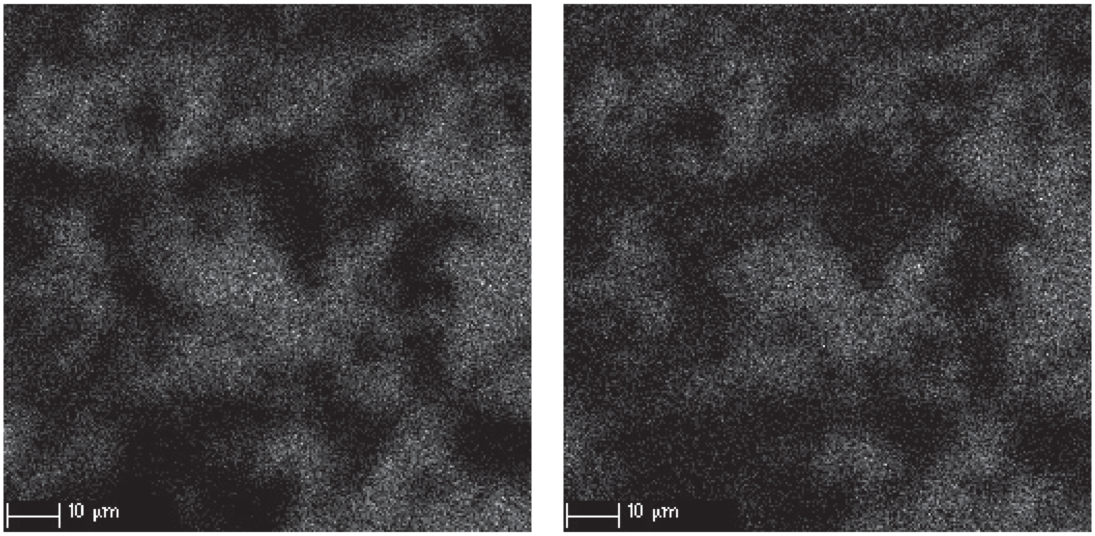

FIG. 1. Distribution of sodium (A) and fluorine (B) in a lacroixite-rich zone of the montebrasite – amblygonite sample (USNM 86843) from Canon City, Fremont County, Colorado, showing the conspicuously heterogeneous distribution of these elements.

with lacroixite and subordinate wardite. The new electron-microprobe data clearly demonstrate that Na does not enter significantly in the chemical composition of a member of the montebrasite – amblygonite series. In the case of samples USNM 86843 and MMHU 98079, Na is essentially located in lacroixite, scattered itself in OH-rich amblygonite. This study corroborates and illustrates once again one of the conclusions of Groat *et al* (1990): Na does not substitute for Li in the atomic structure of the minerals of the montebrasite – amblygonite solid-solution series.

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