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The crystal structure and chemical composition of pollucite

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Auszug

Die Struktur des Pollucits von Rumford, Maine, wurde aus 213, mit einem Diffraktometer gemessenen Interferenzen bestimmt und bis $R = 0{,}050$ für alle Interferenzen verfeinert. Raumgruppe ist *Ia3d*, Gitterkonstante $a = 13{,}69$ Å. Das Strukturgerüst ist das gleiche wie beim Analcim. Die großen Hohlräume sind von Caesiumatomen und Wassermolekülen besetzt. Die Natriumatome befinden sich wie beim Analcim in der Punktlage $24c$, $[\frac{1}{4}\,\frac{1}{8}\,0]$. Sie sind jedoch in größeren, von Wassermolekülen begrenzten Gruppen nach Zufall angesammelt. Die allgemeine Zusammensetzung des Pollucits läßt sich durch eine Formel von der Art $Cs_xNa_yAl_{x+y}Si_{48-x-y}O_{96} \cdot (16-x)\,H_2O$ mit $2y \geqslant 16 - x \geqslant y$ ausdrücken.

Abstract

The structure of pollucite from Rumford, Maine has been determined and refined. The space group is *Ia3d* with cell edge $a = 13.69$ Å. The structure was investigated using 213 independent reflection intensities measured on a single crystal diffractometer. Pollucite has the analcime framework with the cesium atoms occupying the large voids in the framework, as initially suggested by Náray-Szabó. The water molecules occupy the large voids of this same set which are not occupied by the cesium atoms. The sodium atoms are located in equipoint $24c$, at $\frac{1}{4}\,\frac{1}{8}\,0$, in the positions between the water molecules. The water molecules and the sodium atoms occupy the same positions as they do in analcime, but they occur only in randomly distributed clusters of atoms whose outer members are restricted to water molecules. The final structure was refined by least squares to an R value of 0.050 for all reflections. The chemical composition of pollucite can be expressed by a general formula similar to: $Cs_xNa_yAl_{x+y}Si_{48-x-y} \cdot O_{96}(16-x)\,H_2O$, in which $2y \geqslant 16 - x \geqslant y$.

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Introduction

The similarity of pollucite to analcime was first suggested by PISANI[1] in 1864. In 1936 the similarity was made plausible by STRUNZ[2], who showed that these two minerals had not only the same space group and cell dimensions but similar cell compositions as well.

In 1938 the structure of pollucite was investigated by NÁRAY-SZABÓ[3]. With oscillating-crystal photographs of specimens from Buckfield, Maine and Elba he verified that the space group was *Ia3d*, as determined by STRUNZ. The cell edge he found to be $a = 13.74$ kX ($= 13.77$ Å). NÁRAY-SZABÓ assumed a formula for pollucite analogous to that of analcime, namely $CsAlSi_2O_6 \cdot xH_2O$, and he proposed a structure based upon this formula.

The analcime structure had been solved by TAYLOR[4] in 1930. It consists of an open framework of SiO_4 and AlO_4 tetrahedra, within whose interstitial spaces Na and H_2O are located. The analcime framework, when referred to the isometric space group *Ia3d*, can be described by placing Si and Al together in equivalent position 48*g* and O in 96*h*. There is a large void in the framework centered around 16*b* at $\frac{1}{8}\,\frac{1}{8}\,\frac{1}{8}$.

NÁRAY-SZABÓ placed the Si, Al and O atoms in the positions of the analcime framework, but he placed the Cs atoms in equipoint 16*b* rather than 24*c*, where TAYLOR had placed the 16 Na atoms of the analcime cell. The location of the water molecules was not determined. For these assignments of atoms NÁRAY-SZABÓ found good agreement between his powder data and calculated *F*(*hkl*) values.

The structure based upon *Ia3d* makes no distinction between Si and Al atoms, since they are lumped together in 48*g* with a disordered arrangement. NÁRAY-SZABÓ preferred an ordered structure in which these chemically different atoms could be assigned to distinct positions in a subgroup of *Ia3d*. He found by trial that a very good

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[1] M. F. PISANI, Etude chimique et analyse du pollux de l'ile d'Elba. Comptes rendus Acad. Sciences [Paris] **58** (1864) 714—176.

[2] HUGO STRUNZ, Die chemische Zusammensetzung von Pollucit. Z. Kristallogr. **95** (1936) 1—8.

[3] ST. V. NÁRAY-SZABÓ, Die Struktur des Pollucits $CsAlSi_2O_6 \cdot xH_2O$. Z. Kristallogr. **99** (1938) 277—282.

[4] W. H. TAYLOR, The structure of analcite ($NaAlSi_2O_6 \cdot H_2O$). Z. Kristallogr. **74** (1930) 1—19.

agreement with his data was obtained with the tetragonal subgroup *I*4/*acd* when he placed Cs in 16*f*, Si in 32*g*, and distributed the 96 oxygen atoms in three sets of 32*g* positions with different parameters.

Mineralogists now recognize that natural pollucites contain Na as well as Cs. In 1944 NEL[5] suggested that pollucite and analcime were members of a series which are related by the substitution of Cs^+ for Na^+ plus H_2O. The present investigation was initiated to answer two fundamental questions concerning pollucite: (1) Is the symmetry of the crystal isometric or tetragonal? (2) How are the Na atoms and H_2O molecules distributed in the crystal?

Material and cell

Transparent, colorless pollucite from Rumford, Maine was obtained from the M.I.T. reference collection. Several fragments were examined optically and found to be isotropic with an index of refraction near 1.525. The specific gravity determined with a Berman torsion density balance is 2.94.

A minute fragment of pollucite was mounted for examination by the precession method and adjusted so that [110] was the dial axis. With this setting a simple rotation about the dial axis permitted taking precession photographs with the possible symmetry axes [001], $[1\bar{1}1]$ and $[1\bar{1}0]$ as precessing axes. This setting also permitted the interaxial angles to be conveniently measured on the dial. The symmetries found in the precession photographs and the interaxial angles measured on the dial are consistent with those of a cubic crystal having Friedel symmetry $4/m\,\bar{3}\,2/m$. The qualitative information obtained from these photographs also confirmed that the space group of pollucite is *Ia*3*d*.

The value of the cubic-cell edge, $a = 13.69$ Å, was obtained from a photograph taken with a precision back-reflection Weissenberg camera. The unit-cell content and ideal chemical formula of pollucite from Rumford, Maine were determined using the procedures outlined in *Crystal-structure analysis*, by BUERGER[6]. FOOTE's analysis[7] of this

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[5] H. J. NEL, Pollucite from Karibib, South West Africa. Amer. Mineral. **29** (1944) 443—452.

[6] MARTIN J. BUERGER, Crystal-structure analysis. John Wiley and Sons, Inc., 1960, 242—249.

[7] H. W. FOOTE, On the occurrence of pollucite, mangano-columbite and microlite at Rumford, Maine. Amer. J. Sci. **1** (1896) 457—459.

material, combined with a specific gravity of 2.94 and a cell edge of 13.69 Å, yield a cell content of

$$Cs_{11.7}Na_{3.1}Li_{0.25}K_{0.4}Al_{15}Si_{33}O_{96.2} \cdot 4H_2O.$$

For the purpose of computing intensities this was regarded as approximately:

$$Cs_{12}Na_4Al_{16}Si_{32}O_{96} \cdot 4H_2O.$$

Measurement of intensities

Several fragments of pollucite were ground in a sphere grinder. One exceptionally clear and perfect sphere of radius 0.17 mm was selected for intensity measurement. This was mounted for rotation about the *a* axis. No birefringence could be detected with a polarizing microscrope when this sphere was immersed in a liquid whose refractive index was that of pollucite. In addition, no indication of twinning could be detected on Weissenberg photographs.

The integrated intensities were measured on a single-crystal diffractometer having equi-inclination geometry. Cu$K\alpha$ radiation (nickel-filtered) and a proportional counter were used for the intensity measurements. Of the approximately 300 independent reflections in the Cu$K\alpha$ sphere, 213 were accessible to the diffractometer. The intensities were corrected for Lorentz, polarization and absorption factors. Sets of symmetrically equivalent reflections were measured during the course of the measurements in order to detect any possible deviation from cubic symmetry. No significant deviations were found, and no reflections other than those consistent with symmetry $Ia3d$ were observed. These results, in addition to the known isotropic optics and isometric form development of the crystals, left little doubt that pollucite is truly isometric.

Structure determination

The corrected values of the integrated intensities were used as the coefficients, $F^2(hkl)$, for computing a three-dimensional Patterson function. The main features of this function could be interpreted as requiring the heavy Cs atoms to be located in 16*b*, at $\frac{1}{8}\frac{1}{8}\frac{1}{8}$, which confirmed Náray-Szabó's location for this atom.

A Fourier synthesis was made using the observed $F(hkl)$'s and the phases determined by the Cs atoms. The Fourier maps revealed the Si(Al) and O positions, which proved to be those of the analcime framework, as deduced by Náray-Szabó.

Because of this result, the structure proposed by NÁRAY-SZABÓ for the space group *Ia3d* was tested. Structure factors for all reflections were calculated using the coordinates reported by NÁRAY-SZABÓ and the simplified composition $CsAlSi_2O_6$ assumed by him. For the purpose of these calculations, a preliminary choice was made for scale factor and reasonable isotropic temperature factors. The discrepancies in the calculated and observed structure factors gave a residual of $R = 0.249$. This low R value, in conjunction with the results found by the Fourier synthesis, indicated that NÁRAY-SZABÓ's structure was basically correct.

A decision was still required regarding the possible assignments of Na and H_2O to the voids in the framework. The computations based on the analysis given by FOOTE indicated that there are approximately 12 Cs, $4H_2O$, and 4 Na per cell. Because the 12 Cs atoms occupy an equipoint of multiplicity 16, these atoms must be disordered in these positions. In minerals such as analcime and high leucite, these 16*b* positions are all occupied. Thus it was reasonable to expect that the four remaining positions not occupied by Cs atoms in pollucite might be randomly occupied by 4 H_2O molecules or 4 Na atoms. A refinement was made in the isometric system of two models with alternative assignments of atoms to voids. Each model had the equivalent of 4 atoms missing, namely 4 Na in one case and 4 H_2O in the other. These models are briefly described in Table 1.

Table 1. *Two possible models for pollucite in Ia3d*

Model 1 (4 Na missing)			
	12 Cs + 4 H_2O	in equipoint	16*b*
	32 Si + 16 Al	in equipoint	48*g*
	96 oxygen atoms	in equipoint	96*h*
Model 2 (4 H_2O missing)			
	12 Cs + 4 Na	in equipoint	16*b*
	32 Si + 16 Al	in equipoint	48*g*
	96 oxygen atoms	in equipoint	96*h*

Refinement

A least-squares refinement of the pollucite structure was carried out using the full-matrix program SFLSQ 3 written by C. T. PREWITT for the IBM 7094 computer. For the two alternative models described in Table 1, the average scattering curves were computed for the combinatons: 16 Al + 32 Si, 12 Cs + 4 Na, and 12 Cs + 4 H_2O. The water molecule was represented by a fully ionized oxygen atom[8], and all other atoms were considered to be half ionized. Initial isotropic temperature coefficients were arbitrarily choosen and equal weights were given to all reflections. Only 202 of the 213 reflections were used in the refinement of the structure because four reflections suffered greatly from extinction and seven had intensity values below the limit of observation.

In the initial stage of refinement it was realized that both models of Table 1 had essentially the same scattering characteristics and should give equivalent values for the structure factors. The reason for this is that $Na^{+1/2}$ and O^{-2} are nearly isoelectronic ions, each having approximately 10 electrons. In every other aspect these models are the same, and therefore one set of calculations sufficed for both models at the beginning of the refinement.

The calculations were made with one cycle of least-squares, allowing only the scale factor to vary, followed by one cycle with fixed scale factor and isotropic temperature coefficients, allowing the free coordinates to vary, followed by one cycle in which only the individual isotropic temperature coefficients were allowed to vary. This greatly lowered the sum of all the discrepancies in the $F(hkl)$'s from a value of $R = 0.249$ to a value of $R = 0.097$.

Using the last values of the calculated structure factors, a difference Fourier synthesis was computed. The difference map clearly indicated that the omitted atoms occupy equipoint 24*c*. Either 4 Na or 4 H_2O must be statistically distributed over the 24 available sites of each cell.

If the nature of the coordination polyhedron about 24*c* is considered, a decision can be made for the possible assignments of atoms to equipoints 16*b* and 24*c*. For this purpose the interatomic distances in the polyhedron, and the ionic radii of the atoms assigned to it must be taken into account. Equipoint 24*c* has symmetry 222 and lies at the center of a distorted octahedron, as shown in Fig. 1. Four of the corners of this octahedron (those to which the tetrahedra of Fig. 1 are attached) are the centers of oxygen atoms. These are equidistant from 24*c* at 2.5 Å, and lie at the corners of a slightly twisted rectangle containing 24*c*. The two remaining corners of the octahedron are the centers of voids at 16*b*, which must be occupied

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[8] T. SUZUKI, Atomic scattering factor for O^{-2}. Acta Crystallogr. **13** (1960) 279.

by either Cs, Na, H_2O or some combination of these atoms. The centers of these two voids are separated by a distance of nearly 4.8 Å, and they are joined by a two-fold rotation axis that includes 24*c* and is normal to the plane of the oxygen atoms. The four oxygen atoms form the edges of a window in the framework. This window, which connects the two voids in 16*b*, is actually composed of an eight-membered loop of tetrahedra, only two of which are shown in Fig. 1. All of the imaginable possibilities for assigning the different kinds of atoms to the center of the window, namely the point at 24*c*, and to the adjacent voids at 16*b* have been exhausted in Table 2, with the assumption that all three of these positions are occupied and not vacant.

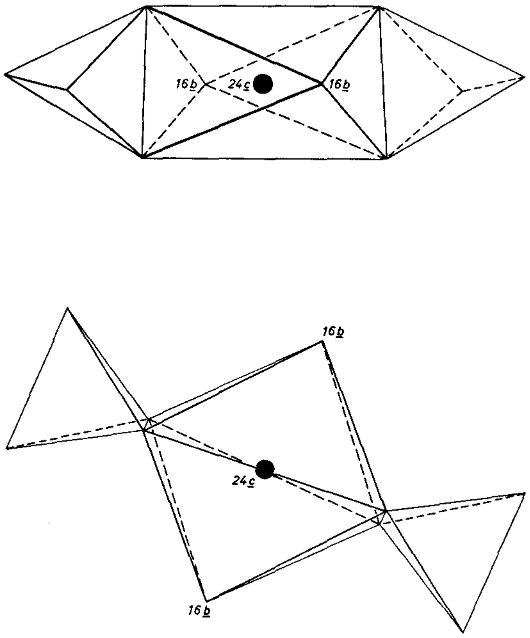

Fig. 1. The coordination octahedron around equipoint 24*c*

The only assignments of atoms listed in Table 2 which can be accepted as real possibilities must be those for which the size of the atom in the window is small enough to fit into the space available between the atoms in the voids at 16*b*. In addition, the arrangement of the atoms must not be one in which two positive ions would be juxtaposed. These restrictions leave for serious consideration only

Table 2. *The possible assignments of atoms to equipoint* 24*c and the two neighboring* 16*b positions*

Sequence of atoms along a two-fold axis between large voids			Sum of radii of atoms in voids	Available space between atoms in 16*b*	Diameter of atom occupying window
large void 16*b*	window 24*c*	large void 16*b*			
← 4.8 Å →					
Cs	H_2O	Cs	3.4 Å	1.4 Å	2.8 Å
Cs	Na	Cs	3.4	1.4	1.9
Cs	Cs	Cs	3.4	1.4	3.4
Na	H_2O	Na	1.9	2.9	2.8
Na	Na	Na	1.9	2.9	1.9
Na	Cs	Na	1.9	2.9	3.4
H_2O	H_2O	H_2O	2.8	2.0	2.8
H_2O	Na	H_2O	2.8	2.0	1.9
H_2O	Cs	H_2O	2.8	2.0	3.4
Cs	H_2O	Na	2.6	2.2	2.8
Cs	Na	Na	2.6	2.2	1.9
Cs	Cs	Na	2.6	2.2	3.4
Cs	H_2O	H_2O	3.1	1.7	2.8
Cs	Na	H_2O	3.1	1.7	1.9
Cs	Cs	H_2O	3.1	1.7	3.4
Na	H_2O	H_2O	2.4	2.4	2.8
Na	Na	H_2O	2.4	2.4	1.9
Na	Cs	H_2O	2.4	2.4	3.4

the two possible assignments Na—H_2O—Na and H_2O—Na—H_2O, and perhaps one other.

Because most of the voids are occupied by Cs, the assignment Cs—H_2O—Na was also tentatively accepted, since the sodium atom might be displaced from the center of the void. If one assumes that the sodium atoms are randomly distributed in the voids so that an occasional Cs atom is missing, but replaced by an Na atom, then the assignments Cs—H_2O—Na and Na—H_2O—Na could be considered to occur in the same structure model for pollucite. Although a water molecule has the symmetry 2*mm*, there can be no objection to placing it in the position 24*c*, whose symmetry is 222, because the water molecules could be statistically oriented to achieve a symmetry compatable with symmetry 222. Furthermore, the four oxygen atoms of the window are all equidistant from the proposed water location at 2.5 Å, which is a suitable distance for hydrogen bonds.

Table 3. *Assignments of atoms in pollucite for refinement in Ia3d*

Model 1

12 Cs + 4 H_2O	in the large voids	16*b* at $\frac{1}{8}\,\frac{1}{8}\,\frac{1}{8}$
3 Na	in the window	24*c* at $\frac{1}{4}\,\frac{1}{8}\,0$
15 Al + 33 Si	in the framework	48*g* at $x,\ \frac{1}{4}-x,\ \frac{1}{8}$
96 O	in the framework	96*h* at $x\,y\,z$

Model 2

12 Cs + 4 Na	in the large voids	16*b* at $\frac{1}{8}\,\frac{1}{8}\,\frac{1}{8}$
4 H_2O	in the window	24*c* at $\frac{1}{4}\,\frac{1}{8}\,0$
16 Al + 32 Si	in the framework	48*g* at $x,\ \frac{1}{4}-x,\ \frac{1}{8}$
96 O	in the framework	96*h* at $x\,y\,z$

Since the difference map revealed that there are atoms in 24*c*, this analysis of the coordination polyhedron around 24*c* leaves only two possible models for the pollucite structure. These are summarized in Table 3. Models 1 and 2 of Table 3 are similar to models 1 and 2 of Table 1 respectively.

The assignment H_2O—Na—H_2O, which leads to model 1 of Table 3, requires that each Na atom be bracketed by two water molecules in the adjacent voids. The similar assignment Cs—Na—H_2O is not possible because of the close juxtaposition of the positive ions. Because FOOTE's analysis indicates that the H_2O: Na ratio is between 1:1 and 4:3, model 1 implies that the H_2O molecules must be arranged so that all the Na ions are not only surrounded by them, but that the H_2O: Na ratio remains between 1: 1 and 4: 3. The only possibility of satisfying these requirements is for the Na ions and the H_2O molecules to occur together in small groups or clusters in the crystal. The interior of each cluster must be composed of Na ions, water molecules and that small portion of the framework that lies within the cluster. The boundaries of the cluster must be composed solely of water molecules, each having one or more Cs neighbors. Thus, model 1 calls for a distribution of the Na and H_2O which is not purely random. For this reason model 1 was thought to be an unlikely possibility at the beginning of this investigation[9]. Nevertheless model 1, with the assignment H_2O—Na—H_2O (i.e. to 16*b*—24*c*—16*b*) proved to be the best model for the structure.

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[9] RICHARD M. BEGER and MARTIN J. BUERGER, The crystal structure of the mineral pollucite. Nat. Acad. Sci. Proc. **58** (1967) 853–854.

The number of Na atoms was now reevaluated from FOOTE's analysis which gives the cell content:

$$Cs_{11.7}K_{0.4}Na_{3.1}Li_{0.25}Al_{15}Si_{33}O_{96.2} \cdot 4H_2O.$$

The sum of Cs + K + H_2O is approximately 16 per cell, which is in agreement with model 1. The sum of the alkali atoms, however, should be equal to the aluminum content, but this sum is 0.5 atoms too high. For reasons which are justified below, the aluminum content was accepted as correct, and the number of Na atoms was set equal to 3 for model 1 in Table 3.

With these changes made, the refinement of the structure of pollucite was resumed. Model 1 and model 2 of Table 3 were treated in an equal manner for the purpose of comparing them and possibly of choosing the better one. When the atoms were included in equipoint 24*c* and given an arbitrary isotropic temperature factor of $B = 0.8$, the R value for both models dropped from .097 to .086.

At this point a correction was applied for anomalous scattering as given in the International Tables. While the values for the other atoms in pollucite are comparatively small, the correction for the Cs atom with Cu$K\alpha$ radiation is relatively large. Because some of the equipoints are occupied by chemically different atoms, the values of $\Delta f'$ and $\Delta f''$ for these groupings of atoms were averaged according to the ratios of the numbers of each kind of atom in the equipoint.

A new weighing scheme was also introduced at this stage. This is based upon the discrepancies between $|F_c|$ and $|F_o|$ and is of a type first suggested by DE VRIES[10]. Equal weights, $w = 1$, were assigned to reflections having $|F_o| \geqslant 200$, and different weights, $w = |F_o|/k$, were assigned to reflections having $|F_o| < 200$. The least-squares program SFLSQ 3 minimizes the value of $w\,(|F_o| - |F_c|)^2$.

After the introduction of these changes, the two models were refined with isotropic temperature factors. The residual reached a minimum of $R = .065$ for model 1 and $R = .067$ for model 2. Since the principal difference between models 1 and 2 is the difference in the scattering power between 3 atoms and 4 atoms in 24*c*, one might possibly select the better of the two if it were possible to refine the occupancy of 24*c*. This proved to be impossible for two reasons: first the precise value of the isotropic temperature factor was not known in advance, and second the isotropic temperature factor and

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[10] A. DE VRIES, On weights for a least-squares refinement. Acta Crystallogr. 18 (1965) 1077.

the occupancy of 24*c* are highly interdependent. The correlation between these two parameters was computed with the correlation matrix[11] in one cycle of least-squares refinement. The computed value of the correlation coefficient was $\varrho = .85$. Another indication of the strong interaction in the occupancy and the B of atoms in 24*c* is the very high error estimates found for the B values of atoms in this equivalent position.

The refinement of the structure was continued with the introduction of anisotropic thermal parameters for all atoms. These changes reduced the R values of models 1 and 2 to .045 and .047 respectively. With the final values of the calculated structure amplitudes a difference Fourier synthesis was computed for both models. The difference maps indicated that both of these models closely represent the characteristics of the real crystal structure.

Discussion of the structure models

A meaningful way for comparing the results of the least-squares refinement of the two models is provided by the significance tests described by HAMILTON[12]. The necessary information for such tests is given in Table 4. Because the R values for model 2 are higher than those for model 1, we may reasonably wish to decide whether or not model 2, as compared to model 1, is inconsistent with the data for pollucite. If we hypothesize that the values of the 20 parameters given by the anisotropic refinement of model 2 correctly describe the structure of pollucite, then we can test the hypothesis with HAMILTON's tables and the ratio of the R values of the two models in Table 4. From HAMILTON's tables for testing the R-value ratio we find for 20 parameters, 182 degrees of freedom, and a 25 per cent level of significance, that this ratio is 1.064; this is nearly equal to 1.063 from Table 4. Therefore, near the 25% level of significance we can reject the hypothesis that model 2 correctly describes the structure of pollucite. In other words, the test tells us that if we reject such models as model 2 in favor of model 1, we will be wrong 25% of the time.

In the case of pollucite, however, there is additional cogent evidence for accepting model 1 and rejecting model 2. Most of the evidence

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[11] S. GELLER, Parameter interaction in least-squares structure refinement. Acta Crystallogr. **14** (1961) 1026—1035.

[12] WALTER C. HAMILTON, Significance tests on the crystallographic R factor. Acta Crystallogr. **18** (1965) 502—510.

Table 4. *Comparison of the results of the refinement for the significance tests on the R value*

Kind of refinement	Isotropic		Anisotropic	
Model	1	2	1	2
Number of structure amplitudes	202	202	202	202
Number of parameters	9	9	20	20
Degrees of freedom	193	193	182	182
Weighted R	4.3	4.4	3.2	3.4
R of model 2 divided by R of model 1	1.024		1.063	
$\mathcal{R}$ (from Hamilton's tables) at				
25 % significance level	1.029		1.064	
50 % significance level	1.021			

was first recognized by Nel[5] in the chemical analyses of pollucite specimens from different localities. In Table 5 all but one of the analyses[13] of pollucite referred to by Nel have been assembled from the original literature. In the same table the cell content has been computed for each analysis, and certain sums and ratios of atoms have been computed for comparison. Table 5 illustrates the relationships deduced by Nel, and in addition some further relationships which lead to important conclusions. From the analyses Nel deduced that:

1. The number of Cs ions is inversely proportional to the number of Na ions in the unit cell.
2. The number of Cs^+ ions plus H_2O molecules is 16.

Both of these relationships are well supported by Table 5. The second relationship ($Cs^+ + H_2O = 16$) is the basis for model 1 of Table 3, which the author accepts as the best model for the pollucite structure.

Nel ventured to suggest that the Na ions replace the Cs ions in the voids, and that each Na thereby makes room for an H_2O molecule. Nel might not have suggested this if he had weighed the fact that the number of Na ions (plus other small ions, e.g. Li) is always less than the number of H_2O molecules. This important observation is emphasized by the author in Table 5, where the $H_2O/(Na + Li +$

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[13] The omitted analysis is of pollucite from Varutrask, and contains P_2O_5 in some form of impurity. The formula reported by Nel[5] for this analysis is $Cs_{11}Na_3Al_{14}Si_{34}O_{96} \cdot 5H_2O$.

Table 5. *Analyses of pollucites*

	1	2	3	4	5	6	7
SiO_2	43.65 %	43.51 %	44.28 %	45.20 %	50.07 %	45.7 %	45.25 %
Al_2O_3	16.84	16.30	16.32	16.98	17.19	17.2	16.38
Fe_2O_3							0.77
Cs_2O	36.14	36.10	35.83	33.02	24.54	30.2	32.31
K_2O	0.38	0.48	0.38	0.61	0.17		0.19
Rb_2O						1.3	
Na_2O	2.09	1.68	1.59	2.04	4.34	2.8	2.28
Li_2O	0.08	0.05			0.09		
CaO		0.22	0.13		0.09		0.72
H_2O	1.58	1.50	1.62	2.04	3.81	2.66	2.66
	100.00	99.84	100.15	99.88	100.30	99.86	100.56

Number of ions on the basis of 48 (Si + Al)

	1	2	3	4	5	6	7
Si	33.00	33.30	33.47	33.26	34.17	33.25	33.65
Al	15.00	14.70	14.53	14.74	13.83	14.75	14.35
Fe							0.43
Cs	11.65	11.78	11.55	10.36	7.14	9.37	10.24
K	0.38	0.47	0.37	0.57	0.15		0.18
Rb						0.56	
Na	3.07	2.50	2.33	2.90	5.74	3.95	3.28
Li	0.24	0.15			0.25		
Ca		0.18	0.11		0.07		0.58
O	96.16	96.28	95.93	95.56	95.79	95.97	96.82
H_2O	3.99	3.82	4.09	5.00	8.67	6.45	6.61

Sums and ratios of atoms for some of the elements

	1	2	3	4	5	6	7
Alkali + Ca + Fe	15.34	15.08	14.36	13.83	13.35	13.88	14.28
Alkali/Al	1.023	1.026	0.988	0.938	0.965	0.941	0.955
$Cs+K+H_2O$	16.02	16.07	16.01	15.93	15.96	16.38	17.02
$H_2O/(Na+Li)$	1.205	1.350	1.676	1.724	1.431	1.633	1.541

1. Rumford, Maine[7].
2. Hebron, Maine[14].
3. Tamminen quarry, Greenwood, Maine[15]
4. Leominister, Massachusetts[15]
5. Oxford Milling and Mining Co. Greenwood, Maine[15].
6. Karibib, South West Africa[5]
7. Island of Elba[2].

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[14] H. G. WELLS, On the composition of pollucite and its occurrence at Hebron, Maine. Amer. J. Sci., ser. III, no. 141 (1891) 213—220.

[15] W. E. RICHMOND and F. A. GONYER, On pollucite. Amer. Mineral. 23 (1938) 783—789.

+ Ca + Fe) ratio is given at the bottom of each column. The ratio is always larger than 1, but less than 2. If certain assumptions can be made, these are the limits of the H_2O/Na ratio which are suggested by model 1 of Table 3.

The water in pollucite must occur in clusters of two or more molecules which enclose a smaller number of Na ions. Each water molecule within a cluster has three potential Na positions around it. The symmetry of the water position is 32, and the Na positions are on the two-fold axes at a distance of 2.4 Å from the water site. The three Na positions are thus related by three-fold symmetry. Although there is no obvious reason why any of the Na positions need be occupied by the ions, experience with sodium-bearing zeolites[16] has shown that the tetrahedral charge distribution of water is compatible with two Na neighbors, and that a water molecule would be less likely to have three, one, or zero Na neighbors. Since the positions of Na and H_2O are the same as in analcime, we may assume that, like analcime, each H_2O molecule tends to have two neighboring Na ions. This assumption, as a first approximation, helps to explain the H_2O/Na ratios as found in pollucite analyses.

In analcime the H_2O/Na ratio is 1, while in pollucite it varies between 1 and 2. An easy explanation of this can be appreciated from the one-dimensional analogy of a chain. This chain, however, is composed of two kinds of links which alternate in regular fashion. The odd-numbered links, including the first and the last, can be thought of as the H_2O molecules, while the even-numbered links are the Na ions. In analcime the chain is of infinite length and none of the links is missing, so that the odd- and even-numbered links are in the ratio 1:1. In pollucite the chain is no longer infinite, but is composed of numerous small segments. Each segment of the chain represents a cluster of H_2O and Na in the crystal.

As the ratio of the odd- to even-numbered links in a chain segment depends upon the length of that segment, so, by analogy, the H_2O/Na ratio in a cluster depends upon the size of that cluster. The obvious upper limit of the ratio is for the smallest possible cluster: H_2O—Na—H_2O where the H_2O/Na ratio is 2. If the H_2O/Na ratio for the whole crystal depends to some extent upon the average size of the H_2O-Na clusters, then one might expect that the chain analogy would allow one to predict what the H_2O/Na ratio should be, solely

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[16] W. H. Taylor, The nature and properties of aluminosilicate framework structures. Proc. Royal Soc. [London] A, **145** (1934) 80—103.

on the basis of the number of H_2O molecules per cell as indicated by the chemical analysis. For example, if there are 4 H_2O molecules per cell, as there are in analyses 1, 2 and 3 of Table 5, then the chain analogy allows us to assume that the average H_2O-Na cluster in the crystal should be represented by the chain segment H_2O—Na—H_2O—Na—H_2O—Na—H_2O. The predicted H_2O/Na ratio would be $4/3 = 1.33$. The ratios calculated[17] for analyses 1 and 2 agree very well with this predicted value, but analysis 3 has a much higher value. The Na content of analysis 3 is lower than expected by the chain analogy. Some of the Na links are missing from the chain segments.

The one-dimensional analogy does not exactly describe the three-dimensional crystal, because the ends of the analogous chain segments are single Cs atoms, whereas the boundaries of the H_2O-Na clusters are many Cs atoms. The Cs atoms may interfere with the arrangement of the Na atoms around some of the H_2O molecules at the boundaries of the clusters. Because an Na ion cannot lie between an H_2O molecule and a Cs ion, an H_2O molecule at the boundary of a cluster may have less than two Na neighbors (i.e. one or none). This interference is especially severe if the H_2O molecule has more than one Cs neighbor or two H_2O neighbors that are satisfied with Na ions in their other windows. Although it is possible that some of the H_2O molecules may be isolated from Na ions by having 3 Cs ions for near neighbors, it is assumed that the H_2O molecules are less likely to have this arrangement because of their tetrahedral charge distribution. This is also a reasonable assumption if the H_2O molecules available are not much greater in number than that which is necessary to enclose the Na ions. Because the occurrence of the Na ions at the boundaries of the clusters may be restricted, and because some of the H_2O molecules may be isolated from the Na ions, the chain analogy allows one to predict only an approximate minimum value of the H_2O/Na ratio. The actual value of this ratio should lie between the predicted minimum and 2.

In pollucite crystals, Cs + H_2O sum to 16 (model 1), and the number of Na atoms is less than the number of H_2O molecules. Consequently, the sum of the alkali atoms must be less than 16 when the crystal contains water. The basis for model 2 of Table 3 is that the alkali atoms should sum to 16 in number. In Table 5 the sum of

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[17] The Na + Li content of analysis 1 is probably a little too high as evidenced by the disagreement in the alkali/Al ratio.

these atoms is always less than 16 by a significant amount, and it is as low as 13.35 in one case. Furthermore, the sums of the alkali atoms appear to be equal to the corresponding aluminum contents in these analyses. Thus the aluminum content of pollucite crystals

Table 6. *Final positional and thermal parameters of pollucite*
(Model 1)

Atom	Atomic coordinates and standard deviations						Equivalent isotropic[18] temperature factor
	x	$\sigma(x)$	y	$\sigma(y)$	z	$\sigma(z)$	
Si,Al	.6621	.0001	.5879	.0001	$\frac{1}{8}$		0.68 Å^2
O	.1037	.0004	.1340	.0005	.7203	.0003	2.28
Cs,H_2O	$\frac{1}{8}$		$\frac{1}{8}$		$\frac{1}{8}$		2.15
Na	$\frac{1}{4}$		$\frac{1}{8}$		0		1.57

Table 7. *Observed and calculated structure factors of pollucite*
(Model 1)

h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c
4	0	0	614.7	648.3	6	5	1	56.5	53.6	13	7	2	62.1	62.6	11	7	4	8.0	3.2
8			361.5	358.5	8			15.9	17.6	15			51.6	56.9	13			5.9	5.7
12			147.1	146.1	10			155.5	156.8	10	8	2	24.3	21.5	8	8	4	128.6	128.9
16			43.2	46.0	12			14.7	11.5	12			15.9	10.7	10			2.2	21.9
2	2	0	58.5	47.5	14			91.3	90.2	14			2.2	1.2	12			88.3	89.9
4			59.9	52.0	16			4.9	5.5	11	9	2	81.5	81.7	14			9.8	10.2
6			52.6	49.0	7	6	1	110.2	112.5	13			80.4	86.0	11	9	4	31.1	30.8
8			70.3	63.5	9			21.5	18.5	12	10	2	17.9	18.4	13			17.4	14.7
10			30.5	31.3	11			44.6	45.3	11	11	2	71.9	73.0	10	10	4	10.9	7.6
12			58.1	54.0	13			26.8	27.4	6	3	3	194.8	188.7	12			27.2	28.2
14			2.2	1.4	15			28.8	31.8	10			157.3	154.0	6	5	5	75.0	67.1
16			44.2	46.4	8	7	1	100.9	99.2	14			90.7	94.7	10			164.0	162.4
4	4	0	349.3	357.8	10			97.4	100.9	5	4	3	29.7	26.5	14			94.1	93.8
6			105.1	103.2	12			32.6	30.9	7			33.1	26.0	7	6	5	91.5	88.0
8			225.7	222.1	14			63.7	68.4	9			34.8	13.4	9			53.6	52.9
10			61.4	59.7	9	8	1	26.1	21.7	11			20.5	18.5	11			80.0	80.9
12			108.7	109.0	11			74.6	74.4	13			14.4	12.1	13			38.9	41.3
14			26.1	28.0	13			41.0	41.2	15			12.9	14.1	8	7	5	71.2	67.0
16			34.1	36.4	10	9	1	77.9	79.8	6	5	3	93.7	91.5	10			71.4	70.1
6	6	0	74.7	70.3	12			2.2	1.2	8			41.8	38.9	12			14.1	10.5
8			102.8	100.2	14			40.2	43.0	10			119.4	119.2	14			38.2	38.4
10			37.8	36.1	11	10	1	32.1	33.0	12			8.4	5.5	9	8	5	22.8	21.3
12			94.6	96.2	13			42.5	45.8	14			62.0	62.4	11			61.1	59.3
14			6.5	2.2	12	11	1	19.8	18.8	7	6	3	162.4	160.7	13			39.0	41.6
8	8	0	154.5	156.0	4	2	2	44.7	35.6	9			20.7	18.2	10	9	5	110.1	112.3
10			62.1	61.1	8			28.1	26.1	11			53.6	54.5	12			2.2	2.1
12			77.2	78.9	12			20.5	18.7	13			23.9	26.0	11	10	5	51.8	53.8
14			6.9	6.9	16			14.3	12.0	15			22.9	24.0	8	6	6	51.7	50.4
10	10	0	26.5	24.4	3	3	2	277.6	261.7	8	7	3	38.6	36.9	12			47.1	44.4
12			47.0	50.5	5			152.4	141.5	10			91.5	91.7	7	7	6	145.2	143.3
2	1	1	40.6	33.3	7			229.4	223.3	12			9.5	7.4	9			24.1	22.9
6			49.6	49.9	9			24.8	19.5	14			48.3	52.6	11			53.8	51.9
10			186.3	187.3	11			81.7	78.1	9	8	3	19.2	21.1	13			28.8	30.1
14			122.4	122.7	13			50.1	51.4	11			28.6	26.3	10	8	6	21.3	23.0
3	2	1	138.6	121.2	15			32.0	34.5	13			14.9	12.9	12			25.3	22.9
5			78.4	71.9	6	4	2	39.9	36.3	10	9	3	26.5	24.0	9	9	6	85.9	91.3
7			133.4	128.7	8			38.7	33.9	12			14.1	12.1	11			58.8	63.1
9			27.9	20.0	10			40.9	22.6	11	10	3	14.2	14.8	12	10	6	21.4	22.9
11			41.4	40.0	12			40.1	38.4	13			15.8	16.4	11	11	6	35.6	38.9
13			39.4	38.8	14			7.2	3.2	12	11	3	4.3	2.0	10	7	7	52.6	55.3
15			20.9	22.5	16			32.8	36.6	4	4	4	209.1	202.7	9	8	7	43.7	45.7
4	3	1	69.8	67.2	5	5	2	96.1	89.7	8			155.3	151.9	11			9.8	3.2
6			106.4	104.0	7			126.6	117.8	12			98.6	101.5	13			9.9	9.4
8			78.4	76.4	9			66.2	64.3	7	5	4	54.9	49.1	10	9	7	17.0	12.4
10			155.3	155.3	11			91.6	90.1	9			26.6	19.0	12			16.5	14.9
12			27.8	27.2	13			52.1	54.9	11			46.3	44.9	11	10	7	14.3	12.6
14			99.3	100.1	15			56.0	57.2	13			35.4	35.0	8	8	8	139.5	142.3
16			6.5	3.3	8	6	2	32.9	31.6	15			30.2	30.0	12			104.1	113.2
5	4	1	21.7	21.8	10			2.2	0.1	6	6	4	56.7	50.6	11	9	8	40.4	43.7
7			81.3	76.9	12			27.4	25.1	8			41.8	34.4	10	10	8	10.3	11.9
9			24.8	21.9	14			2.2	0.4	10			20.7	19.6	10	9	9	103.1	112.4
11			57.4	56.5	7	7	2	212.4	212.1	12			57.6	55.7					
13			36.6	35.7	9			46.1	41.9	14			9.9	9.4					
15			37.4	38.6	11			94.8	92.6	9	7	4	32.4	32.4					

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[18] W. C. Hamilton, On the isotropic temperature factor equivalent to a given anisotropic temperature. Acta Crystallogr. **12** (1959) 609—610.

is always less than 16 and the Si content is consequently higher than 32. The relationships exhibited by the analyses substantiates model 1, which explains the chemical variation in pollucite crystals to a remarkable degree.

According to a recent report by NEWNHAM[19] the near-infrared absorption spectrum of pollucite is similar to the water-vapor spectrum. NEWNHAM concluded that the H_2O molecules cannot be strongly bonded to the silicate framework, but must be so arranged in the structure that they may behave like free water molecules. On the basis of this evidence one may conclude that the water molecules are better placed in the voids than in the windows of the framework. Thus the infrared evidence also favor model 1.

The atomic parameters and the standard deviations for model 1 are given in Table 6. The structure-factor calculation with all 213 reflections gave model 1 a residual value of $R = .050$. The final observed and calculated structure amplitudes are given in Table 7.

Description and discussion of the structure and composition

The general nature of the composition and structure of pollucite has been known since the work of STRUNZ and NÁRAY-SZABÓ, but a detailed discussion of this mineral has not previously been given. Pollucite has symmetry $Ia3d$ and is similar to analcime, which, to a first approximation, also has this symmetry in spite of the fact that it tends to be birefringent and may give x-ray spectra inconsistent with $Ia3d$. Pollucite and analcime have essentially the same cell dimensions and the same aluminosilicate framework. If the positional parameters for the Si,Al and O atoms given in Table 6 are compared with those given in the recent refinement of analcime by KNOWLES, RINALDI and SMITH[20], the values are found to be nearly identical. The fractional coordinates differ only in the third or fourth decimal place. The water molecules and Na atoms occupy the same equivalent positions in pollucite as they do in analcime. Thus the bond angles and interatomic distances relating the atoms Si, Al, O, Na and H_2O are substantially identical in the two minerals.

In pollucite the aluminosilicate framework is composed of 4-, 6-, and 8-membered loops of tetrahedra, which are normal to $\bar{4}$, $\bar{3}$ and

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[19] R. E. NEWNHAM, Crystal structure and optical properties of pollucite. Amer. Mineral. 52 (1967) 1515—1518.

[20] C. R. KNOWLES, F. F. RINALDI and J. V. SMITH, Refinement of the crystal structure of analcime. Indian Mineral. 6 (1965) 127—140.

two-fold rotation axes respectively. Examples of these loops are designated by numbers in Fig.2. The framework is best visualized in terms of 6-membered loops of tetrahedra. With the origin at $\bar{3}$ there is one independent 6-membered loop in each octant of the cell. In this manner it is easy to see that there are $8 \times 6 = 48$ Si,Al per cell. Likewise, since every oxygen atom is common to two tetrahedra, there are $\frac{4}{2} \times 8 \times 6 = 96$ oxygen atoms per cell.

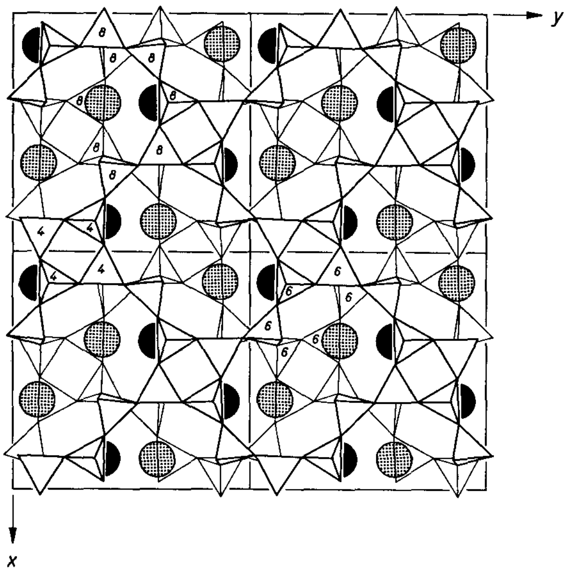

Fig. 2. Projection on (001) of the aluminosilicate framework and the Cs,H_2O atoms in pollucite. The lower halves of four cells are shown. Cs,H_2O atoms are represented by solid circles at $\frac{1}{8}c$ and stippled circles at $\frac{3}{8}c$

There have been many previous attempts by others to assign a general formula to the composition of pollucite. The author suggests that a general formula similar to one of the following two might be useful:

$$Cs_xNa_yAl_{x+y}Si_{48-x-y}O_{96} \cdot (16-x)\,H_2O, \text{ in which } 2y \geqslant 16-x \geqslant y$$

or

$$Cs_{16-x}Na_yAl_{16-x+y}Si_{32+x-y}O_{96} \cdot x\,H_2O, \text{ in which } 2y \geqslant x \geqslant y.$$

Within the framework there are 16 large voids at $\frac{1}{8}\frac{1}{8}\frac{1}{8}$ etc., on the three-fold axes. Each of these voids is framed by two 6-membered

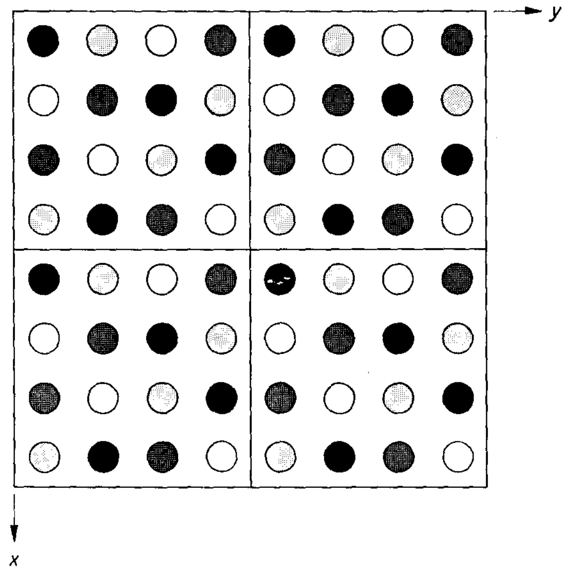

Fig. 3. Projection on (001) of the Cs atoms and H_2O molecules in pollucite. Four cells are shown. Solid black circles are atoms at $\frac{1}{8}c$, heavy stippled circles at $\frac{3}{8}c$, light stippled circles at $\frac{5}{8}c$, and open circles are at $\frac{7}{8}c$

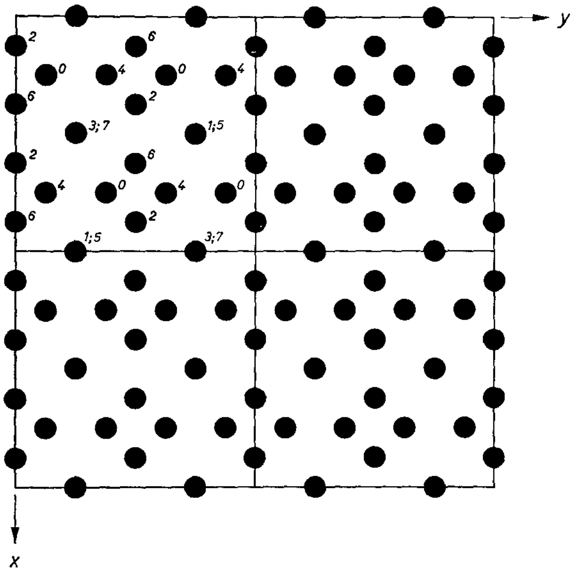

Fig. 4. Projection on (001) of the Na atoms in pollucite. Four cells are shown. The fractional z coordinates are in eights (e.g. 5 signifies $\frac{5}{8}c$)

loops of tetrahedra centered on the three-fold axis, and by three 8-membered loops related by the three-fold rotation. The voids form channels along the nonintersecting three-fold axes, and the walls of these channels are composed of 6-membered loops of tetrahedra. The channels along the three-fold axes connect with each other by large openings in the 8-membered loops, which the author has previously referred to as windows. The term window is suggestive of the fact that one channel communicates with another by way of an 8-membered loop of tetrahedra, and that this opening is closed or open depending upon whether an Na atom is present or absent in the center of the loop.

In pollucite the large voids are filled by Cs ions and H_2O molecules. The symmetry of the Cs position is 32, and in accordance with this symmetry each Cs atom has three nearest neighbor Cs atoms at 4.84 Å on the two-fold axes, and two more distant neighbor Cs atoms at 5.93 Å on the three-fold axis. The distribution of the Cs ions (and

Table 8. *Designation of atoms in pollucite*

Atoms	Coordinates in terms of the atoms in Table 6		
	x	y	z
Cs(1)	$\frac{1}{8}$	$\frac{1}{8}$	$\frac{1}{8}$
Cs(2)	$\frac{3}{8}$	$\frac{1}{8}$	$\frac{7}{8}$
H_2O	$\frac{1}{8}$	$\frac{1}{8}$	$\frac{1}{8}$
H_2O	$\frac{3}{8}$	$\frac{1}{8}$	$\frac{7}{8}$
Na(1)	$\frac{1}{4}$	$\frac{1}{8}$	0
Si(1) or Al(1)	$\frac{1}{2}+x$	$\frac{3}{4}+x$	$\frac{1}{8}$
Si(2) or Al(2)	$\frac{3}{8}$	$-x$	$\frac{3}{4}-x$
Si(3) or Al(3)	$\frac{3}{4}+x$	$\frac{5}{8}$	$\frac{1}{2}+x$
O(1)	$\frac{1}{2}-y$	$\frac{1}{2}+z$	$-x$
O(2)	x	$\frac{1}{2}-y$	$\frac{1}{2}+z$
O(3)	$\frac{1}{2}+z$	x	$\frac{1}{2}-y$
O(4)	$\frac{3}{4}+y$	$\frac{1}{4}-x$	$\frac{3}{4}-z$
O(5)	$\frac{3}{4}-z$	$\frac{3}{4}+y$	$\frac{1}{4}-x$
O(6)	$\frac{1}{4}-x$	$\frac{3}{4}-z$	$\frac{3}{4}+y$
O(7)	y	$\frac{1}{2}+z$	$-x$
O(8)	$-x$	y	$\frac{1}{2}+z$
O(9)	$\frac{1}{2}+z$	$-x$	y
O(10)	$\frac{3}{4}-z$	$\frac{1}{4}-y$	$\frac{1}{4}+x$
O(11)	$\frac{1}{4}+x$	$\frac{3}{4}-z$	$\frac{1}{4}-y$
O(12)	$\frac{1}{4}-y$	$\frac{1}{4}+x$	$\frac{3}{4}-z$
O(13)	$\frac{1}{4}-x$	$\frac{1}{4}-z$	$\frac{1}{4}-y$
O(14)	$-z$	$\frac{1}{2}-x$	y

Table 9. *Interatomic distances in pollucite*

Atom pair	Distance	Standard deviation
Cs(1) polyhedron	Cs(1)—O(*n*)	
O(1)	3.561 Å	.006 Å
O(2)	3.561	.006
O(3)	3.561	.006
O(4)	3.561	.006
O(5)	3.561	.006
O(6)	3.561	.006
O(7)	3.394	.005
O(8)	3.394	.005
O(9)	3.394	.005
O(10)	3.394	.005
O(11)	3.394	.005
O(12)	3.394	.005
Na(1) octahedron	Na(1)—O(*n*)	
O(1)	2.498 Å	.005 Å
O(6)	2.498	.005
O(7)	2.498	.005
O(11)	2.498	.005
H_2O(1)	2.420	
H_2O(2)	2.420	
Si(1) or Al(1) tetrahedron	Si(1) or Al(1)—O(*n*)	
O(2)	1.655 Å	.005 Å
O(12)	1.655	.005
O(13)	1.630	.004
O(14)	1.630	.004
O(1) polyhedron	O(1)—Atom (*n*)	
Si(2)	1.630 Å	.004 Å
Si(3)	1.655	.005
Cs(1) or H_2O(1)	3.561	.006
Cs(2) or H_2O(2)	3.394	.005

H_2O) is illustrated in Fig.3, and the distribution of the Na ions is shown in Fig.4. The Na ions and water molecules in pollucite occur together in clusters of atoms whose outer members are water molecules. The clusters are randomly distributed in the crystal as island-like regions, however, the Na and H_2O have definite crystallographic positions within these regions. The general distribution of the Cs, and H_2O in relation to the aluminosilicate framework can be seen

Table 10. *Bond angles in pollucite*

Atoms	Multiplicity	Bond angles	Standard deviations
Cs(1) polyhedron			
O(2)—Cs(1)—O(12)	6	44.2°	.2°
O(2)—Cs(1)—O(8)	6	75.5	.2
O(2)—Cs(1)—O(4)	3	89.0	.1
O(2)—Cs(1)—O(3)	6	76.5	.1
O(2)—Cs(1)—O(10)	6	70.4	.1
O(2)—Cs(1)—O(9)	6	150.9	.1
O(12)—Cs(1)—O(8)	3	94.6	.1
O(12)—Cs(1)—O(3)	6	116.4	.1
O(12)—Cs(1)—O(10)	6	112.0	.2
O(12)—Cs(1)—O(9)	3	151.5	.1
O(8)—Cs(1)—O(3)	6	89.1	.1
O(8)—Cs(1)—O(10)	3	45.0	.2
O(4)—Cs(1)—O(3)	3	133.3	.1
Na(1) octahedron			
O(6)—Na(1)—H_2O(1)	4	92.8°	.1°
O(6)—Na(1)—H_2O(2)	4	87.3	.1
O(1)—Na(1)—O(6)	2	174.5	.1
O(1)—Na(1)—O(7)	2	117.1	.2
O(1)—Na(1)—O(11)	2	63.2	.1
H_2O(1)—Na(1)—H_2O(2)	1	180	
Si(1) or Al(1) tetrahedron			
O(2)—Si(1)—O(12)	1	104.6	.4
O(2)—Si(1)—O(13)	2	111.8	.3
O(2)—Si(1)—O(14)	2	111.6	.3
O(13)—Si(1)—O(14)	1	105.5	.4
O polyhedron			
Si(2)—O(1)—Si(3)	1	144.5	.3
Si(2)—O(1)—Cs(1)	1	116.3	.2
Si(2)—O(1)—Cs(2)	1	104.8	.2
Si(3)—O(1)—Cs(1)	1	91.4	.2
Si(3)—O(1)—Cs(2)	1	97.4	.2
Cs(1)—O(1)—Cs(2)	1	88.2	.1

in Fig.2. The interatomic distances and bond angles for pollucite are given in Tables 9 and 10 respectively, and the coordinates of the atoms referred to in these tables are given in Table 8.

Acknowledgements

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