

TETRANATROLITE FROM MONT ST-HILAIRE, QUÉBEC

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ABSTRACT

Tetranatrolite is a new name for "tetragonal natrolite", a mineral first reported from Ilimaussaq, Greenland by Andersen *et al.* (1969). It is a widespread constituent in the pegmatitic dykes andmiaroles in the nepheline syenite at Mont St-Hilaire, Québec, occurring most commonly as epitactict overgrowths on natrolite crystals and as small euhedral crystals or silky fibrous sprays on natrolite, analcime or microcline. It occurs less commonly as irregular earthy patches on natrolite or analcime. The mineral is white, translucent to opaque with vitreous to dull lustre. The hardness could not be determined, and cleavage was not observed. The mineral is readily soluble in 1:1 HCl, slowly in 1:1 HNO₃ and is only slightly attacked by 1:1 H₂SO₄. Optically the mineral is uniaxial positive, ω 1.481 and ϵ 1.496. Single-crystal photographs show the mineral to be tetragonal, $I4_2d$ or $I4_1md$, a 13.098(2), c 6.635(2) Å. With D_{meas} 2.276 g/cm³, we find $Z = 4$, whence $D_{\text{calc}} = 2.234$ g/cm³. Strongest lines in the powder X-ray-diffraction pattern are 6.549(5)(200), 5.912(4)(101), 4.635(4)(220), 4.387(5)(211), 4.143(4)(310), 3.189(5)(321), 2.867(10)(411), 2.438(4)(431), 1.816(4)(640,413), 1.721(4)(730). Electron-microprobe analysis gave: SiO₂ 46.9, TiO₂ 0.06, Al₂O₃ 25.6, CaO 1.48, MgO n.d., FeO 0.11, Na₂O 14.0, K₂O 1.12, H₂O⁺ (TGA to 1000°C), 9.59, H₂O⁻ (TGA to 135°C) 1.31, sum 100.18 (by weight). The analysis corresponds to (Na_{1.75}Ca_{0.10}K_{0.09})Fe_{0.01}Al_{1.95}Si_{3.02}O₁₀•2.06H₂O or, ideally, Na₂Al₂Si₃O₁₀•2H₂O. A TGA curve shows a minor weight-loss between room temperature and 135°C and a major weight-loss between 135 and 435°C. The infrared spectrum of the mineral strongly suggests (Si,Al) disorder. Hydrothermally the mineral decomposes to nepheline and analcime at 445°C and 1.0 kbar, and to analcime at 330°C and 0.5 kbar. No change was observed at 265°C and 0.5 kbar. The mineral is a dehydration product of a higher hydrate.

Keywords: tetranatrolite, St-Hilaire, Québec, natrolite, pegmatite, epitactict overgrowth, Si,Al disorder, nepheline syenite.

SOMMAIRE

Tetranatrolite est un synonyme de "natrolite tétragonale", nom sous lequel un minéral fut décrit par Anderson *et al.* (1969) qui l'avaient trouvé à Ilimaussaq (Groënland). Ce minéral est un constituant très répandu dans les dykes pegmatitiques et dans les cavités miarolitiques de la syénite à néphéline du mont St-Hilaire, dans la province de Québec. Il se présente le plus communément sous forme d'enduits épitactiques sur cristaux de natrolite ou de petits cristaux idiomorphes ou encore d'agrégats de fibres soyeux sur natrolite, analcime ou microcline. Moins souvent, on le trouve en taches terreuses irrégulières sur natrolite ou analcime. C'est un minéral blanc, translucide à opaque, d'éclat vitreux à terne, dont la dureté n'a pu être déterminée et où nul clivage n'a été décelé. Il est facilement soluble dans 1:1 HCl, difficilement dans 1:1 HNO₃ et à peine attaqué par 1:1 H₂SO₄. Optiquement, il est uniaxe positif, avec ω 1.481 et ϵ 1.496. Les clichés de cristal unique révèlent sa symétrie tétragonale, $I4_2d$ ou $I4_1md$, a 13.098(2), c 6.635(2) Å. Pour une densité mesurée de 2.276, on trouve $Z = 4$, d'où la densité calculée de 2.234. Les raies les plus intenses du diagramme de poudre sont les suivantes: 6.549(5)(200), 5.912(4)(101), 4.635(4)(220), 4.387(5)(211), 4.143(4)(310), 3.189(5)(321), 2.867(10)(411), 2.438(4)(431), 1.816(4)(640,413), 1.721(4)(730). L'analyse à la microsonde électronique donne: SiO₂ 46.9, TiO₂ 0.06, Al₂O₃ 25.6, CaO 1.48, MgO n.d., FeO 0.11, Na₂O 14.0, K₂O 1.12, H₂O⁺ (ATG jusqu'à 1000°C) 9.59, H₂O⁻ (ATG jusqu'à 135°C) 1.31, somme 100.18 (en poids). Cette analyse donne la formule (Na_{1.75}Ca_{0.10}K_{0.09})Fe_{0.01}Al_{1.95}Si_{3.02}O₁₀•2.06H₂O ou idéalement, Na₂Al₂Si₃O₁₀•2H₂O. La courbe d'ATG montre une faible perte de poids entre la température ambiante et 135°C et une perte plus forte de 135 à 435°C. Le spectre infrarouge indique un désordre (Si,Al). La tétranatrolite subit une décomposition hydrothermale en néphéline et analcime (445°C, 1.0 kbar), ou en analcime seulement (330°C, 0.5 kbar); à 265°C et sous 0.5 kbar, on n'observe aucun changement. Ce minéral provient de la déshydratation d'un hydrate supérieur.

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Mots-clés: tétranatrolite, St-Hilaire, Québec, natrolite, pegmatite, enduit épitactique, désordre, Si,Al, syenite néphélinique.

INTRODUCTION

A tetragonal analogue of natrolite was first found in the albitite veins cutting the naujaite inclusions in lujavrite in the westernmost part of Tugtup Agtakorfia, Greenland, and was described by Andersen *et al.* (1969). The mineral was found as aggregates of sheaf-like groups of small prismatic crystals, each of which displayed a single prism and either a pinacoid or a dipyrmaid. The mineral was reported as commonly associated with aegirine, analcime, natrolite, sodalite, epistolite, schizolite, lepidolite, sphalerite and a number of beryllium minerals.

The X-ray powder-diffraction pattern of the mineral was similar to that of natrolite but could be indexed on the basis of a body-centred tetragonal cell with a 13.043, c 6.619 Å. The optical properties of the mineral were also found to be consistent with those for a uniaxial mineral. Andersen *et al.* made no attempt to name this mineral at that time.

In studies of the minerals from Mont St-Hilaire, Québec, it has been frequently noted that crystals of a variety of natrolite, usually well formed and transparent when first collected, alter after short exposure to air, producing a white, translucent to opaque, friable and somewhat porous crust. The rate of alteration

is apparently affected by the humidity of the immediate environment. The well-defined boundaries between the altered crust and the transparent core material are either smooth or irregular surfaces, apparently without transitional zones (Figs. 1, 2). When the crust material is removed, the freshly exposed transparent core material does not show sign of further alteration during a period of more than a year under similar conditions. The freshly collected material may be preserved in water, but the altered crust material does not recover its transparency after being immersed in water for several months.

Studies of this alteration material from Mont St-Hilaire by the present authors show that it is a tetragonal dimorph of natrolite, similar in composition and physical properties to the tetragonal natrolite from Greenland reported by Andersen *et al.* (1969). The name *tetranatrolite* was proposed for this mineral jointly by the present authors with Dr. Andersen and his associates. The mineral and its name were approved by the Commission on New Minerals and Mineral Names, I.M.A. Type specimens from Mont St-Hilaire are preserved at the National Museum of Natural Sciences, Ottawa (37131) and at the Royal Ontario Museum, Toronto (M35545). A specimen of the mineral

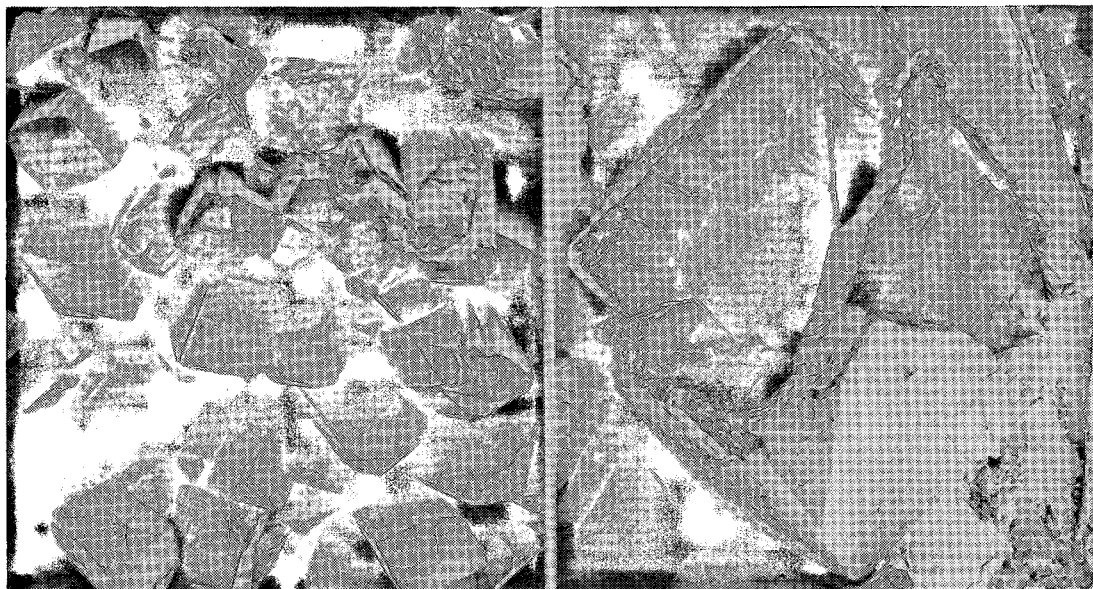


FIG. 1. Tetranatrolite overgrowths on natrolite crystals (3 mm across) from Mont St-Hilaire.

FIG. 2. Tetranatrolite overgrowth on a natrolite crystal with a portion of tetranatrolite removed, showing sharp boundaries. Cross-section of crystal: 3 mm.

from Ilmaussaq, Greenland, an ideotype, is also deposited with the Royal Ontario Museum (M36160).

OCCURRENCE AND PHYSICAL PROPERTIES

Tetranatrolite is a widespread constituent in the pegmatite dykes and miarolitic cavities in the nepheline syenite at Mont St-Hilaire, and is closely associated with natrolite, microcline, analcime, aegirine and a variety of accessory minerals. The mineral occurs most commonly as a friable crust up to 1 mm thick on natrolite crystals of various sizes (Fig. 1) and as small euhedral crystals (0.1 mm) or fine, silky fibrous sprays on larger crystals of natrolite, analcime and microcline. It occurs less commonly as irregular, earthy patches on massive natrolite and analcime. Tetranatrolite-coated natrolite crystals are very simple in habit, as is typical of natrolite crystals from this locality, displaying prism {110}, pyramid {111}, with or without a small pinacoid {100}. Re-entrant angles in the prismatic zone, indicating multiplicity of the underlying natrolite crystals, are very common.

The mineral is white, translucent to opaque, with a vitreous to dull lustre. Owing to its peculiar mode of occurrence, the hardness of the mineral could not be determined, and cleavage was not observed. Several determinations of the density of the mineral by the flotation method gave 2.276(5) g/cm³, slightly higher than 2.254(5) g/cm³, the density of the closely associated natrolite, and 2.210(2) g/cm³, the density of tetranatrolite from Greenland (Andersen *et al.* 1969). Tetranatrolite dissolves

readily in 1:1 HCl and slowly in 1:1 HNO₃ with gelatinous residues; it is only slightly attacked by 1:1 H₂SO₄.

The optical properties of tetranatrolite and the associated natrolite, summarized in Table 1, were obtained from crystal fragments previously oriented by X-ray goniometry using a spindle stage and sodium light. All refractive-index liquids used were checked using an Abbé refractometer. The maximum and minimum indices of refraction for tetranatrolite from Mont St-Hilaire are slightly higher than those for the material from Greenland and are significantly higher than those for natrolite from Mont St-Hilaire and the Green River Formation, Colorado (Table 1). Composite tetranatrolite-natrolite crystals show optical continuity in thin section, with the ϵ direction of tetranatrolite parallel to the Z principal vibration direction of natrolite, suggesting an epitactic relationship between the two minerals.

X-RAY STUDIES

Both the tetranatrolite crust and the associated natrolite core were studied by X-ray diffraction, using Weissenberg, precession and powder methods. From the diffraction symmetry and systematic extinctions observed on single-crystal photographs the space group for tetranatrolite was established to be $I\bar{4}2d$ or $I4_1md$, thus confirming the space group $I\bar{4}2d$ for the material from Greenland, deduced on the basis of systematic extinctions in the indexed powder-diffraction data (Andersen *et al.* 1969). The systematic extinctions observed for natrolite were

TABLE 1. PROPERTIES OF TETRANATROLITE AND NATROLITE

	Tetranatrolite			Natrolite	
	(1)	(2)	(3)	(4)	(5)
Space group	$I\bar{4}2d$		$I\bar{4}2d$	$Fdd2$	$Fdd2$
a (Å)	13.098(2)	13.047(2)	13.043(1)	18.308(7)	18.285(8)
b				18.632(8)	18.636(8)
c	6.635(2)	6.614(2)	6.619(1)	6.589(3)	6.586(3)
Vol. (Å ³)	1138.28	1125.86	1126.02	2x1123.80	2x1122.12
Z	4	4	4	8	8
w or α	1.481(1)		1.480(1)	1.478(1)	1.4775(10)
β				1.480(1)	1.4810(10)
e or γ	1.496(1)		1.493(1)	1.492(1)	1.4895(10)
Optic sign	(+)		(+)	(+)2V=59°	(+)2V=62°
D _{obs} (g/cm ³)	2.276(5)		2.210(2)	2.254(5)	2.25(1)
D _{calc}	2.234	2.256		2.254	

- (1) Mont St-Hilaire, Québec (this study). (2) Ilmaussaq, Greenland (this study).
 (3) Ilmaussaq, Greenland (Andersen *et al.* 1969). Refractive indices by double variation.
 (4) Mont St-Hilaire, Québec (this study). (5) Green River Formation, Colorado (Pabst 1971). Refractive indices for 589 μ m, 2V measured directly in white light.

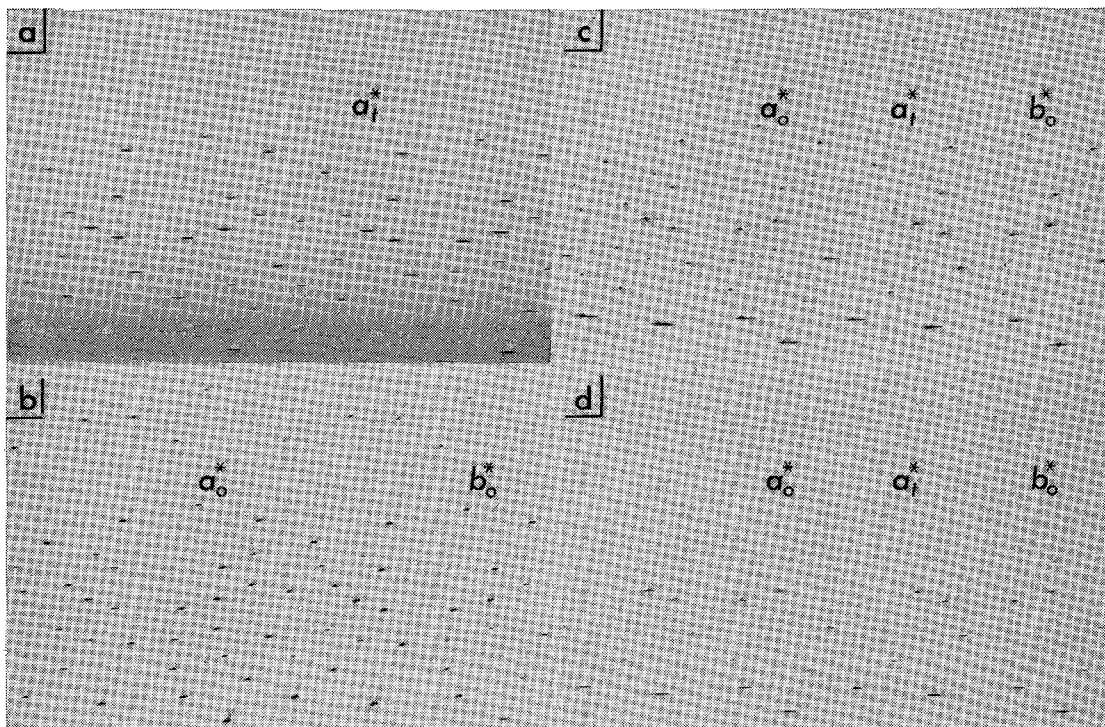


FIG. 3. C-axis Weissenberg photographs of (a) tetranatrolite, (b) natrolite, (c) twinned natrolite and (d) composite tetranatrolite-natrolite crystal.

consistent with the space group $Fdd2$ assigned to natrolite by other authors (Meier 1960, Pabst 1971). Cell parameters for tetranatrolite and natrolite from Mont St-Hilaire (Table 1) were obtained from single-crystal photographs and refined by a least-squares method using powder-diffraction data. Single-crystal photographs were used as a guide for indexing.

The single-crystal-diffraction photographs of tetranatrolite (Fig. 3a) are similar to those of natrolite, which is orthorhombic but pseudotetragonal in both cell geometry and X-ray diffraction intensities (Fig. 3b). The similarity is best shown by the Weissenberg photograph (Fig. 3d) of a composite tetranatrolite-natrolite crystal fragment. In this photograph the epitaxial nature of the overgrowth of tetranatrolite on natrolite is also evident. The single-crystal-X-ray reflections of tetranatrolite tend to be elongate and somewhat diffuse; the diffuseness is more pronounced in the high 2θ region.

Twinning is very common in natrolite from Mont St-Hilaire, as suggested by the frequent observations of re-entrant angles on the crystals (Fig. 1). The twinning in the Mont St-Hilaire natrolite may be explained by (110) as the

twin plane, or by [001] as the twin axis of 90° rotation. Both types of twinning have been observed for natrolite from other localities (e.g. Hey 1932, Pabst 1971). Theoretically, the two types of twinning can be distinguished by single-crystal X-ray diffraction. Twinning by (110) as the twin plane brings a and b of one individual to coincide approximately (deviation 1°) with b and a of the other, whereas twinning by $[001]_{90^\circ}$ produces exact coincidence of a and b of the two individuals. However, the poor quality of the X-ray photographs of the twinned crystals from Mont St-Hilaire (Fig. 3c) does not permit an unequivocal determination of the twin law. Owing to the very small difference between a and b , twinning by either twin law produces doubling of reflections along the 2θ axis as is shown in Figure 3c. The maximum splitting occurs along a^* and b^* and minimum splitting along the row line $[110]^*$. The absence of the doubling of reflections in the photographs of single crystals of tetranatrolite (Fig. 3a) eliminates the possibility that the tetragonal symmetry might be caused by twinning of orthorhombic natrolite.

The X-ray powder-diffraction pattern of tetra-

TABLE 2. X-RAY POWDER DIFFRACTION DATA FOR TETRANATROLITE AND NATROLITE FROM MONT ST-HILAIRE, QUÉBEC (1)

hkl	Tetranatrolite				Natrolite			
	Mont. St. Hilaire		Greenland (2)		Mont. St. Hilaire		Greenland (2)	
	d _{calc} Å	d _{obs} Å	I _{obs}	d _{calc} Å	d _{obs} Å	I _{obs}	d _{calc} Å	d _{obs} Å
200	6.549	6.549	5	6.530	10	6.527	7	220
101	5.919	5.912	4	5.902	20	5.880	7	111
220	4.631	4.635	4	4.612	5	4.660	3	040
211	4.391	4.387	5	4.377	10	4.579	2	400
310	4.142	4.143	4	4.124	5	4.366	6	{131
						4.150	2	240
						4.104	2	420
						3.262	1	440
						3.194	4	151
						3.152	4	511
321	3.186	3.189	5	3.171	10	3.102	2	{022
112	3.123	3.120	1	3.114	5	2.940	4	{202
202	2.959	2.960	2	2.949	5	2.899	1	{222
420	2.929	2.926	1	2.914	1	2.862	10	{260
						2.843	10	{620
411	2.865	2.867	10	2.851	20	2.862	10	{351
						2.843	10	{531
312	2.589	2.588	1	2.580	5	2.575	2	{242
						2.445	3	{422
						2.410	3	{460
431	2.436	2.438	4	2.425	5	2.445	3	{171
						2.410	3	{711
402	2.330	2.329	1	2.322	1	2.323	1	{080
						2.288	1	{800
440	2.315	2.312	1			2.288	1	{371
						2.258	1	{082
332	2.260	2.256	1	2.250	1	2.236	1	{280
						2.196	2	{602
						2.174	3	{262
422	2.196	2.195	3	2.187	3	2.174	3	{622
600	2.183	2.183	1			2.075	1	{160
						2.057	1	{480
213	2.069	2.070	1	2.061	2	2.057	1	{133
						1.962	1	{313
303	1.973	1.973	1			1.962	1	{840
541	1.955	1.954	1			1.877	2	{333
323	1.889	1.887	1	1.881	3	1.877	2	{191
						1.825	2	{153
640	1.816	1.816	4	1.807	5	1.825	2	{513
413	1.815					1.806	1	{210.0
						1.795	2	{353
701	1.800	1.800	1	1.792	1	1.795	2	{533
						1.758	1	{10.2.0
622	1.757	1.757	1	1.749	2	1.758	1	{771
						1.741	1	{482
						1.726	2	{842
721	1.736	1.737	1			1.741	1	{591
						1.703	2	{951
730	1.720	1.721	4			1.680	1	{4.10.0
						1.680	1	{10.4.0
								{553

(1) CuKα radiation (λ = 1.5418 Å), 114.6 mm camera, visual intensities, Si standard. Indices for tetranatrolite are based on the $\sqrt{2}d$ cell, and for natrolite, the $Fd\bar{2}d$ cell. Indices of natrolite and tetranatrolite may be compared directly if the indices for tetranatrolite are transposed to those for the F-cell by the matrix $\begin{pmatrix} 1 & 0 \\ 0 & 1/10 \end{pmatrix}$.

(2) Andersen et al. 1969. CuKα radiation (λ = 1.5418 Å), quartz internal standard, Guinier-Hägg camera. In the original data the line at 2.425 Å was indexed as 501, and the line at 2.061 Å indexed as 620. Data obtained by the present authors on the material from Greenland are essentially identical to those quoted here.

natrolite is essentially identical to that of tetranatrolite from Greenland (Andersen et al. 1969) and is very similar to the pattern of natrolite (Table 2). The diffraction lines 220, 310, 321, 411, 431, 422, 622 and 730 are characteristic of tetranatrolite, as the corresponding lines for natrolite are resolved into doublets resulting from the splitting of hkl and khl pairs. The splittings described may be observed on carefully prepared films using 114.6 mm Debye-Scherrer cameras. A focusing camera may bring

out the splitting of other less well resolved pairs (Anderson et al. 1969, Plate 1).

CHEMICAL COMPOSITION

Electron-microprobe analyses of eleven composite tetranatrolite-natrolite grains were performed at 15 kV with a specimen current of about 50 nA. Standards used were pyroxene (for Ca, Si), jadeite (Na, Al), ilmenite (Fe, Ti) and synthetic phlogopite (K, Mg). The water content was determined by TGA to 1000°C. The averaged analyses (Table 3) may be recalculated, on the basis of 10 oxygen atoms per formula, to $(Na_{1.75}Ca_{0.10}K_{0.00})Fe_{0.01}Al_{1.95}Si_{3.02}O_{10} \cdot 2.06H_2O$ for tetranatrolite and to $(Na_{2.00}Ca_{0.01})Al_{1.00}Si_{3.06}O_{10} \cdot 2.08H_2O$ for natrolite. H_2O^- (weight loss below 135°C) in tetranatrolite was considered to be adsorbed water; it was not used in the calculation. With $Z = 4$ for the body-centred cell, the calculated density of tetranatrolite is 2.234 g/cm³, which is slightly lower than the measured density 2.276 g/cm³. However, if H_2O^- is included in the calculation, the calculated density becomes 2.263 g/cm³.

The empirical formulae for tetranatrolite and the associated natrolite are both very close to the ideal formula $Na_2Al_2Si_3O_{10} \cdot 2H_2O$. Tetranatrolite, however, is relatively enriched in K and Ca with respect to natrolite. The enrichment of Ca and K is probably characteristic of tetranatrolite. A survey of the 27 analyses of natrolite

TABLE 3. ANALYSES OF TETRANATROLITE AND NATROLITE

	(1)	(2)	(3)
S10 ₂	46.9	48.81	47.8
T10 ₂	0.06	n.d.	0.05
Al ₂ O ₃	25.6	25.83	25.2
CaO	1.48	0.69	0.10
MgO	n.d.*	n.d.	n.d.*
FeO	0.11	tr	0.04
Na ₂ O	14.0	14.60	16.1
K ₂ O	1.12	tr	0.05
H ₂ O ⁺	9.59	10.70	9.73
H ₂ O ⁻	1.31		
Total	100.17	100.63	99.07

(1) Tetranatrolite and (3) natrolite associated with (1), Mont St-Hilaire, Québec. Analyses by electron microprobe, total Fe as FeO, H₂O by TGA to 1000°C. BaO and SrO were looked for but not detected.

(2) Tetranatrolite, Ilfmaussaq, Greenland (Andersen et al. 1969).

* not detected.

compiled by Foster (1965) shows that only two samples (analyses 25 and 26) contain more than 2.0 wt. % combined CaO and K₂O.

SI/AL DISORDER AND INFRARED SPECTRA

It has been shown that infrared spectra are sensitive to the degree of Si/Al order or disorder, as exemplified by the infrared studies on the feldspars (Laves & Hafner 1956, Hafner & Laves 1957, Thompson & Wadsworth 1957, Martin 1970) and cordierite (Langer & Schreyer 1969). The main effects in the infrared spectra characteristic of Si/Al disorder in the feldspars and cordierite were found to be (1) a reduction in the multiplicity of bands due to the increase of symmetry and (2) a considerable increase in half-widths (*i.e.*, broadening) of the bands owing to the decreasing similarity of the close-neighbor relationship of Si and Al atoms (Laves & Hafner 1962).

Tetranatrolite is presumably a disordered modification of natrolite. In the structure of natrolite (Meier 1960) there are three symmetrically nonequivalent tetrahedral atoms Si(I), Si(II) and Al, the site symmetries of which are C₂, C₁ and C₁ respectively. A tetragonal structure with *I42d* symmetry may be derived from natrolite by disordering Si(II) and Al. Si(I), however, is not required by symmetry to participate in the disorder. In the disordered structure there are only two symmetry-nonequivalent atoms, Si(I) and (Si,Al), with site symmetries S₄ and C₁, respectively. Thus, pronounced differences would be expected between

the infrared spectra of the ordered natrolite and the presumably disordered tetranatrolite, similar to those differences observed for the ordered and disordered feldspars and cordierite.

Infrared spectra of natrolite and tetranatrolite from Mont St-Hilaire (Fig. 4) were obtained using a Beckman 4250 spectrometer and KBr pellets containing 2.0 mg of sample dried at 110°C for 12 hours. The spectrum of natrolite from Mont St-Hilaire is comparable in fine details to the spectra of natrolite from Puy-de-Dôme (France), Poona (India) and Bohemia (Czechoslovakia) given in van der Marel & Beutelspacher (1976, p. 294-295). The spectrum of tetranatrolite shows a general resemblance only to that of natrolite and is characterized by broad, more spread-out groups of bands. The broadening of bands is particularly pronounced in the OH-stretching (3200–3600 cm⁻¹) and the T–O-stretching (900–1100 cm⁻¹) regions. It also shows a decrease in multiplicity of bands compared with the spectrum of natrolite. The effect is more pronounced in the T–O-stretching and T–O-bending (450–620 cm⁻¹) regions. The infrared spectrum of tetranatrolite is, therefore, consistent with what is expected of an Al/Si disordered structural model. The minor shift of the bands toward lower frequencies in the tetranatrolite spectrum is due, at least in part, to a slightly higher Al:Si ratio (Flanigen *et al.* 1971).

TGA AND HYDROTHERMAL STUDIES

The thermogravimetric analysis of tetra-

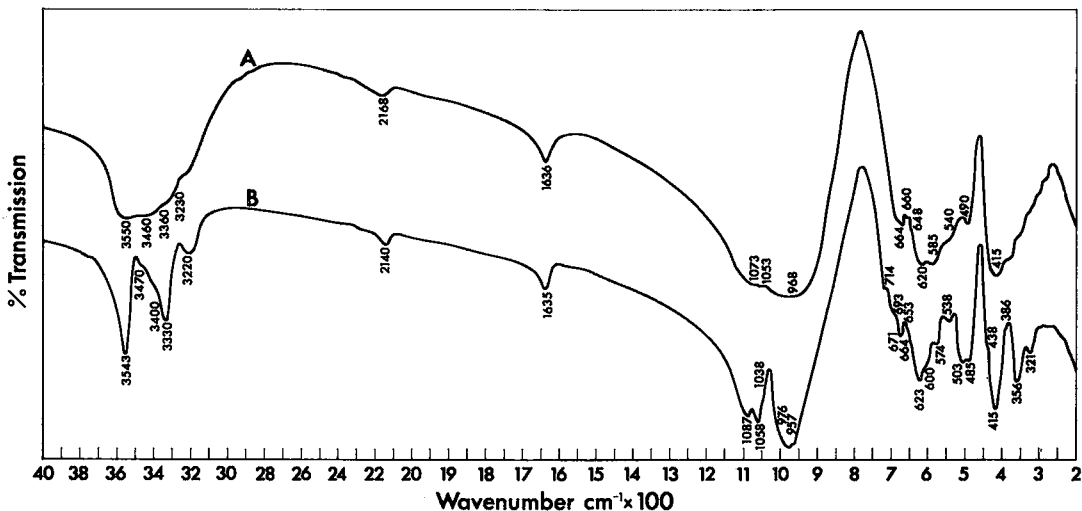


FIG. 4. Infrared spectra of tetranatrolite (A) and natrolite (B) from Mont St-Hilaire.

trolite, at a heating rate of 10°C/min. in air, showed a slow weight-loss of 1.31% between room temperature and 135°C and a major, rapid weight-loss of 9.59% between 135 and 435°C. Rehydration took place immediately on cooling from 435°C to room temperature, but the initial weight-loss, thought to be due to adsorbed water, was not recoverable within 12 hours after cooling to room temperature. The X-ray powder pattern of the rehydrated material is basically identical to that of the unheated material, except that some lines tend to be diffuse. Rehydration is found to be impossible if the mineral is heated to 615°C. The dehydrated material quenched from that temperature gives an X-ray powder pattern essentially similar to that of metanatroilite (Peng 1955). The material quenched from 820°C is amorphous. The TGA curve of natrolite associated with tetranatroilite shows a rapid single-stage weight-loss of 9.73% between 200 and 369°C.

Tetranatroilite from Mont St-Hilaire treated hydrothermally at 265°C and 0.5 kbar for 7 days remained unchanged. Treated at 330°C and 0.5 kbar for 10 days, the mineral decomposed to analcime with a trace of an unidentified compound, and at 445°C and 1 kbar for 7 days the mineral decomposed to nepheline and analcime with a trace of another unidentified compound.

DISCUSSION

Although X-ray and infrared data suggest that tetranatroilite is a disordered modification of natrolite, its phase relationship with natrolite remains obscure. High-temperature structural studies of natrolite (Peacor 1973) showed that the $Al_2Si_5O_{10}$ framework stayed strongly ordered at least up to 198°C. Continuous-heating X-ray-diffraction studies by van Reeuwijk (1972), using a Guinier-Lenné camera, showed that natrolite on dehydration at 280°C transformed to metanatroilite, which changed to another phase at 510°C. However, none of these high-temperature phases can be related to tetranatroilite. Our attempt by hydrothermal experiments to reorder Al and Si in tetranatroilite also was unsuccessful. Thus, it seems that factors other than temperature and pressure, such as stoichiometry, may have played important roles in the formation of tetranatroilite. The introduction of Ca into the structure of natrolite may promote the disorder of Al and Si during crystallization and, subsequently, may serve to stabilize the disordered structure.

Further studies of fresh, composite tetra-

trolite-natroilite crystals preserved in water revealed the existence of yet another new mineral, a higher hydrate form of natrolite. This new phase, which dehydrates partially to form tetranatroilite, is described in the following paper by Chao (1980).

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