RELATIONSHIP OF THE MINERALS AVELINOITE, CYRILOVITE, AND WARDITE*

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ABSTRACT

Avelinoite, the iron analogue of wardite, the isostructural aluminum compound, occurs in bright-yellow crystal aggregates in the altered zone surrounding frondelite, at the Sapucaia pegmatite mine, Minas Gerais, Brazil. Crystals are tetragonal trapezohedral (422); $a:c=1:2.650$; the forms {001}, {113}, and {012} are present. The optical constants are: uniaxial negative, $\omega=1.803$, $\epsilon=1.769$. The measured specific gravity is 3.081 ± 0.005. The formula, $NaFe_3'''(PO_4)_2(OH)_4\cdot 2H_2O[Z=4]$ is derived from the chemical analysis, in per cent: insoluble 1.04, Na_2O 4.70, K_2O 0.63, MnO 0.99, CaO 0.10, FeO none, Fe_2O_3 47.87, Al_2O_3 1.36, P_2O_5 29.06, H_2O 14.45; total 100.20. The space group for avelinoite, like that for wardite, is $P4_12_12$-$(D_4^4,)$ or $P4_32_12$-(D_4^8); the cell size of avelinoite is $a_0=7.32$, $c_0=19.4$ Å.

Cyrilovite from West Moravia studied by the writer is identical with avelinoite; the published data for chemical composition and *d*-spacings of cyrilovite are in error, and sodium is an essential constituent of the mineral.

INTRODUCTION

In December 1954 a summary description of avelinoite, a new hydrous sodium ferric phosphate mineral from the Sapucaia pegmatite mine in Minas Gerais, Brazil, appeared in *Science* (Lindberg and Pecora, 1954), where its definitive features and isostructural relation to wardite, its aluminum counterpart, were described. In April 1955, however, a reprint came to my attention of a paper by Novotný and Staněk (1953), that described cyrilovite, a new hydrous ferric phosphate mineral from the pegmatite at Cyrilov, near Velké Meziřiči, West Moravia. In June 1955, data for both avelinoite and cyrilovite were summarized in *Mineralogical Abstracts* (p. 512–513). The differences in the chemical analyses of the two minerals, hence the formulas derived therefrom, and the differences in *x*-ray powder-diffraction data† are very apparent; but similar cell dimensions, morphological elements, and optical constants led to a suspicion of the identity of the two. The manuscript on avelinoite was therefore delayed with the hope of procuring some type cyrilovite for laboratory examination. Through the courtesy of the late John P. Marble and George Switzer of the U. S. National Museum, specimens of cyrilovite (U. S. National Museum 107,425) were recently obtained. It is the writer's opinion that the new mineral named cyrilovite

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* Publication authorized by the Director, U. S. Geological Survey.

† Mineralogical Abstracts state that *x*-ray powder data for cyrilovite agree with goniometric results. The *x*-ray single-crystal and goniometric studies agree, but the single-crystal and powder data are inconsistent.

is inadequately described, that the published analysis, formula, and x-ray power-diffraction data are in error. A determination for sodium in cyrilovite by spectrographic methods shows that sodium is present in significant quantity, and, therefore, the published formula of avelinoite would apply to cyrilovite. New x-ray power-diffraction data for cyrilovite are consistent with the single-crystal data for cyrilovite and with the powder-diffraction data for avelinoite.

Avelinoite and cyrilovite are thus identical mineral species, isomorphous and isostructural with wardite, the sodium aluminum counterpart. There are no known intermediate member of the series, and no evidence of solid solution exists.

The name avelinoite was originally given to the mineral with formula $NaFe_3'''(PO)_2(OH)_4\cdot 2H_2O$ in honor of Avelino Ignacio de Oliveira, eminent Brazilian geologist and Director of the National Department of Mineral Production, Rio de Janeiro.

AVELINOITE

Crystallography

Avelinoite from the Sapucaia pegmatite mine occurs in crystal aggregates together with metastrengite and leucophosphite (Lindberg, next article) in an altered zone surrounding frondelite (Lindberg, 1949). The individual crystals vary in size and are usually smaller than 0.1 mm. Many are intergrown; many others are so minute that it is not possible to measure on the goniometer faces that are observable with a high-power microscope. Crystals are squat and when single tend to lie on the basal pinacoid (Fig. 1). Morphological data for avelinoite are given in Table 1 and are there compared with data for cyrilovite. The pinacoid {001} and the dipyramid {113} are the dominant forms; all the faces of these forms tend to be present and equally well developed. The dipyramid {012} is inconspicuous and is not always present. The crystal faces are very

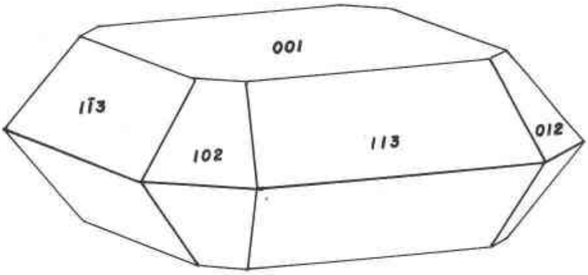

FIG. 1. Avelinoite from the Sapucaia pegmatite mine.

small and the signals observed are of poor quality; the signals observed from the form {113} are slightly better than those observed from {012}. The axial ratio, as calculated from the single-crystal x-ray measurements, $a_0:c_0=1:2.650$ is considered more accurate than that which may be derived from the measured ρ for the forms {012} and {113}; the range in measured ρ varies $3\frac{1}{2}°$ for {012} and 5° for {113}.

TABLE 1. MORPHOLOGICAL DATA FOR AVELINOITE AND CYRILOVITE
Crystal class: Tetragonal trapezohedral (422)

Avelinoite $a:c=1:2.650$; $p_0:r_0=2.650:1$						Cyrilovite[1] $a:c=1:2.650$; $p_0:r_0=2.650:1$			
Form	Number of observations	ρ measured		ϕ Calculated	ρ Calculated	Form	ρ Measured	ϕ Calculated	ρ Calculated
		Range	Mean						
001	6				0°00′	001			0°00′
012	10	52°00′–55°25′	53°33′	0°00′	52°58′	012	53°50′	0°00′	52°59′
113	18	48°20′–53°00′	50°35′	45°00′	51°20′	110	90°20′	45°00′	90°00′
						114	43°03′	45°00′	43°03′
						55·12	57°41′	45°00′	57°18′

[1] Data of Novotný and Staněk (1953). In the original measurements crystals of cyrilovite were oriented with ϕ at 45° to the orientation of the primitive cell; the forms given here have been re-oriented in the direction of the primitive cell, using the true c-axis, which is twice the length of the morphologically determined c-axis. Equivalent forms in both orientations will have equal ρ values; ϕ values will be at 45°00′.

The crystal class for both avelinoite and for wardite is tetragonal trapezohedral (422). The crystal forms observed on both avelinoite and wardite are consistent with this crystal class. Hurlbut (1952) noted that the wardite crystals from Beryl Mountain, New Hampshire, have higher symmetry than that required by the tetragonal pyramidal class (4), to which class wardite had been previously assigned.

Physical properties

Avelinoite is orange to brownish yellow. The powder is yellow.

The specific gravity, as measured on the sample used for analysis by means of an Adams-Johnston pycnometer of fused silica, is 3.081 ± 0.005. That calculated from x-ray data is 3.09 for the avelinoite end member, and 3.07 for avelinoite containing 4 per cent of the wardite component assuming the same volume for each. Since chemical analysis shows substitution not only of Al for Fe, but also of K and Mn for Na, measured and observed specific gravities are considered to be in good agreement. Avelinoite is uniaxial negative with $\omega=1.803$ and $\epsilon=1.769$. The absorption is deep yellow with $O \geqq E$. The small size of the crystals made determination of the hardness impracticable.

Chemical properties

Avelinoite is a hydrous sodium iron phosphate with a simplified formula corresponding to $NaFe_3'''(PO_4)_2(OH)_4\cdot 2H_2O$. Chemical analysis (Table 2) shows substitution of K for Na and Al for Fe. The role of manganese is uncertain. It may occur in avelinoite as a substitution for sodium, or as a surface staining of unknown nature, or as an impurity from admixed frondelite. Frondelite, $Mn''Fe_4'''(PO_4)_3(OH)_5$, occurs in the analyzed sample of avelinoite, both as very fine discrete grains and as small residual fibers in the core of avelinoite crystals. Subtraction of small quantities of frondelite from the analysis of avelinoite affects the ratios derived from avelinoite such that (1) the ratios for alkali,

TABLE 2. CHEMICAL ANALYSES OF AVELINOITE AND CYRILOVITE

	Theoretical composition	Cyrilovite[1]	Avelinoite[2]	Recalculated to 100% without insoluble	Ratios	Oxygen equivalents[3]	Metal equivalents	Metal atoms in unit cell[3]
Insoluble			1.04					
Na_2O	6.40		4.70	4.74	0.0765	0.0765	0.1529	2.97
K_2O			0.63	0.64	0.0068	0.0068	0.0136	0.26
MnO			0.99	1.00	0.0141	0.0141	0.0141	0.27
CaO			0.10	0.10	0.0018	0.0018	0.0018	0.03
FeO			None	None				
Fe_2O_3	49.43	50.89	47.87	48.28	0.3023	0.9069	0.6046	11.72
Al_2O_3			1.36	1.37	0.0134	0.0403	0.0268	0.52
P_2O_5	29.30	33.96	29.06	29.30	0.2064	1.0320	0.4128	8.00
H_2O	14.87	15.14	14.45	14.57	0.8087	0.8087	1.6174	31.38
Total	100.00	99.99	100.20			2.8871		
Sp. gr.		3.085	3.081					

[1] Analysis of Novotný and Staněk (1953). Spectrographic analysis, in per cent, U. S. National Museum 107,425, by Harry Bastron, U. S. Geological Survey, gives in addition 0.3 Mn; 1.4 Al, 0.2 Sc, 1.2 Ti, 0.04 Zr, 0.2 Mg; 0.6 Ca, 0.06 Sr, 0.004 Ba, 4 Na, X Si; 4 Na is equivalent to 5.4 per cent Na_2O.

[2] M. L. Lindberg, Analyst. Cell constants: $Na_4Fe_{12}'''(PO_4)_8(OH)_{16}\cdot 8H_2O$. Actual ratios $(Na_{2.97}K_{0.26}Mn_{0.27}Ca_{0.03})$ $(Fe_{11.72}'''Al_{0.52})(PO_4)_8(OH)_{15.38}\cdot 8H_2O$. The insoluble consists of quartz. Spectrographic analysis, in per cent, by Janet Fletcher of the U. S. Geological Survey shows in addition 0.0X Mg, Ti, Be; 0.00X Ba; 0.000X Sr.

[3] On basis of 56 oxygen atoms per cell.

water, and trivalent ions more closely approach theoretical values, (2) the ratio of phosphate ion is slightly lowered, and (3) the amount of manganese is lowered in proportion to the amount of frondelite subtracted. If all the manganese present is from frondelite, a maximum impurity of 11 per cent of frondelite would be present in the analyzed powder of avelinoite. Grain counts indicate approximately 4 per cent frondelite, plus a much smaller amount of surface staining of unknown nature.

Avelinoite is soluble in hot dilute HCl, in hot dilute H_2SO_4, and, with difficulty, in hot dilute HNO_3. In the closed tube, it gives off water and fuses.

X-ray Data

Single-crystal rotation and Weissenberg pictures about the *a*- and *c*-axes of avelinoite indicate a mineral of tetragonal symmetry with the following reflections present: (*hk*0), (*h*0*l*), and (*hkl*) all orders; (*h*00), $h=2n$; (00*l*), $l=4n$. These reflections are identical with those recorded by Larsen (1942, p. 292) for wardite, the isostructural aluminum mineral. The space group for both avelinoite and for wardite as determined from these reflections is $P4_12_12$-(D_4^4) or $P4_32_12$-(D_4^8), not $P4_1$-(C_4^2) as previously recorded. The new space group is in the tetragonal trapezohedral class (422). The symmetry of the measured crystals is consistent

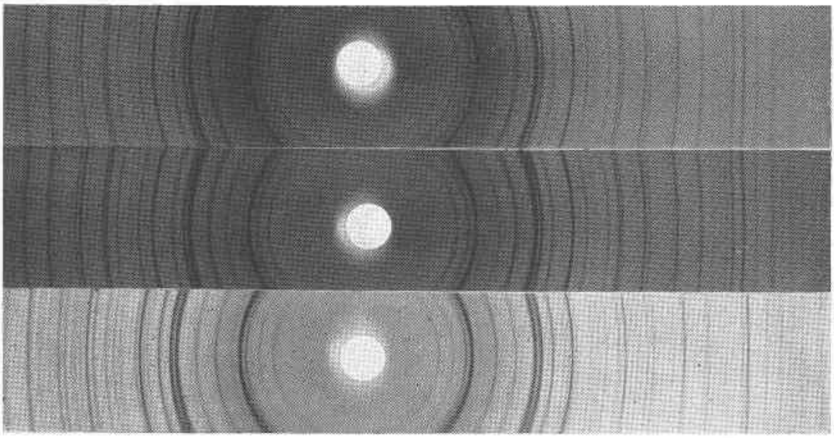

Plate 1. Powder diffraction patterns of cyrilovite, avelinoite, and wardite. (1) Cyrilovite, Cyrilov, Velké Meziříči, West Moravia; U. S. National Museum 107,425; iron radiation, manganese filter. (2) Avelinoite, Sapucaia pegmatite, Brazil; iron radiation, manganese filter. (3) Wardite, Beryl Mountain, New Hampshire; iron radiation, manganese filter; center of film offset in relation to avelinoite to emphasize difference in cell size.

TABLE 3. *X*-RAY DIFFRACTION DATA FOR AVELINOITE, CYRILOVITE, AND WARDITE

Single-Crystal Data

Avelinoite	Cyrilovite[1]	Wardite[2]
$a_0=7.32$, $c_0=19.40$ Å $a:c=1:2.650$ volume = 1040 Å^3 cell weight = 1930 cell contents: $Na_4F_{12}(PO_4)_8(OH)_{16}\cdot 8H_2O$ Sp. gr. = 3.081 ± 0.005 (measured) Sp. gr. = 3.09 (calculated)	$a_0=7.32$, $c_0=19.4$ Å $a:c=1:2.650$	$a_0=7.04$, $c_0=18.88$ Å $a:c=1:2.6818$ $Na_4Al_{12}(PO_4)_8(OH)_{16}\cdot 8H_2O$ 2.81 (meas.) 2.81 (calc.)

Powder Diffraction Data

Avelinoite[3]				Cyrilovite[4]		Cyrilovite[5]			Wardite[6]		
I	d (Å) meas.	Index	d (Å) calc.	I	d (Å) meas.	I	d (Å) meas.	sin θ	I	d (Å) meas.	Index
½	6.87	101	6.85	½	6.83				2	6.63	101
3	5.85	102	5.84	3	5.85	VW	5.50	0.1627	2	5.68	102
3	5.19	110	5.18	2	5.19	M	5.04	0.1774	3	4.99	110
10	4.85	004	4.85	10	4.85	VS	4.64	0.1927	10	4.74	004
		103	4.85								103
1	4.56	112	4.57						1	4.42	112
3	4.05	104	4.04	2	4.05	VW	3.89	0.2297	3	3.94	104
		113	4.04								113
½	3.65	200	3.66								
5	3.60	201	3.60	4	3.60	M	3.53	0.2536	2	3.47	201
½	3.31	210	3.27								
3	3.230	211	3.228	2	3.226	M	3.16	0.2829	3	3.110	211
8	3.186	203	3.186	8	3.186	M	3.02	0.2963	6	3.085	203
7	3.101	115	3.105	5	3.101	W	2.862	0.3125	2	3.025	115
		212	3.102						7	2.994	212
3	2.913	204	2.921	3	2.920	?	2.623	0.3410	4	2.825	213
		213	2.920						1	2.629	214
8	2.658	205	2.663	8	2.664	?	2.519	0.3551	7	2.591	205
3	2.502	215	2.502	3	2.505	W	2.445	0.3657	2	2.539	107
		222	2.501						1	2.388	117, 008
2	2.296	108	2.302	2	2.301	W	2.252	0.3971	2	2.256	108
3	2.210	207	2.209	3	2.207	S	2.179	0.4105	2	2.156	207
3	2.182	304	2.181	3	2.180	?	2.144	0.4171	4	2.108	
		313	2.180						1	2.063	
½	2.156	225	2.154	½	2.156				1	2.027	
½	2.116	217	2.116	½	2.116				1	2.004	
1	2.067	109	2.067	1	2.067				2	1.961	
		305	2.066						2	1.926	
4	2.020	226	2.021	4	2.020	M	1.989	0.4496	1	1.827	
		321	2.019						½	1.841	
2	1.990	315	1.988	2	1.990	?	1.962	0.4558	1	1.815	
½	1.943	306	1.948	½	1.943				4	1.764	
½	1.875	10.10	1.876	½	1.871				½	1.674	
1	1.854	209	1.857	½	1.854				½	1.666	
4	1.833	307	1.832	3	1.833	M	1.807	.04949	1	1.656	
		400	1.830						1	1.639	
3	1.714			2	1.718	M	1.694	0.5280	1	1.590	
½	1.668			½	1.664				½	1.573	
½	1.658			½	1.654	?	1.646	0.5434	½	1.559	

TABLE 3—(*continued*)

Powder Diffraction Data

Avelinoite[3]				Cyrilovite[4]		Cyrilovite[5]			Wardite[6]		
I	*d* (Å) meas.	Index	*d* (Å) calc.	I	*d* (Å) meas.	I	*d* (Å) meas.	sin θ	I	*d* (Å) meas.	Index
1	1.638			½	1.634				2	1.534	
1	1.615			1	1.615	?	1.601	0.5587	5	1.516	
3	1.589			3	1.589	W	1.570	0.5826	2	1.460	
5	1.553			4	1.553	VS	1.535	0.5980	½	1.426	
2	1.510			2	1.510	W	1.496	0.6182	½	1.422	
2	1.462			2	1.462	W	1.447	0.6324	½	1.399	
1	1.450			½	1.450				1	1.392	
1	1.434			½	1.432	W	1.414	0.6324	½	1.386	
1	1.411			½	1.411	W	1.395	0.6413	½	1.381	
½	1.403			½	1.403				2	1.368	
1	1.380			½	1.380				1	1.356	
2	1.357			1	1.358				2	1.333	
2	1.349			1	1.349	M	1.337	0.6690	1	1.319	
½	1.332								1	1.300	
½	1.306								½	1.284	
3	1.296			2	1.296	M	1.284	0.6965	½	1.272	
½	1.284								½	1.268	
½	1.271								½	1.255	
3	1.254			2	1.253	M	1.240	0.7211	½	1.250	
1	1.234								½	1.216	
1	1.218			½	1.218	W	1.209	0.7398	1	1.197	
½	1.211								1	1.175	
½	1.202			½	1.202	W	1.191	0.7509	1	1.142	
½	1.196			½	1.195	VW	1.180	0.7579	½	1.087	
1	1.184			½	1.184	?	1.159	0.7718	½	1.081	
½	1.165			½	1.164	VW	1.147	0.7800	½	1.074	
½	1.152			½	1.151	VW	1.138	0.7860	½	1.055	
½	1.150								½	1.051	
1	1.142			1	1.142	?	1.133	0.7898	½	1.031	
½	1.127			½	1.127	?	1.128	0.7929	1	1.017	
½	1.103			½	1.102	W	1.019	0.8774	1	1.004	
½	1.087								½	0.9965	
½	1.056								2	0.9897	
½	1.044										
½	1.034										
½	1.021										
2	1.013			1	1.014	M	1.009	0.8861			
1	1.004					?	.9993	0.8951			
3	0.9940			2	.9938	S	.9908	0.9028			
						M	.9803	0.9124			
2	0.9843			1	.9822	M	.9657	0.9262			

[1] Cyrilovite, Cyrilov, Velké Mežiřiči, West Morovia. Data of Němec (Novotný and Staněk, 1953).

[2] Wardite, Beryl Mountain, N. H. Data of Hurlbut (1952).

[3] Avelinoite, Sapuacia Pegmatite. Iron radiation, manganese filter Debye-Sherrer camera diameter 114.59 mm.

[4] Cyrilovite, U.S.N.M. 107,425; Cyrilov, Velké Mežiřiči, West Moravia. Iron radiation, manganese filter Debye-Sherrer camera diameter 114.59 mm. Data of Lindberg.

[5] Cyrilovite (same locality); values of sin θ from Table 2 of type description (Novotný and Staněk, 1953) transformed to *d*-spacings by Lindberg, using Kα = 1.7889, cobalt radiation. Sin θ = 0.2963 was substituted for sin θ = 0.2987 to conform with θ = 17°14′.

[6] Wardite, Beryl Mtn., N. H., Harvard No. 103,379. Iron radiation, manganese filter Debye-Sherrer camera diameter 114.59 mm.

with this class. Donnay (1954, p. 380) corrected the space group of wardite to be consistent with reflections listed by Larsen, but did not change the crystal class C_4^2 (p. 40) to D_4^4 or D_4^8 (p. 46), as noted by Heritsch (1955, p. 249).

The cell dimensions and powder diffraction data for avelinoite, cyrilovite, and wardite are given in Table 3. The indexing of planes of high *d*-spacing in avelinoite is on the basis of reflections also observed in the single-crystal studies; planes with low *d*-spacing have not been indexed because of the large number of possible choices. Calculated *d*-spacings for comparable reflections in wardite will be given in a later paper by Lindberg describing the relationship of millisite to the wardite-avelinoite (cyrilovite) series. The isostructural relationship and differences in cell size of avelinoite and of wardite are illustrated in Plate 1, where their powder-diffraction patterns are compared.

CYRILOVITE

In 1953 Novotný and Staněk published a description of a new mineral, cyrilovite. This mineral now appears, by re-examination by the present author of type material (U. S. National Museum 107,425), to be the same species as avelinoite although the original data of cyrilovite were only in partial agreement with the data for avelinoite. The chemical composition, formula, and *x*-ray powder-diffraction data for cyrilovite cited in the original paper are inconsistent with data for avelinoite and the identity of the two minerals cannot be established from published data alone.

Crystals of both cyrilovite and avelinoite are tetragonal; the direction for the *a*-axis selected by Novtný and Staněk (1953) is at a 45° angle to the smallest primitive unit cell for cyrilovite and also at 45° to the direction selected by Lindberg for avelinoite. After adjustment for this difference in orientation and doubling of the *c*-axis, the $a{:}c$ ratio of both minerals is the same. The forms given by Novotný and Staněk for cyrilovite become in the primitive lattice $\{001\}$ the same, $\{010\}=\{110\}$, $\{111\}=\{012\}$, $\{011\}=\{114\}$, $\{053\}=\{55.12\}$.

The optical properties for cyrilovite are $\omega=1.805$, $\epsilon=1.777$ (Novotný and Staněk, 1953). The measured specific gravity of cyrilovite is 3.085 (Novotný and Staněk) compared with 3.081 ± 0.005 for avelinoite.

The chemical analysis of cyrilovite, as determined upon a sample weighing 0.018 gram (Novotný and Staněk, 1953), is recorded in Table 2 where it may be compared with the analysis of avelinoite. Sodium was recorded in spectrographic traces only. Harry Bastron of the U. S. Geological Survey re-examined cyrilovite (U. S. National Museum 107,425) spectrographically and reports approximately 4 per cent Na which is equivalent to 5 per cent Na_2O, indicating that sodium is an essential component in cyrilovite; the published analysis of the other components, which total 99.99 per cent without considering the sodium content, is in error. The published analysis of cyrilovite cannot therefore be used to determine the correct formula for cyrilovite.

The cell dimensions of cyrilovite obtained from single-crystal studies as determined by Němec (Novotný and Staněk, 1953, p. 334) agree exactly with those of avelinoite; cyrilovite, tetragonal, $c_0 = 19.4$, $a_0 = 7.32$ Å; avelinoite, tetragonal, $c_0 = 19.4$, $a_0 = 7.32$ Å; however, the *x*-ray powder-diffraction data for cyrilovite, obtained by Černohorský (Novotný and Staněk, 1953, p. 329) are not in agreement with the data for avelinoite. A comparison of the powder diffraction data for the two minerals (Table 3) shows:

1. The powder pattern of avelinoite may be indexed using the values of a_0 and c_0 derived from single-crystal data.
2. The powder pattern of cyrilovite (original data) cannot be indexed by using the values of a_0 and c_0 derived from single crystal data.
3. To the extent that the intensities of reflections are indicated, the succession of strong and weak lines for cyrilovite is not the same as for avelinoite, and entirely different reflections would be used in cataloging for identification.

In order to investigate the discrepancies in the powder data for cyrilovite, as compared with avelinoite and with the single crystal data, an *x*-ray powder-diffraction pattern of type material (U. S. National Museum 107,425) was taken (pl. 1). The *d*-spacings and their relative intensities so obtained (Table 3) are consistent with those of avelinoite and indicate that there is a fundamental error in the published data for cyrilovite. The *d*-spacings obtained by Černohorský (data, Table 3, transformed from sin θ by the present author) are too small for the cell size and indicate a cell size smaller than that of wardite, the isostructural aluminum compound. That significant differences in cell size for avelinoite and wardite exist is illustrated in Plate 1; reflections aligned on the right of center are considerably offset to the left of center.

CONCLUSIONS

Avelinoite and cyrilovite are believed to be the same mineral species. The published description of cyrilovite only suggests the possibility of identity of the two minerals. Examination of type material of cyrilovite established the presence of sodium in cyrilovite in quantity sufficient to enter the formula. The published formula of avelinoite may be applied to cyrilovite. The new formula and the corrected *x*-ray powder data for cyrilovite, which make the powder and single-crystal data consistent, also permit the relationship of cyrilovite to wardite to be recognized.

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