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THE PRESENT STATUS OF THE ANALCIME-POLLUCITE SERIES

P. ČERNÝ
Dept. Earth Sciences, University of Manitoba, Winnipeg, Man. Canada R3T 2N2

ABSTRACT

The compositions of natural pollucites known until recently cover the range $Poll_{49}$ - $Poll_{82}$. The finds of Cs-rich analcimes fill most of the gap $Poll_0$ - $Poll_{49}$. The largest of the three remaining gaps is that at the pollucite end, $Poll_{82}$ - $Poll_{100}$. The following nomenclature is proposed for the analcime-pollucite series: analcime for $Poll_0$ - $Poll_5$; cesian analcime for $Poll_5$-$Poll_{50}$; sodian pollucite for $Poll_{50}$ - $Poll_{95}$, and pollucite for $Poll_{95}$ - $Poll_{100}$.

The stoichiometric relations in most analyzed analcime-pollucites are compatible with the requirements of Beger's (1969) structure model: $Cs + H_2O = 1$, $Na < H_2O$, $Cs + Na < 1$, $Al < 1$ and $Si/Al > 2$ (all per 6 oxygens of anhydrous subcell). Most of the H_2O contents are higher than predicted by Nel (1944; $Na/H_2O = 1$) but lower than expected by Neuvonen & Vesasalo (1960; $H_2O = Na + 2\times$ excess Si over 2). The tetrahedral composition $Al_{0.90}Si_{2.10}$ is rather constant in all members of the series, independent of the $Na/Cs/H_2O$ ratios. The average content of 0.90 univalent cations fits the tetrahedral content of $Al_{0.90}$.

The unit cell edge 13.64 - 13.71Å seems to be rather constant, but the intensities of *x*-ray powder diffraction reflections show considerable variations with the change in the $Na + H_2O$ and Cs contents. All sodian pollucites are isotropic and produce cubic *x*-ray diffraction patterns, but the cesian analcimes show sectorial birefringence and splitting of those reflections that are doubled in monoclinic wairakite.

The relations between chemical composition and physical properties in the whole series follow the trends established earlier for sodian pollucites, but require a re-examination to provide more reliable determination curves.

INTRODUCTION

This study was prompted by the discovery of Cs-rich analcime in the Tanco pegmatite at Bernic Lake, southeastern Manitoba. Several samples were studied simultaneously by R. V. Gaines and J. Ito, and by the present author, and the results were summarized by the latter (Černý 1972). The compositions of these samples fall into the Na-rich half of the analcime-

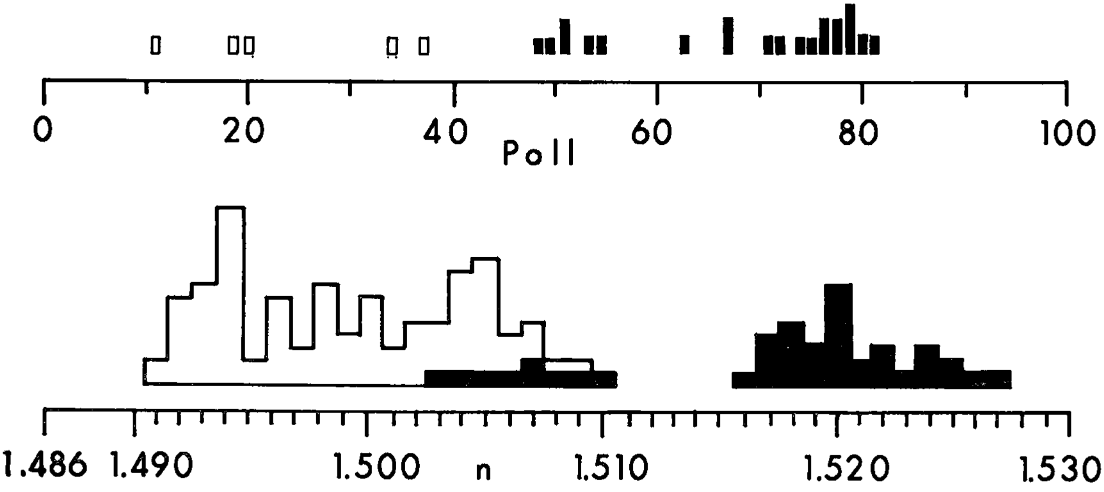

FIG. 1. Population of the analcime-pollucite series as known from natural specimens. Upper part: chemically analyzed specimens shown in terms of Poll; lower part: refractive indices of cesian analcimes and pollucites. The Poll and *n* scales are correlated only very approximately, using $n = 1.486$ for $Poll_0$ and taking $n = 1.520$ as characteristic of the group $Poll_{74}$ - $Poll_{80}$ (see Table 1). Black columns — isotropic sodic pollucites; white columns — birefringent cesian analcimes from Bernic Lake. The shortest columns in each graph represent the height equal to one sample.

pollucite series, and populate it almost comcontinuous at least in terms of chemical completely (Fig. 1). The existence of such a series, position, has been assumed impossible structurally (Newnham 1967; Henderson & Taylor 1969; and other earlier authors), but is compatible with the structure proposed by Beger (1969).

This latter structure model, with cubic symmetry and space group *Ia*3*d*, has the analcime tetrahedral framework with cesium in the large voids around the 16*b* equipoint (at ⅛, ⅛, ⅛). The water molecules populate the same large voids that are not occupied by cesium. The sodium cations are located in equipoint 24*c* (at ¼, ⅛, 0), between the water molecules. The water molecules and the sodium cations occupy the same positions as they do in analcime, but they occur only in randomly-distributed clusters whose outer members are restricted to water molecules.

This structure model is compatible with some peculiarities of pollucite stoichiometry, some of which have been recognized earlier: in contrast to the idealized formula $(Cs,Na)AlSi_2O_6 \cdot xH_2O$, the actual compositions shows $Cs + H_2O = 1$, $Na < H_2O$, $Cs + Na < 1$, and $Si/Al > 2$.

The purpose of the present study was (1) to re-examine the populatiton of the analcime-pollucite series in light of the recent find of cesian

TABLE 1. UNIT CELL CONTENTS AND PHYSICAL PROPERTIES OF THE ANALCIME-POLLUCITE MINERALS

	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Si	2.100	2.093	(2.100)	(2.100)	(2.100)	2.146	2.071	2.076	2.075		2.127	2.095	2.061	2.095
Al	.900	.916	(.900)	(.900)	(.900)	.854	.915	.925	.916		.861	.883	.937	.929
Fe^{3+}	-	-	-	-	-	-	.004	.014	.004		-	.010	-	-
ΣR^{IV}	3.000	3.009	(3.000)	(3.000)	(3.000)	3.000	2.990	3.015	2.995		2.988	2.988	2.998	3.024
Na	.900	.761	.730	.714	.460	.516	.300	.261	.298	[4.47]	.357	.261	.181	.248
Cs	-	.098	.169	.177	.310	.316	.433	.410	.465	[23.10]	.445	.494	.551	.590
Rb	-	tr.	tr.	-	.004	.002	-	-	.036	[.03]	-	-	.017	.016
K	-	.019	.013	.002	.110	.010	.058	.032	.027	[.48]	.009	.055	.103	.029
Li	-	-	-	-	-	-	.021	.073	.058		.015	.072	.028	-
Mg	-	-	-	-	-	-	.024	.040	.012		-	-	-	-
Ca	-	.002	-	.002	.020	.006	.048	.011	.017	[.05]	.004	.026	.001	-
Mn	-	-	-	-	-	-	-	tr.	-		-	-	-	-
Σ cat.	.900	.880	.912	.895	.904	.850	.884	.827	.913		.830	.908	.881	.883
H_2O	1.100	.96	.73	1.02	.52	.61	.50	.41	.16		.54	.40	.41	.41
Si/Al	2.33	2.28	(2.33)	(2.33)	(2.33)	2.51	2.26	2.24	2.27		2.47	2.37	2.19	2.25
Poll*	.0	11.1	18.6	19.8	34.3	37.2	49.0	49.6	50.9	51.3	53.5	54.4	62.5	66.8
CRK**	.0	13.3	19.9	20.0	46.9	38.6	55.6	53.4	57.8	54.6	54.6	59.7	76.1	71.9
n	1.486	1.492	1.493-6	1.496-7	1.501-9	1.505	-	-	-	1.503-10	1.507	1.527	1.520	1.517
opt.	-	biref.	biref.	biref.	biref.	biref.	-	-	-	isotr.	isotr.	isotr.	isotr.	isotr.
a(Å)	13.70	13.694	-	-	13.684	13.693	-	-	-	-	13.64	-	13.674	-
density	-	2.35	-	2.45	2.60	2.63	-	-	-	2.73	2.68	-	-	2.865

	15	16	17	18	19	20	21	22	23	24	25	26	27	28
Si	2.101	2.138	2.084	2.106	2.088	2.119	-	2.072	2.061	2.140	2.131	2.071	2.124	2.092
Al	.894	.870	.888	.913	.924	.869	-	.918	.940	.851	.868	.944	.900	.909
Fe^{3+}	tr.	-	.027	.002	-	.002	-	-	-	-	-	tr.	.003	-
ΣR^{IV}	2.995	3.008	2.999	3.021	3.012	2.990	-	2.990	3.001	2.991	2.999	3.030†	3.027	3.001
Na	.164	.211	.204	.110	.182	.172	[2.0]	.166	.192	.163	.171	.143	.118	.146
Cs	.595	.600	.634	.590	.658	.647	[32.0]	.732	.728	.747	.687	.628	.615	.722
Rb	.047	-	-	.024	-	-	[∿ .2]	-	-	.031	.014	.021	-	-
K	.029	.035	.011	-	.036	.036	[∿ .2]	.030	.002	.004	.006	tr.	-	.023
Li	.038	-	-	.036	-	-	-	.008	.015	.010	-	.002	.014	-
Mg	-	-	-	.012	-	-	-	-	-	-	-	-	.007	-
Ca	-	-	.036	.021	-	-	-	.011	-	-	.002	-	.014	.001
Mn	-	-	-	-	-	-	-	-	-	-	-	.001	-	-
Σ cat.	.873	.846	.885	.793	.876	.855	-	.947	.937	.955	.880	.796	.768	.89
H_2O	.27	.36	.41	.23	.31	.37	[1.8]	.24	.25	.23	.23	.29	.16	.25
Si/Al	2.35	2.45	2.35	2.31	2.26	2.44	-	2.26	2.20	2.51	2.45	2.20	2.36	2.30
Poll	66.8	70.9	71.7	74.2	74.9	75.7	75.8	77.3	77.6	78.2	78.4	78.9	80.1	80.5
CRK	76.8	75.0	72.9	77.4	79.0	79.9	78.5	80.5	77.9	81.7	80.7	81.7	81.0	82.8
n	1.522	-	-	1.524	1.520	1.521	1.520	1.525	-	1.520	-	1.517	-	1.52
opt.	isotr.	isotr.	isotr.	isotr.	isotr.	isotr.	isotr.	isotr.	isotr.	isotr.	-	isotr.	isotr.	isotr.
a(Å)	-	-	-	-	13.65	-	13.682	-	-	13.66	-	13.688	-	13.65
density	2.917	2.890	2.896	2.94	2.89	2.896	2.94	2.981	2.94-3.03	2.88	-	2.832	-	2.97

*Poll = Cs x 100/(Cs+Li+Na+K+Rb+Ca+Mg+Mn)
**CRK = (Cs+Rb+K) x 100/(Cs+Li+Na+K+Rb+Ca+Mg+Mn)

( ) from assumed values of SiO_2 and Al_2O_3
† includes .015 P

[ ] wt. % of oxides

1. Synthetic analcime, interpolated from Saha (1959, 1961)
2. Bernic Lake #1 (Černý 1972)
3. Bernic Lake #63568 (Černý 1972)
4. Bernic Lake #2-1 (Černý 1972)
5. Bernic Lake #2-2 (Černý 1972)
6. Bernic Lake #2 (Černý 1972)
7. Tin Mountain (Connolly & O'Hara 1929)
8. Siberia (Vlasov *et al.* 1964)
9. Far East (Vlasov *et al.* 1964)
10. Puklice (Miškovský 1955; new data given here)
11. Greenwood B (Richmond & Gonyer 1938)
12. Kalbinski Range (Ginzburg 1946)
13. Nagatare (Sakurai *et al.* 1972)
14. Karibib (Nel 1944)
15. Varuträsk (Quensel 1937)
16. Elba (Rammelsberg, quoted in Wells 1891)
17. Elba (Gossner & Reindl 1932)
18. Sayan Mts. (Melentjev 1961)
19. Leominster (Richmond & Gonyer 1938)
20. Jeclov (Miškovský 1960)
21. Maine (Newnham 1967)
22. Hebron (Wells (1891)
23. Rumford (Foote 1898, quoted in Gossner & Reindl 1932)
24. Bernic Lake (Nickel 1961)
25. Buckfield (British Museum, Natural Sciences lab. No. 3487)
26. Luolamäki (Neuvonen & Vesasalo 1960)
27. Northwestern European U.S.S.R. (Sosedko 1954, quoted in Melentjev 1961)
28. Greenwood A (Richmond & Gonyer 1938)

analcimes; (2) to check the crystallochemical requirements of Beger's structure on a broader scale; and (3) to re-examine the relations between chemical composition and physical properties in the series.

Collection and Treatment of Data

Chemical analyses, refractive indices, densities, and unit cell edges were collected from the literature, and *x*-ray powder diffraction data recorded for different members of the series. The chemical analyses were recalculated to atomic contents on the basis of 6 oxygens per anhydrous subcell; these are given along with physical properties in Table 1. For simplicity, the chemistry of individual specimens is characterized by their molecular percentage of the pollucite end member ("Poll") calculated as the atomic ratio Cs × 100/(Cs + Na + K + Rb + Ca + Mg). The minor components K, Rb, Ca, and Mg are arbitrarily grouped with Na.

Rb and K could be grouped with Cs, because of the large size of Rb and the preferred occurrence of K in the 16*b* equipoint sites of anhydrous compounds. The pollucite percentage based on this grouping is shown in Table 1 as the "CRK" index, (Cs + Rb + K) × 100/(Cs+ Na + K + Rb + Ca + Mg). However, the Rb and K contents are very small in all samples and the crystallochemical role of K in pollucite is uncertain. Thus the simpler Poll index is used in this study; it can be seen in Table 1 that the differences between the Poll and CRK indices are small, except for samples 5, 13 and 15.

Names of species and varieties are used in this study, and suggested for general use, in the following way: analcime for $\text{Poll}_{0\text{-}5}$; cesian analcime for $\text{Poll}_{5\text{-}50}$; sodian pollucite for $\text{Poll}_{50\text{-}95}$; and pollucite for $\text{Poll}_{95\text{-}100}$.

Chemical Composition

Population of the analcime - pollucite series

As shown in Figure 1, the classic pollucites cover the Poll_{62} - Poll_{82} range, and a separate group of Na-richer minerals shows compositions between Poll_{49} and Poll_{55}. The few chemically analyzed cesian analcimes from Bernic Lake are scattered over the earlier gap Poll_0 - Poll_{49}. Judging from the refractive indices measured on 101 specimens, the cesian analcimes range continuously from about Poll_{10} to Poll_{52}. Thus there seem to be just three relatively small gaps remaining at present: Poll_0 - Poll_9, Poll_{55} - Poll_{62}, and Poll_{82} - Poll_{100}. Only continued study of natural specimens will show if one or more of them are true. It is particularly interesting to note that, so far, no natural pollucite has been found with less then 18 mol. % analcime. The first gap, Poll_0 - Poll_9, seems to be populated by synthetic phases (Dr. T. Iiyama, personal comm. 1972).

Si/Al ratio

The slight deviation of the Si/Al ratio from the idealized value of 2 in several pollucites was recognized by Richmond & Gonyer (1938) and by Nel (1944). However, both authors did not

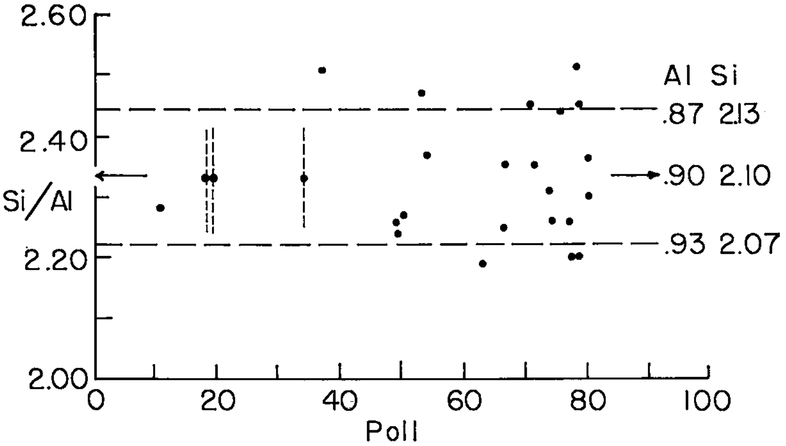

FIG. 2. Si/Al plotted against Poll. Note the concentration of most data points within the narrow Si/Al range 2.22 - 2.44. Dashed vertical lines indicate possible error in Si/Al for analyses in which these components were not directly determined (*cf.* Černý 1972).

realize how persistent this deviation was, and how constant its magnitude was, in all pollucites known to the dates of their respective studies.

Figure 2 shows the plot of the Si/Al ratios against the Poll contents. It is evident that the spread of the Si/Al values is well-centered around 2.33, *i.e.* $Al_{0.90}Si_{2.10}$, and two thirds of them lie between 2.22 ($Al_{0.93}Si_{2.07}$) and 2.44 ($Al_{0.87}Si_{2.13}$). This ratio appears to be independent of the Na and Cs contents. Thus the tetrahedral framework composition $(Al_{0.90}Si_{2.10}O_6)^{-0.90}$ can be considered typical for the whole series, undergoing only minor variations in individual members. As shown in the introduction, such a tetrahedral composition is required by the Beger structure model.

Presumably pure ferric analogues of pollucite were synthesized by Kopp *et al.* (1963), Kume & Koizumi (1965), and Kopp & Clark (1966), but natural pollucites are known to carry negligible Fe (sample 17 in Table 1).

Cationic content

As in the previously-known sodian pollucites, Na and Cs are also the most important cations in the cesian analcimes. K, Rb, Mg, and Ca are present only in very subordinate quantities (maximum atomic contents only 0.11, 0.05, 0.04, and 0.05, respectively, per 6 oxygens). Figure 3 shows the atomic contents of Cs plotted against those of Na plus minor cations; all the data closely follow the line connecting the 0.90 points, in accordance with the average anionic charge of -0.90. Deviations are caused by either slight variation in the Si/Al ratio, or by cationic deficiency which is rather common in zeolitic minerals (and may be apparent only because of lack of information about the possible H^+ and/or H_3O^+ contents). Formulas with high alkali excesses over Al (up to around 0.10) suggest faulty chemical analyses.

H_2O

The variations in the content of H_2O, not considered in detail by Richmond & Gonyer (1938), were related to the Na content by Nel (1944) in a simple way: the molecular content of H_2O equals the atomic content of Na (although his general formula does not reflect this relation properly): $Cs_{1-x}Na_{x-y}Al_{1-y}Si_{2+y}O_6 \cdot xH_2O$. Neuvonen & Vesasalo (1960) have proposed a variation of the H_2O content in pollucite with the Si content, as found for analcime by Saha (1959): the molecular content of H_2O equals the atomic content of Na plus twice the excess of Si over 2: $Cs_{1-x-y}Na_xAl_{1-y}Si_{2+y}O_6 \cdot (x+2y)H_2O$.

The Na/H_2O ratios derived this way are, however, lower than those expected in the Na + H_2O clusters in Beger's structure. Figure 4 shows that most Na/H_2O ratios determined for

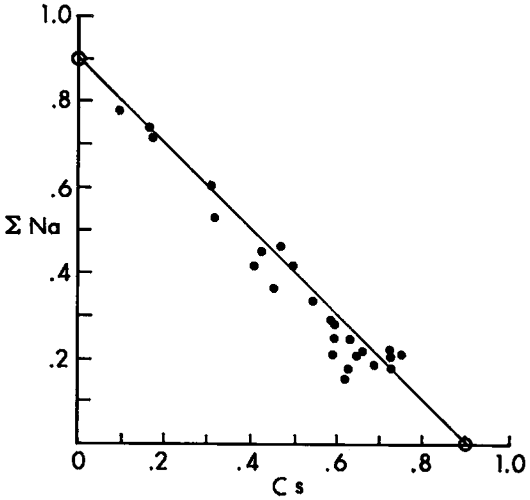

FIG. 3. Atomic contents of Cs plotted against those of Na plus all minor cations (Σ Na). Most data follow very closely the line connecting $Na_{0.90}$ and $Cs_{0.90}$, in accordance with the average tetrahedral Al content of .90.

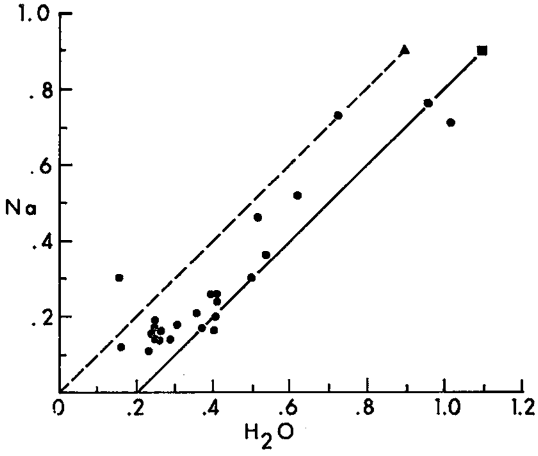

FIG. 4. Atomic contents of Na plotted against molecular contents of H_2O. Dashed line (zero to black triangle) follows Nel's assumption (number of H_2O molecules equals that of Na cations). Solid line (0.2 to black square) follows Neuvonen's & Vesasalo's proposal (number of H_2O molecules equals that of Na plus twice the excess of Si over 2). Most data are scattered between the two lines.

natural analcime-pollucites plot between the lines based on Nel's and Neuvonen's & Vesasalo's proposals, and thus approach closely the range expected in Beger's structure. It must be kept in mind that the analytical error in determination of water in pollucite tends to lead to somewhat lower-than-actual contents of H_2O (Barrer & McCallum 1953), and particularly the H_2O contents of Cs-rich members of the series should be influenced by this error.

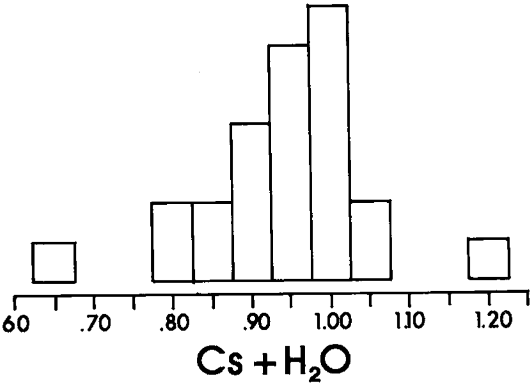

FIG. 5. Frequency of the Cs + H_2O contents in analcime-pollucites. Note that the distribution is not normal around 1.00 but frequencies are higher for slightly lower values.

As shown in Figure 5, most of the Cs + H_2O contents per 6 oxygens of anhydrous subcell are close to 1. This fact, first pointed out by Nel (1944), is one of the characteristics following from the Beger structure model. In accordance with the lower-than-actual H_2O contents expected in most analyses, the distribution of Cs + H_2O totals shown in Figure 5 is not normal around 1.00, but frequencies are higher for slightly lower values.

General formula

The stoichiometric relations discussed above conform with the general formula proposed by Beger (1969), which reads (after reduction to the presently used subcell) $Cs_xNa_yAl_{x+y}Si_{2x-y}O_6 \cdot (1\text{-}x)H_2O$.
Beger's limitation $2y \geqq 1\text{-}x \geqq y$ may be complemented by the empirical restriction of $x+y$ to ~ 0.90.

PHYSICAL PROPERTIES

Refractive index and density

Both these characteristics are known to increase with increasing Cs content. Their plots

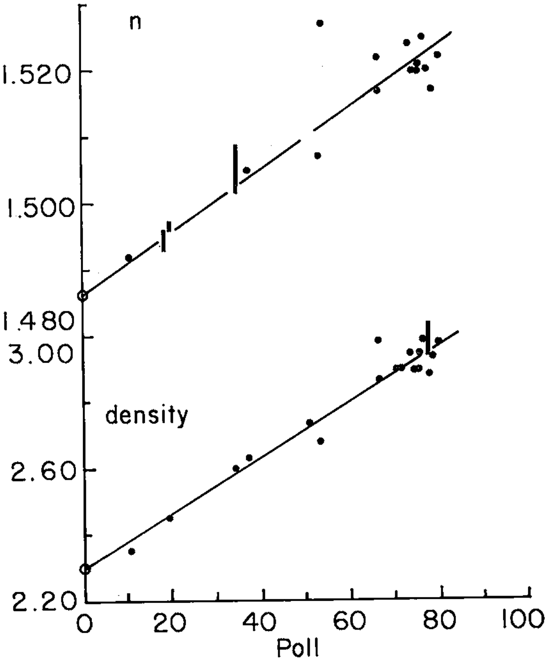

FIG. 6. Refractive indices and specific gravities plotted against Poll. The best fit lines are visual estimates, anchored at the analcime end of the series at 1.486 and 2.25 (see Table 1).

against mol. % Poll, shown in Figure 6, show a considerable scatter of data at the Cs-rich end of the series, which becomes, however, much less evident in a plot of *n* versus density, shown in Figure 7. This suggests that in many studies either the material used for checking the physical properties was not representative of the bulk material used for chemical analysis, or the determination of alkalis was not accurate. Since no data are available for a synthetic pollucite with $Al_{0.90}Si_{2.10}$, and those known for other compositions deviate from expected values so much that they are evidently not reliable (Kume & Koizumi 1965; Kopp & Clark 1966; Kopp *et al.* 1963; Plyushev 1959), it is not possible to anchor the lines in Figures 6 and 7 at the Poll end of the scale in a similar way as at the Anal end (which was calculated from data by Saha 1959, 1961). The slope of these lines is only a visual estimate.

Isotropic and anisotropic phases

All sodian pollucites described in the literature and examined to date by the author are isotropic, but the newly-discovered cesian analcimes from Bernic Lake show sectorial birefringence (Černý 1972). Analcime, of course, is

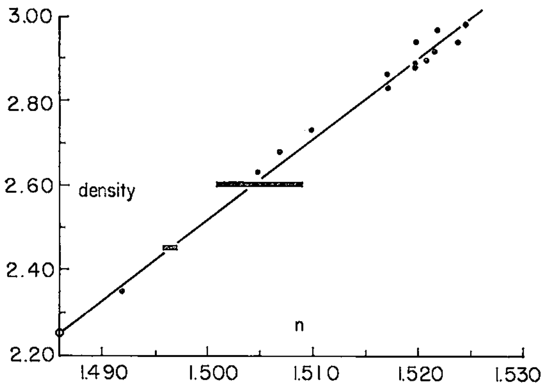

FIG. 7. Densities plotted against refractive indices. The line of best fit is visually estimated, with the 2.25/1.486 values for pure analcime used as the only fixed point (see Table 1). Note that the correlation between these two physical properties is distinctly better than that between them and Poll (Fig. 5).

known to be both isotropic and anisotropic (Coombs, 1955; Knowles *et al.* 1965; and others).

Unit cell edge

Saha (1959, 1961) found a rather broad variation of *a* with changes in the Si/Al ratio in analcime. This suggests that in the natural Anal-Poll series with its restricted Si/Al range *a* should be almost constant, provided the influence of the widely-changing Na/Cs ratio is negligible. The range of *a* values quoted in literature is indeed only 13.64 - 13.71Å. A more detailed discussion of these relations is not possible at present, as the available data come from different authors and were obtained by different (and frequently unspecified) methods. It would not be surprising if the actual range of *a* was much smaller.

X-ray powder diffraction data

The relative intensities of *x*-ray powder diffraction reflections show considerable variations in the Anal-Poll series. Similar to the variations in alkali-rich beryls, recognized by Evans & Mrose (1966) and treated quantitatively by Bakakin *et al.* (1970), the substitution (and different allocation) of heavy Cs for Na influences profoundly the diffraction and absorption of *x*-rays in these minerals. Figure 8 shows *x*-ray powder diffraction patterns recorded under identical conditions for a series of samples with different Poll contents. Besides the increase in intensity of reflections 321, 440, 532, 631, and 721, and the decrease in that of 211, 220, 400, 422, 431, 521, 640 and 800 with increasing Poll content, the sodian pollucite patterns are generally weaker.

The *x*-ray powder diffraction patterns of optically isotropic sodian pollucites display cubic symmetry but those of the anisotropic cesian analcimes from Tanco show a broadening of the same reflections (422, 521, 640, and 800) that are doubled in the anisotropic, monoclinic wairakite $CaAl_2Si_4O_{12} \cdot 2H_2O$ (Seki 1966, 1971; Seki & Oki 1969; Harada *et al.* 1972).

SUMMARY AND CONCLUSIONS

The preceding review establishes the natural analcime-pollucite group of minerals as an al-

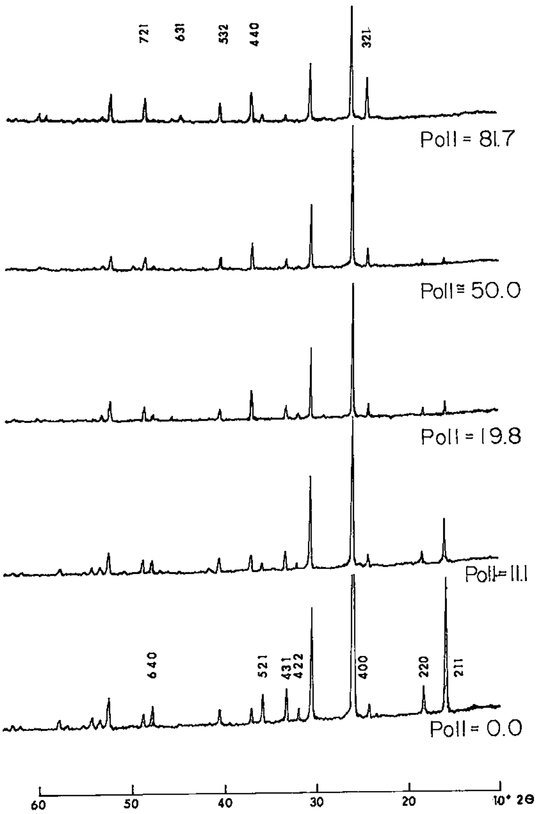

FIG. 8. *X*-ray powder diffractograms of five members of the analcime-pollucite series, taken under identical conditions. Reflections showing changes in intensity with changing chemical composition are indexed in those "end"-member patterns towards which their intensity increases. The cut-off height of the analcime 400 peak is at about two thirds of its true intensity.

most continuous isomorphous series. Only three small gaps seem to be present but these may disappear with continued research. The most prominent is that at the pollucite end of the series; the cesium-richest members in existence are sodian pollucites with more than 18 mol. % analcime.

The following nomenclature is proposed for the analcime-pollucite series: analcime for $Poll_0$-$Poll_5$, cesian analcime for $Poll_5$ - $Poll_{50}$, sodian pollucite for $Poll_{50}$ - $Poll_{95}$, pollucite for $Poll_{95}$ - $Poll_{100}$.

The stoichiometric relations derived from all published chemical analyses comply with the basic requirements of Beger's (1969) structure model: $Cs + H_2O = 1$; $Na < H_2O$, and thus $Cs + Na < 1$, which necessitates $Al < 1$ and $Si/Al > 2$. Most of the analyses show Al very close to 0.90 and Si/Al very close to 2.33.

The sodian pollucites are isotropic in accordance with the cubic space group *Ia3d* established by all structure refinements performed to date. However, the cesian analcimes are anisotropic, and their *x*-ray powder diffraction data are similar to those of monoclinic wairakite. A more detailed work on some of these Na > Cs phases may be useful to establish the possible structural causes of the anisotropism; these may be rather delicate and difficult to detect, as in the case of isotropic and birefringent analcimes (Knowles *et al.* 1965).

The relations between chemical composition and physical properties in the whole series follow the patterns established earlier for sodic pollucites. However, the presently available data show a rather wide scatter. A reexamination of refractive index, density, and *a* vs. chemical composition is being carried out at our Department.

To verify the continuity of the analcime-pollucite series, all new finds of different members of the series should be checked for their physical properties, and thoroughly examined if close to the gaps shown in Figure 1. This applies particularly to analcimes with possible low Cs contents from Li-rich pegmatites (Neuvonen & Vesasalo 1960; Rijks & v. d. Veen 1972), and to late hydrothermal "pollucites" from vugs (*e.g.* Richmond & Gonyer 1938; Miškovský 1955; Černý 1972).

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