PSEUDO-ISOMORPHISM AS ILLUSTRATED IN THOMSONITE

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Introduction

Thomsonite may seem hardly of sufficient importance to justify such extended discussions of its composition as have recently appeared;[1] yet, as it illustrates so well certain principles of mineralogical research, this further contribution to the subject will, it is hoped, not be considered superfluous.

That the analysis of a mechanical mixture will give the same results as that of an isomorphous mixture of the same components in corresponding proportions is such a self-evident proposition that its general recognition and acceptance would surely be expected; yet every now and then articles appear in which it is overlooked.   Before the development of modern microscopic methods of study of minerals this was more or less excusable; but at the present day it is surely not going too far to hold that *no claim of the isomorphism of two or more compounds should be accepted unless accompanied by optical evidence.*

The Composition of Thomsonite Proper.

In the paper noted, Gordon expressed surprise that the writer should consider a mineral to have a definite formula when by laboratory experiments its lime can be replaced by soda, potash, etc.   As a matter of fact, numerous minerals of quite definite formulas can be so changed, and this has no bearing on the matter at all.   He then (unconsciously) obscured the very replaceability he was trying to prove by recalculating a series of analyses of miscellaneous minerals, loosely classified as thomsonite, on the basis of $Na_2O = 1$.   To really demonstrate mutual replaceability it would have been necessary at least to use the *sum* of the Na_2O and CaO as unity (or a small integer); but even this is not the most informing procedure.

The method brought to the attention of mineralogists by Schaller[2] some years ago has not been used as widely as seems

[1] The writer, *Am. Min.*, **8**, 121-125 (1923); S. G. Gordon, *Proc. Acad. Nat. Sci. Philadelphia*, **76**, 103-107 (1924); A. N. Winchell, *Am. Min.*, **10**, 90-97 (1925).
    [2] *U. S. Geol. Survey Bull.*, **610**, 163-164 (1915).

desirable. As pointed out by several writers who discussed the subject at the time, least square methods of reduction would be more acceptable from the mathematical viewpoint, but most analytical data are not of sufficient accuracy to justify this, so the original plan is here favored. In brief, this is to divide each ratio by the nearest integer obtained by inspection, find the average of the several quotients, and divide each ratio in turn by this average.

By thomsonite is here understood the mineral to which the name was originally given. To arrive at its formula, the Schaller method will be employed, but only the three analyses made on material of known optical character will be admitted. The transparent columnar mineral from Scotland, the type of the species, has been found by the writer to be optically homogeneous; the most modern analysis of it, that by Tschermak,[3] may be used. The data are as follows (in all the analyses any K_2O is added to Na_2O, and Fe_2O_3 to Al_2O_3):

	ANALYSIS	RATIO	INT.	r/i	$r/av.$
Na_2O	4.01	0.065	1	0.0650	1.04×1
CaO	13.55	0.242	4	0.0605	0.97×4
Al_2O_3	31.63	0.310	5	0.0602	0.97×5
SiO_2	37.73	0.628	10	0.0628	1.01×10
H_2O	13.52	0.750	12	0.0625	1.04×12
Sum	100.44			av. 0.0622	

The mineral described by Gordon from Franklin Furnace, New Jersey, had optical properties identical with those of the preceding material, and so is undoubtedly also thomsonite. Its $CaO : Na_2O$ ratio and $Al_2O_3 : CaO + Na_2O$ ratio are both somewhat abnormal, (yielding a long line in Winchell's diagram) perhaps owing to the presence of a small amount of calcium carbonate in the sample analyzed; but the deviation from the formula of the type thomsonite is not great, as the tabulation shows:

	ANALYSIS	RATIO	INT.	r/i	$r/av.$
Na_2O	3.68	0.059	1	0.0590	0.94×1
CaO	15.94	0.284	4	0.0710	1.13×4
Al_2O_3	30.34	0.297	5	0.0594	0.95×5
SiO_2	36.44	0.606	10	0.0606	0.97×10
H_2O	13.52	0.750	12	0.0625	1.00×12
Sum	99.92			av. 0.0625	

[3] *Sitzb. Akad. Wien*, **126,** 545 (1917).

The thomsonite from Peekskill, N. Y., described by Phillips[4] shows indices agreeing with those of the two preceding minerals within the limits of accuracy of the immersion method, and its analysis gives:

	ANALYSIS	RATIO	INT.	r/i	$r/av.$
Na_2O	4.02	0.065	1	0.0650	1.04×1
CaO	13.80	0.246	4	0.0615	0.99×4
Al_2O_3	31.65	0.310	5	0.0620	0.99×5
SiO_2	37.44	0.623	10	0.0623	1.00×10
H_2O	13.22	0.734	12	0.0612	0.98×12
Sum	100.13			av. 0.0624	

It is concluded from these data that the formula of the mineral to which the name thomsonite was originally assigned, and of material from other localities agreeing with it in optical properties —$a = 1.525$, $\beta = 1.530$, $\gamma = 1.540$, all ± 0.005—is $Na_2O.4CaO.5Al_2O_3.10SiO_2.12H_2O$. This is the formula previously obtained by the writer by a somewhat different procedure. Winchell adopted twice this formula, in order to show apparent isomorphism, and then added one extra H_2O, on the basis of Gordon's plotting of the H_2O contents of thomsonite and related minerals. That plot is misleading, however, in that all the ratios given are referred to $Na_2O = 1$, which exaggerates deviations in the constituents with the higher ratios, and, moreover, the line is not drawn accurately through the center of the points plotted, so the indication as to water content of the analyses above worked out appears to the present writer more dependable. This is, namely, 12 H_2O to the single molecule, or 24 H_2O to the doubled one.

THE ALLEGED THOMSONITE SERIES

Many analyses are on record, of material which the analysts called thomsonite, in which both the silica and soda are higher than in thomsonite as above defined. This has been interpreted by several students as due to isomorphism of the valence-for-valence replacement type, and their conclusions are accordingly without significance; but Winchell has endeavored to account for it on the basis of volume-for-volume replacement, so that his results are worth further consideration. As the one who first

[4] *Am. Min.*, **9**, 240-241 (1924).

urged upon mineralogists the fundamental significance of the volume-for-volume type of isomorphism, the present writer should be glad to welcome any new illustrations of this relationship, and desires to state that in his opinion, if any isomorphism occurs in the thomsonite series, then Winchell's explanation of it may well be the correct one. He is, however, still awaiting optical evidence of the existence of such a series. The situation is not analogous to that of the plagioclases to which Winchell refers, for in it there is optical evidence of isomorphism.

The lowest refractive index of normal thomsonite never having been observed to go much below 1.520, whereas the highest indices of most other zeolites are decidedly less than this, a simple way to study the matter suggests itself, namely, to examine fragments immersed in a liquid with $n=1.518$. This the writer has done in the case of over twenty-five specimens labelled thomsonite in mineral collections, widely separated localities being represented. In every case except those above cited, more or less admixture with zeolites having all their indices below that of the liquid has been observed. In some instances, such as the mineral from Magnet Cove, from Kaaden, Bohemia, and from Monte Somma, Italy, the amount of the low-index zeolite was so small that their analyses might have been safely used in arriving at the formula of thomsonite proper. In other cases, as that from Minnesota, about half the grains had the high indices of thomsonite, and the remainder were a mineral of lower indices. In still others such as the well-known Table Mountain, Colorado, specimens, and the fibrous masses from Nova Scotia and from the Faroes, the bulk of the material had all three indices lower than the liquid. In some cases the low-index mineral could be definitely identified as mesolite or natrolite; in others it was a seemingly unrecognized species, the prior name for which, as pointed out in the writer's earlier paper, is faroelite. The important point is that material labelled thomsonite in collections (and analyzed under that name) is not uniform, but represents either thomsonite proper, faroelite, (a somewhat similar zeolite with all three indices lower than the lowest of thomsonite), or mechanical mixtures of these with one another or with other zeolites not related to thomsonite at all. There is no optical evidence whatever that any isomorphous series is represented.

The Composition of Faroelite

Unfortunately, only a single analysis of this mineral on material of entirely satisfactory optical character is available, namely, that made by Hillebrand (Doelter No. 50), the locality being Table Mountain, Colorado, and the specimens being preserved in the U.S. National Museum. The data are:

	Analysis	Ratio	Int.	r/i	$r/av.$
Na_2O	5.62	0.091	1	0.091	0.94×1
CaO	11.34	0.202	2	0.101	1.04×2
Al_2O_3	29.71	0.291	3	0.097	1.00×3
SiO_2	41.21	0.686	7	0.098	1.01×7
H_2O	12.20	0.677	7	0.097	1.00×7
Sum	100.08			av. 0.097	

As far as it goes, then, this analysis indicates faroelite to have the composition $Na_2O.2CaO.3Al_2O_3.7SiO_2.7H_2O$, or $Na_2Ca_2Al_6Si_7O_{26}.7H_2O$. This is a somewhat simpler formula than was previously suggested by the writer on the basis of averaging several analyses; however, as further optical study has indicated the presence of admixture in materials corresponding to the descriptions of the samples analyzed in most of these cases, this new result is probably nearer the true formula of the mineral, although of course further analyses on optically controlled material will be necessary before it can be regarded as settled. As previously stated, the refractive indices of this mineral are: $a = 1.512, \beta = 1.513, \gamma = 1.518$, all ± 0.005.

Re-interpretation of Winchell's Thomsonite Diagram

As the result of his fairly extensive optical studies, made on as many samples corresponding in description to those analyzed as has been practicable, the writer sees in Winchell's diagram (*op. cit.*, p. 95) *not* a continuous series, but three different things: thomsonite, in part with minor amounts of admixture, in the cluster of analysis-points lying below line 60; faroelite, likewise with admixtures, in the other close group of points between horizontal lines 60 and 70; and, in the widely outlying points, mechanical mixtures with large quantities of other zeolites, as for instance with natrolite in No. 61 (which lies on a line connecting thomsonite with natrolite), and with mesolite in No. 52 (in which material mesolite can be readily recognized optically).

SUMMARY AND CONCLUSIONS

Now that the immersion method for the optical study of minerals is available, no claim of isomorphism between two or more compounds should be accepted unless accompanied by optical evidence.   Extensive optical study of specimens labelled thomsonite in collections has failed to yield evidence of isomorphism, but instead has shown the presence of two distinct species, often more or less admixed with other zeolites. By applying the Schaller method of reducing the ratios derived from a mineral formula, and using only analyses on optically satisfactory material, it is shown that thomsonite proper, the mineral to which the name was originally assigned, has the formula $Na_2O.4CaO.5Al_2O_3.10SiO_2.12H_2O$.   Some specimens labelled thomsonite consist of faroelite, which may be $Na_2O.2CaO.3Al_2O_3.7SiO_2.7H_2O$, but needs further study.   Winchell's diagram, when considered in the light of the optical data, confirms the existence of these two species, and demonstrates graphically what had already been suspected from optical observations, that certain of the analyses in the literature ascribed to thomsonite have been made upon mechanical mixtures.   The alleged thomsonite series represents, in the writer's opinion, only pseudo-isomorphism.