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HABITS, CRYSTAL FORMS AND COMPOSITION OF THOMSONITE

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ABSTRACT

The variation in the chemical composition of thomsonite (Si/Al = 1.0 to 1.3) resembles a solid solution with mesolite (Si/Al = 1.5). Although these two zeolites are commonly associated, the most silica-rich thomsonite does not occur with mesolite. Blocky, complex crystals, occurring at Yellow Lake, British Columbia, as well as the type thomsonite from Old Kilpatrick, Scotland and the blocky crystals from Vesuvius, have Si/Al ratios near one. Bladed crystals of thomsonite occur at a number of localities and in a range of sizes. The Si/Al ratio ranges from about 1.05 for the coarsest blades to 1.13 for the finest. The third habit, the variety faroelite, occurs in the form of botryoidal growths and waxy balls, which are composed of extremely small crystals, and have the highest observed Si/Al ratios (1.2 to 1.3). The small crystal size, simple crystal forms and the disorder of the crystals suggest rapid, disequilibrium growth that has led to random replacement of AlO_4 with SiO_4 tetrahedra. This substitution causes the composition of such thomsonite to approach that of mesolite.

SOMMAIRE

Par la variation de sa composition chimique, la thomsonite (Si/Al = 1.0 à 1.3) ressemble à une solution solide avec mésolite (Si/Al = 1.5). Bien que ces deux zéolites se trouvent communément associées, la thomsonite la plus riche en silice n'est pas accompagnée de mésolite. Les cristaux complexes et trapus de Yellow Lake (Colombie britannique), les cristaux de la thomsonite type d'Old Kilpatrick (Écosse) et ceux du Vésuve ont tous un rapport Si/Al voisin de l'unité. Des cristaux de thomsonite en lames se trouvent dans nombre de localités et en une gamme de tailles. Le rapport Si/Al varie d'environ 1.05 pour les lames les plus épaisses à 1.13 pour les plus fines. La variété dite faroelite représente un troisième facies, sous forme d'agrégats botryoïdes et de boules cireuses, comprenant des cristaux extrêmement petits, pour lesquels le rapport Si/Al est le plus élevé que l'on ait observé (1.2 à 1.3). La petite taille de ces cristaux, leurs formes simples et les indices de désordre suggèrent une croissance rapide, en déséquilibre, qui se manifeste par le remplacement de tétraèdres AlO_4 par SiO_4. Cette substitution donne des thomsonites qui approchent de la mésolite en composition.

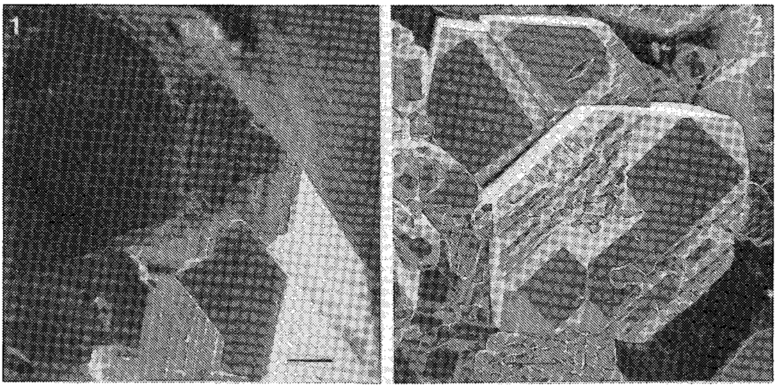
(Traduit par la Rédaction)

INTRODUCTION

With a few notable exceptions, most zeolite minerals have variable compositions. The cations, which are exchangeable, can be expected to show some variability, but the Si/Al ratio of the framework also commonly varies within definable limits. It is clear from experimental work in the synthesis of zeolites that silica activity in the growth medium plays a major role in the Si/Al ratio of the growing phase; for example, the Si/Al ratio in synthetic analcime is controlled by the composition of the glass from which it grew (Saha 1959).

The interpretation of natural zeolites is less direct. However, studies of the compositions of zeolites that have grown sequentially have led to the conclusion that most natural zeolites grow within a narrow range of silica activity, because the Si/Al ratios vary little within a single locality: *e.g.*, ferrierite–clinoptilolite (Wise & Tschernich 1976a) and offretite–erionite–levyne (Wise & Tschernich 1976b). The greater variability observed between localities is probably a result of different temperatures. Variation in the composition of thomsonite, however, does not appear to be explained by these mechanisms. For example, the Si/Al ratios of thomsonites in different habits at a single locality show some variation. A common association in the Pacific Northwest is thomsonite and mesolite. The thomsonite here as in world-wide occurrences shows a range of Si/Al ratios (from 1.0 to nearly 1.3), whereas Si/Al ratios of the associated mesolite are nearly constant at 1.5.

By 1845 it was known that thomsonite occurred in three different habits (Hey 1932). Long, radiating prisms rich in crystal forms, such as those found at Old Kilpatrick, Scotland,



FIGS. 1. Scanning electron micrograph of blocky thomsonite, Yellow Lake, British Columbia. Flat face (upper left) is early-growth *c* face that was stopped by a calcite cover. The complex faces in the lower part of the photograph are from later growth. Length of bar, 0.1 mm. 2. Scanning electron micrograph of the complex forms of blocky thomsonite, Yellow Lake, British Columbia. Length of bar, 0.1 mm.

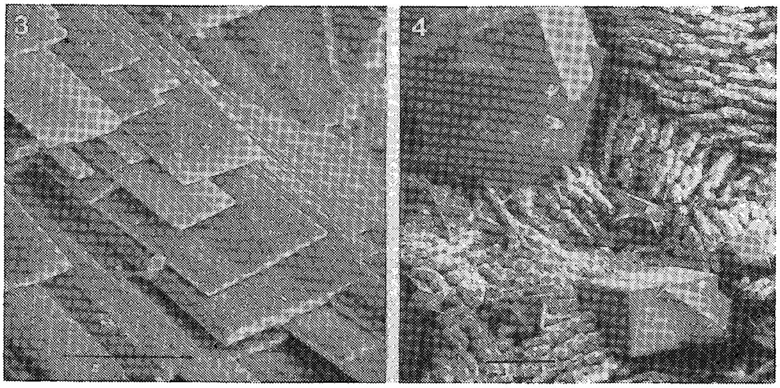
were those upon which the original species definitions were based. Radiating clusters of fibrous crystals, actually blade-shaped, such as those from County Antrim, Ireland, were later found to be similar physically and chemically. The waxy to fibrous botryoidal spherules, like those found at Nolso, Faroe Islands, and Goble, Oregon, were also later equated with thomsonite. It was also recognized in 1845 that the botryoidal form was the richest in silica, whereas the coarse blocky crystals were the poorest.

A detailed study of thomsonite habits, chemistry, crystal forms and association in various localities in the Pacific Northwest has been carried out. The purpose of this paper is to explain the observations that (1) the smallest crystals have the simplest forms, the highest silica content and the most disordered structure, and (2) the trend in chemical composition resembles a solid solution with mesolite, a commonly associated mineral.

HABITS AND CRYSTAL FORMS OF THOMSONITE

Blocky crystals with complex forms

Thomsonite from Yellow Lake near Olalla, southern British Columbia, forms stout to elongate, pseudotetragonal prismatic crystals.



FIGS. 3. Scanning electron micrograph of bladed thomsonite from Skookumchuck Dam, Washington. Crystals are bounded by simple forms with a few vicinal faces modifying broad *b* faces. Length of bar, 0.1 mm. 4. Scanning electron micrographs of chabazite crystals on a cavity lining of thomsonite, Beech Creek quarry, Grant Co., Oregon. Length of bar, 0.1 mm.

These crystals have grown in flattened vesicles in porphyritic trachyte flows. The thomsonite is characterized by a large number of different crystal forms (Tschernich & Wise, in prep.) and is the only example of this habit in the Pacific Northwest.

The Yellow Lake thomsonite is associated with brewsterite, mesolite, calcite and analcime, and has formed crystals ranging in width from about 0.5 to several millimetres. In a few instances the growth of thomsonite was interrupted by the crystallization of calcite. The earlier generation of thomsonite is composed of simple crystals bounded by smooth *a*, *b* and *m* prism faces and a smooth *c* face (Fig. 1). Thomsonite not covered by calcite continued to grow and developed complexly terminated crystals (Figs. 1 & 2) with such forms as {101}, {502}, {301}, {801} and {410}.

Bladed crystals

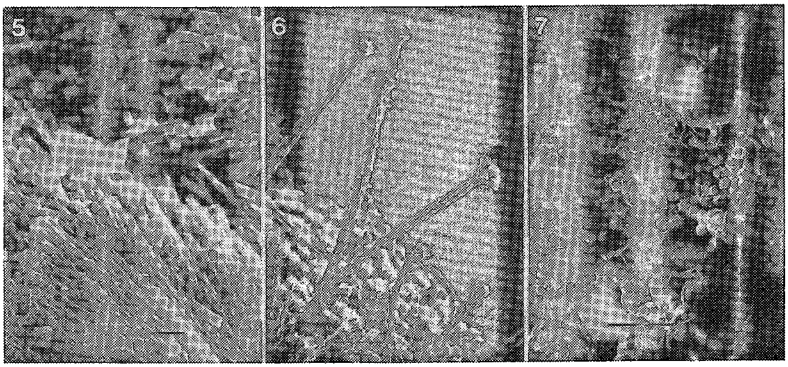
Thomsonite from many localities in the Pacific Northwest occurs as simple blades, flattened parallel to {010} and elongated parallel to *c*. In some growth sequences thomsonite occurs in two generations. It is commonly the first zeolite to crystallize but it also may grow again at a later stage. The crystals from both generations have similar sizes and habits and both are followed by mesolite crystallization.

Thomsonite from Drain, Oregon, Point Sal, California, and Skookumchuk Dam, Washington, are coarsely crystalline with crystals up to several millimetres in two dimensions, but seldom thicker than 0.2 mm. The smaller, bladed crystals are illustrated in Figure 3. The forms are simple: *a, b, m* and *c*. This habit appears to represent fairly rapid growth from abundant nuclei.

At Point Sal thomsonite formed early and was followed by calcite and analcime, and later by natrolite. At Drain, Oregon, analcime crystallized first, followed by thick cavity linings (several cm) of large, bladed crystals of thomsonite. These were followed by needles of mesolite, upon which are perched thomsonite blades. Thomsonite was both preceded and followed by mesolite at Skookumchuck Dam (Tschernich 1972), but the sample analyzed here was followed by mesolite.

Thomsonite forms fine-grained cavity linings of bladed crystals at several localities, but is particularly common at the Beech Creek quarry, Grant County, Oregon. These linings have a pale, blue-grey, opalescent appearance. The growth surface is dull and seems rough (Fig. 4); the broken surface reveals crystals less than 10 μm thick, but with lengths up to 500 μm. The crystals are generally aligned with the *c* axis perpendicular to the growth surface (Fig. 5). The crystals have the simplest form, only the *a, b* and *c* faces.

At Beech Creek there is an interesting variant of the simple cavity lining, in the form of long, composite rods with radiating sprays at their ends. These rods, which have lengths up to 1



FIGS. 5. Scanning electron micrograph of thomsonite crystals forming cavity lining, Beech Creek quarry, Grant Co., Oregon. Note the crystals have only *a, b* and *c* faces. A fracture through the lining forms the lower third of the photograph. Length of bar, 0.01 mm. 6. Scanning electron micrograph af thomsonite forming rod-like growths, possibly on mesolite needles, Beech Creek quarry, Grant Co., Oregon. Crystals are simple and similar to those forming the lining. Length of bar, 0.1 mm. 7. Scanning electron micrograph of thomsonite forming balls and irregular growths on clay cavity lining, Beech Creek quarry, Grant Co., Oregon. Length of bar, 0.1 mm.

mm are composed of simple crystals about 1 μm thick and 10 μm long (Fig. 6). The rods may have formed around a mesolite fibre, although we have not been able to unambiguously identify mesolite in the rods.

Botryoidal and irregular growths

Smooth, waxy balls and botryoidal growths occur at several localities in the Pacific Northwest, specifically at Beech Creek and Goble, Oregon. This habit is similar to the spherical concretion descriptions applied to the variety faroelite (Faroe Islands) and the variety lintonite (Grand Marais, Minnesota).

The balls at Beech Creek commonly have irregular root-shaped growths protruding from their surfaces (Fig. 7). The thomsonite crystals forming the balls and root-shaped growths are less than 0.1 μm thick, and are oriented with their *c* axis more or less perpendicular to the growth surfaces.

Some conclusions

Experimental work on crystal growth and crystal forms (Tiller 1969, Lofgren 1974) has verified some of the general conclusions drawn from various observations of crystal habits. Crystals with smooth faces and numerous forms have grown slowly from solutions with relatively low degrees of supersaturation (or undercooling). In contrast, crystals with dendritic forms have developed through rapid growth from highly supersaturated solutions. These latter crystals depart from equilibrium structures, and contain a much higher density of defects and disorder in the atomic arrangement.

The range of thomsonite habits and crystal sizes seems to reflect a range of rates of crystal growth. The extreme number of very small crystals with simple, unmodified forms, characteristic of the botryoidal habit, suggests that growth was rapid, although not truly dendritic. The simple and only slightly modified forms and the small thickness-to-length ratios of bladed crystals probably represents a less rapid growth rate than in the formation of the botryoidal habit. The slowest growths are represented by the morphologically complex, blocky crystals.

CHEMICAL COMPOSITIONS

Samples of the various habits of thomsonite were analyzed by EMX methods. Crystals and clusters of crystals were embedded in epoxy, and ground until the centre of the crystal or cluster was exposed. The mount was polished and coated with 250 Å of carbon. The samples were analyzed on an ARL electron microprobe, using an accelerating voltage of 15 kV for all elements with a sample current of 10 na. Standards used were labradorite (Ca and Al), K-feldspar (K and Si), albite (Na). Emission data were reduced with a modified version of EMPADR VII (Rucklidge & Gasparrini 1969). Several points on each sample were analyzed to test for compositional changes across the sample. The analyses, listed in Table 1, are representative of the group from each sample; all analyzed points are plotted in Figure 8 along with thomsonite analyses from the literature, principally from Hey (1932). The variation within a single crystal or crystal cluster is surprisingly small, whether along the length of a single blade or from the core to edge of a spherule.

TABLE 1. SELECTED ANALYSES, CELL CONTENTS, AND CELL DIMENSIONS OF VARIOUS HABITS OF THOMSONITE FROM THE PACIFIC NORTHWEST

	1	2	3	4	5	6	7
	Blocky crystals	Fine blades	Coarse blades	Coarse blades	Fine blades	Botryoidal growths	Waxy balls
	Yellow Lake, B.C.	Spray, Ore.	Skookum-chuck' Dam Wash.	Drain, Ore.	Beech Creek, Ore.	Beech Creek, Ore.	Goble, Ore.
SiO_2	35.55	37.66	38.64	40.30	40.38	39.95	41.49
Al_2O_3	30.67	30.54	30.19	29.94	30.31	28.65	28.59
CaO	9.60	12.84	11.28	12.91	12.16	11.82	11.89
SrO	6.52	0	0	0	0	0	0
Na_2O	3.86	4.10	5.36	4.16	4.34	4.12	4.23
K_2O	0	.04	0	0	0	0	0
Total	86.20	85.18	85.47	87.31	87.19	84.54	86.20
Remainder is H_2O; Fe, Mg, Ba were sought but not detected.							
Cell contents with 40 oxygen							
Si	9.93	10.24	10,45	10.61	10.65	10.85	11.04
Al	10.10	9.79	9.62	9.29	9.43	9.18	8.96
Ca	2.87	3.74	3.27	3.64	3.44	3.44	3.39
Sr	1.06	--	--	--	--	--	--
Na	2.09	2.16	2.81	2.12	2.22	2.17	2.18
K	--	.01	--	--	--	--	--
Si/Al	.98	1.05	1.09	1.14	1.13	1.18	1.23
Cell dimensions in (Å) all values ±0.01							
a	13.06[1]	--[2]	13.09[3]	13.09[3]	--[2]	--[2]	13.07[3]
b	13.12	--	13.09	13.09	--	--	13.07
c	13.23	--	13.20	13.20	--	--	13.14

1/ *From Weissenberg and rotation photographs.*
2/ *Insufficient material for x-ray powder pattern.*
3/ *Calculated from 240 and 204 peaks on diffractometer patterns; there is no distinction between 240 and 420, and therefore, a=b.*

The idealized composition of thomsonite is $NaCa_2Al_5Si_5O_{20}\cdot 6H_2O$, in which Ca/(Ca+Na) is 0.67 and Si/(Si+Al) is 0.5. Most of the plotted points in Figure 8 trend from the ideal composition toward that of mesolite (ideally $Na_2Ca_2Al_6Si_9O_{30}\cdot 8H_2O$).

There is a close correspondence between the Si/Al content of the thomsonite and crystal size for most of the samples. The large, stout prisms

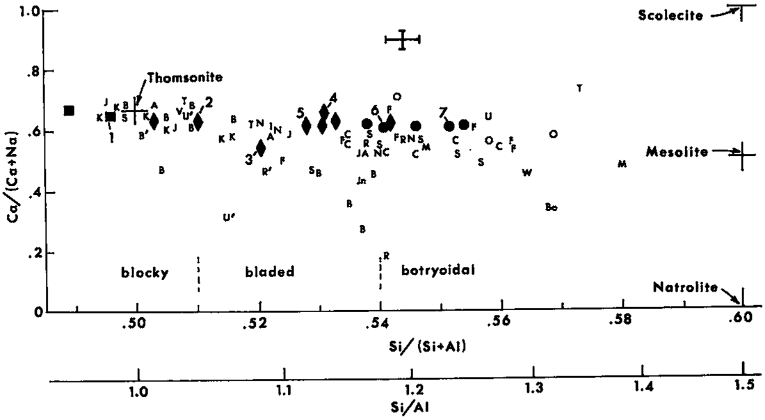
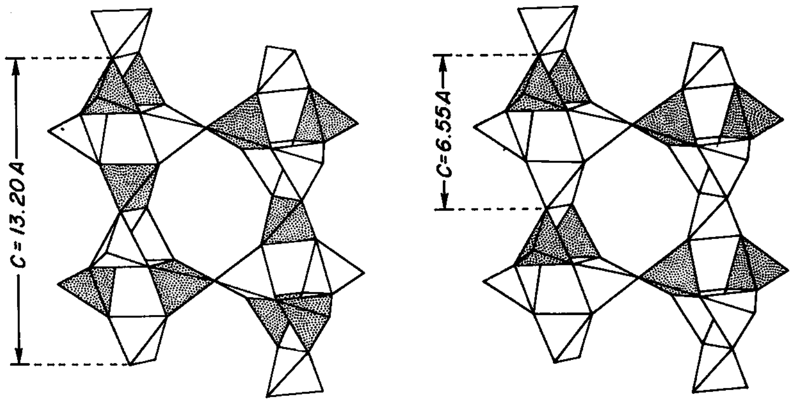


FIG. 8. Compositional plot of thomsonite in various habits. Chemical data obtained in this study on blocky, bladed and bortyodial thomsonite are represented by squares, diamonds and circles, respectively. The numbers refer to analyses in Table 1. Other data points from Hey (1932) with localities indicated by letters: Old Kilpatrick, Scotland *K*, other Scottish localities *S*, Bohemia *B*, West Patterson, New Jersey *J*, Arkansas *A*, Table Mountain, Colorado *C*, Grand Marais, Minnesota *M*, Goble and Ritter Hot Springs, Oregon *O*, Nova Scotia *N*, County Antrim, Ireland *R*, Faroe Islands *F*, Vesuvius, Italy *V*, other Italian localities *T*, Bombay *Bo*, various localities in U.S.S.R. *U*, and New South Wales, Australia *W*. A few other analyses that further define the compositional range are from Yefimov *et al.* (1966) *U′*, Antonin (1942) *B′*, Tomkeieff (1934) *R′*, and Harada *et al.* (1969) *Jn*. The approximate compositional ranges of the three habits are divided by dashed lines. The error bars indicate 1% variations expected in the best analyses.

(Yellow Lake) have compositions close to the ideal formula. The bladed crystals tend to be more siliceous, and botryoidal crystals (Beech Creek, Faroe Islands and Minnesota) have the highest Si contents. This correspondence suggests that there is a genetic relationship between crystal size and silica content. It seems likely that rapid growth gives rise to very small crystals and high silica contents. There is also a parallel relationship between Si content and



THOMSONITE

$NaCa_2Al_5Si_5O_{20}\cdot 6H_2O$

MESOLITE

$Na_2Ca_2Al_6Si_9O_{30}\cdot 8H_2O$

FIG. 9. Comparison of the framework structures of thomsonite and mesolite (Fischer & Meier 1965). Stippling represents Al tetrahedra.

complexity of the forms, with the simpler crystals having the higher Si/Al ratios. At Yellow Lake the earlier crystals with simple forms (Fig. 1) have a slightly higher Si/Al ratio than the part that grew last with complex forms.

Ideal thomsonite with equal amounts of Al and Si develops an ordered framework structure in which SiO_4 and AlO_4 tetrahedra alternate, leading to a *c* dimension twice that of mesolite (Fig. 9). The aluminum avoidance principle of Goldsmith & Laves (1955) states that Al–O–Al bonds are much less stable than Si–O–Si bonds, and therefore tend not to occur. As mentioned earlier, structural disorder is characteristic of crystals that have grown rapidly. Therefore, disorder in the thomsonite framework is more likely to involve random substitution of Si for Al in the tetrahedral sites rather than randomizing the AlO_4 and SiO_4 tetrahedra, which must generate abundant Al–O–Al bonds.

X-RAY EVIDENCE OF DISORDER

X-ray powder patterns of thomsonite in the various habits show small but consistent differences. The poorer crystallinity of the botryoidal thomsonite is reflected in somewhat broader peaks. For example, the 420 and 204 reflections are easily resolvable ($2\theta(CuK\alpha) = 30.54°$ and $30.33°$ respectively) in the bladed or blocky crystals, but are poorly resolved in patterns of botryoidal thomsonite. The *c* dimension, calculated from the 204 peak, decreases from about 13.23 Å for Si-poor thomsonite to about 13.15 Å for the more siliceous ones (Goble sample, #7, Table 1).

Because ordering gives rise to a doubled *c* dimension (Fig. 9), disordering will cause *hkl* reflections having *l* odd to diminish in intensity. For example, the diffractometer peak at 20.4°, composed of reflections from the 221, 122, and 212 planes, has less than half the intensity (relative to 200) in the botryoidal thomsonite than in the blocky crystals. This intensity loss is primarily a result of the intensity of the 221 reflection reducing nearly to zero.

CONCLUSIONS

The evidence presented indicates that the botryoidal and spherical habits of thomsonite are composed of very small, morphologically simple crystals. Moreover, these same crystals have higher Si contents than the larger crystals and show disorder effects in X-ray diffractometer patterns. Increases in silica contents of many zeolites are commonly a result of high silica activities in the solution from which they have grown. However, it is unlikely that a high silica activity in the solutions causes thomsonite crystals to be small, because thomsonite in this habit is not followed by a zeolite richer in Si, such as mesolite. Mesolite forms with or following thomsonite in many localities, but in these cases the thomsonite is either in the form of somewhat larger blades or very much larger blocky, complex crystals.

We conclude, therefore, that the rapidly grown thomsonite, in small disordered crystals, accepts excess silica as a result of nonequilibrium growth. This excess silica entering the chain structure causes the Si/Al ratio to approach that of mesolite. It is probable that crystallization of this high-Si thomsonite lowers the silica activity of the solutions, thus inhibiting mesolite formation.

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