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THORNASITE, A NEW HYDROUS SODIUM THORIUM SILICATE FROM MONT ST-HILAIRE, QUEBEC*

VALERIE E. ANSELL AND GEORGE Y. CHAO
Department of Geology, Carleton University, Ottawa, Ontario K1S 5B6

ABSTRACT

Thornasite, from Mont-Hilaire, Quebec, occurs as small, anhedral crystals in patches of white, powdery brockite associated with yofortierite and analcime. The mineral is rhombohedral, space group $R\bar{3}m$ or $R32$, with cell parameters a 29.08(1), c 17.30(1) Å, $Z = 18$. The strongest lines of the X-ray powder-diffraction pattern [d in Å(I)(hkl)] are: 14.54(20)(110), 8.17(30)(012), 7.27(100)(220), 5.09(20)(042), 4.17(70)(422), 3.239(30)(262), 2.959(20)(802), 2.890(25)(811,713). The grains are colorless to very pale green with a white streak and are translucent to transparent with a vitreous to waxy lustre. Thornasite is brittle with a subconchoidal to uneven fracture. Under long- and short-wave ultraviolet radiation, the mineral fluoresces a bright apple-green. Density 2.62(2)(meas.), 2.627 g/cm^3 (calc.). Optically, the mineral is uniaxial (+) with ω 1.510(1) and ϵ 1.512(1). The average of five electron-microprobe analyses gives SiO_2 57.93, Al_2O_3 1.42, ThO_2 22.45, UO_2 0.88, Na_2O 1.55, K_2O 1.10, CaO 0.17, F 0.40, Cl 0.05, H_2O^- 8.62, H_2O^+ 5.14, $-(F + Cl) \equiv O$ 0.018, sum 99.53 wt. % corresponding to: $(Na_{0.56}K_{0.26}Ca_{0.03})_{\Sigma 0.85}(Th_{0.94}U_{0.04})_{\Sigma 0.98}(Si_{10.69}Al_{0.31})_{\Sigma 11.00}O_{24.12}F_{0.23}Cl_{0.02}\cdot 8.5H_2O$ or, simply, $(Na,K)ThSi_{11}(O,H_2O,F,Cl)_{33}$. The compatibility index is 0.096. Thornasite is slightly metamict.

Keywords: thornsite, hydrous sodium thorium silicate, new mineral species, Mont St-Hilaire, Quebec.

SOMMAIRE

La thornasite du mont St-Hilaire, Québec, se présente en petits cristaux xénomorphes sur une mosaïque de brockite blanche pulvérulente associée à yofortiérite et analcime. Elle appartient au système rhomboédrique, groupe spatial $R3m$ ou $R32$; la maille a pour dimensions a 29.08(1), c 17.30(1) Å, $Z = 18$. Les raies les plus intenses du cliché de poudre [d en Å(I)(hkl)] sont: 14.54(20)(110), 8.17(30)(012), 7.27(100)(220), 5.09(20)(042), 4.17(70)(422), 3.239(30)(262), 2.959(20)(802), 2.890(25)(811,713). Les grains passent d'incolores à un vert très pâle à trait blanc; ils sont transparents à éclat vitreux à cireux. La thornasite est fragile à fracture subconchoïdale, raboteuse. Dans l'ultra-violet, de longue ou courte longueur d'onde, elle est fluorescente en vert-pomme brillant. Poids spécifique 2.62(2)(mes.), 2.627 (calc.) g/cm^3. Optiquement uniaxe positive: ω 1.510(1), ϵ 1.512(1). La moyenne de cinq analyses à la microsonde donne SiO_2 57.93, Al_2O_3 1.42, ThO_2 22.45, UO_2 0.88, Na_2O 1.55, K_2O 1.10, CaO 0.17, F 0.40, Cl 0.05, H_2O^- 8.62, H_2O^+ 5.14, $-(F + Cl) \equiv O$ 0.018, total 99.53% (en poids). Ces données correspondent à: $(Na_{0.56}K_{0.26}Ca_{0.03})_{\Sigma 0.85}$ $(Th_{0.94}U_{0.04})_{\Sigma 0.98}(Si_{10.69}Al_{0.31})_{\Sigma 11.00}O_{24.12}F_{0.23}Cl_{0.02}\cdot 8.5H_2O$ ou, idéalement, à $(Na,K)ThSi_{11}(O,H_2O,F,Cl)_{33}$. L'indice de compatibilité est de 0.096. La thornasite est légèrement métamicte.

Mots-clés: thornasite, silicate hydraté de sodium et thorium, Mont St-Hilaire, Québec, nouvelle espèce minérale.

INTRODUCTION

The new mineral species *thornasite,* previously designated UK27 (Chao & Baker 1979), was discovered in small amounts (less than 100 mg) in material collected from the floor of the De-Mix Quarry, Mont St-Hilaire, Quebec. The mineral occurs as anhedral grains, imbedded in patches (4–8 mm in diameter) of white, powdery brockite associated with yofortierite and analcime. Accessory minerals include: eudialyte, serandite, polylithionite, leifite, natrolite, microcline, albite, mangan-neptunite, steacyite, catapleiite, epididymite and calcite. Serandite is altered to different degrees to a black substance that has stained other minerals on the samples.

The relationship of thornasite to brockite is not known. Brockite could be an alteration product of thornasite, or both minerals may be the alteration product of another mineral.

The name recalls the composition. The mineral and name have been approved by the Commission on New Minerals and Mineral Names, I.M.A. The type specimens are deposited with the Royal Ontario Museum, Toronto (M42070) and the National Museum of Natural Sciences, Ottawa (50770).

X-RAY CRYSTALLOGRAPHY

Single crystals of thornasite were studied using the precession technique. The c-axis cone axis, the 0–4th level of c-axis precession and the 0-level of a-axis precession photographs, taken with Mo$K\alpha$ radiation ($\lambda = 0.7107$ Å) indicate that thornasite is rhombohedral, space group $R\bar{3}m$, $R32$ or $R3m$. The mineral was found to be pyroelectric. Therefore, thornasite lacks a centre of symmetry, and the correct space-group is $R3m$ or $R32$. The single-crystal and powder-

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TABLE 1. X-RAY POWDER-DIFFRACTION DATA FOR THORNASITE

h	k	l	dcalc Å	dobs Å	I/I$_o$	h	k	l	dcalc Å	dobs Å	I/I$_o$
1	1	0	14.540	14.54	20	2	10	4	2.004		
3	0	0	8.395	8.41	10	1	12	1	1.997	1.987	<1
0	1	2	8.183	8.17	30	3	11	1	1.960	1.968	1
2	2	0	7.270	7.27	100	5	10	1	1.892	1.892	<1
1	3	1	6.477	6.47	10	1	10	6	1.840		
1	1	3	5.362	5.37	<1	2	6	8	1.839	1.840	<1
0	4	2	5.091	5.09	20	8	8	0	1.818	1.818	2
2	3	2	4.805	4.81	15	7	9	1	1.803	1.801	2
2	4	1	4.589	4.57	<1	8	0	8	1.783		
5	1	1	4.376	4.37	5	3	8	7	1.777	1.780	4
4	2	2	4.170	4.17	70	5	11	0	1.776		
1	5	2	4.009	4.01	10	1	13	3	1.772		
1	6	1	3.749	3.745	<1	6	10	2	1.761		
3	2	4	3.463	3.457	15	14	0	2	1.761	1.762	7
5	2	3	3.305	3.303	5	14	1	1	1.725	1.726	1
2	6	2	3.239	3.239	30	13	3	1	1.701	1.698	7
2	4	4	3.201	3.196	<1	5	11	3	1.698		
3	1	5	3.101	3.099	<1	7	9	4	1.672		
8	0	2	2.958	2.959	20	14	2	0	1.668		
8	1	1	2.906	2.890	25	10	6	4	1.651	1.666	3
7	1	3	2.888			0	14	4	1.661		
9	0	0	2.798			9	7	5	1.606	1.606	1
1	8	2	2.790	2.788	15	3	14	1	1.596	1.596	<1
6	3	3	2.780			11	7	1	1.586		
2	8	0	2.748	2.748	7	13	1	6	1.564	1.566	<1
6	4	2	2.740			10	2	8	1.563		
6	2	4	2.717	2.716	<1	0	16	2	1.549	1.549	2
3	7	2	2.693	2.687	5	6	12	3	1.530	1.530	1
5	5	3	2.597			1	15	4	1.519	1.513	1
5	4	4	2.585	2.598	5	1	5	11	1.486		
0	8	4	2.545	2.546	7	4	6	10	1.485	1.485	<1
0	10	2	2.418	2.419	3	16	1	3	1.474	1.474	1
4	6	4	2.402	2.399	<1	15	3	3	1.459		
4	7	3	2.379	2.377	4	16	2	2	1.453	1.458	2
6	0	6	2.377							1.444	<1
4	8	2	2.295	2.288	<1					1.410	<1
4	4	6	2.259	2.258	4					1.391	<1
10	1	3	2.208	2.205	<1					1.341	1
10	2	2	2.188	2.187	7					1.327	<1
10	0	4	2.177							1.292	<1
3	9	3	2.159	2.159	7					1.275	<1
12	0	0	2.099	2.092	2					1.232	<1
8	4	4	2.085							1.188	1
7	7	0	2.077	2.076	2					1.174	<1
8	5	3	2.070							1.148	<1
4	10	0	2.016	2.017	7					1.136	<1
8	6	2	2.013								

CuKα radiation (λ=1.5418Å), 114.6 mm Gandolfi camera, Si internal standard, visual intensities.

diffraction photographs are generally of poor quality. The reflections tend to be diffuse, indicating that thornasite is slightly metamict. Attempts to improve the crystallinity of thornasite by heating at 300°C resulted in complete collapse of the structure. The powder pattern (Table 1) was indexed with the aid of precession photographs. Cell parameters were first determined from single-crystal photographs and then refined by a least-squares method using X-ray powder-diffraction data. The refined values are: a 29.08(1), c 17.30(1) Å, c/a = 0.5949:1 and Z = 18.

Physical and Optical Properties

Thornasite occurs as anhedral grains up to 0.7 mm in diameter. The grains are transparent to translucent, colorless with a very pale green tint, vitreous to waxy lustre and a white streak. The surfaces of the grains commonly appear etched and have a white, opaque, powdery coating of brockite that may be removed by means of an ultrasonic bath. Very fine-grained unidentified inclusions occur at or near the surface. The mineral is brittle, with a subconchoidal to uneven fracture. Individual grains are too small to test for hardness. Attempts to determine microhardness were unsuccessful owing to the brittle character. Thornasite fluoresces bright apple-green under long- and short-wave ultraviolet light.

The average of three determinations of density, by heavy-liquid flotation technique and using different grains, is 2.62(2) g/cm^3. The calculated value, based on the chemical formula and the cell parameters, is 2.627 g/cm^3. Thornasite is only slightly attacked by 1:1 HCl at room temperature and is unaffected by either 1:1 HNO_3 or H_2SO_4.

Optically, the mineral is uniaxial positive, with ω 1.510(1) and ϵ 1.512(1), as determined in sodium light at room temperature using a spindle stage and crystals previously oriented by precession techniques. The extinction of the grains between crossed nicols is generally not sharp, and some crystals show patchy extinction. The uniaxial interference figures are very diffuse, perhaps because of the slightly metamict nature of thornasite.

Chemical Formula

Thornasite was analyzed using a Cambridge MK5 microprobe at an accelerating voltage of 15 kV and a specimen current of 50 nA (measured on pure iron). A beam diameter of 10 μm was used to reduce damage to the mineral, in view of the volatility of sodium under the electron beam. The following standards were used: thorianite (Th), pyroxene (Si), hornblende (Na, K, Al, Ca), tugtupite (Cl), riebeckite (F) and synthetic brannerite (U). Both wavelength-dispersion and energy-dispersion microprobe scans revealed no elements heavier than oxygen other than those recorded in Table 2.

The infrared-absorption spectrum (Fig. 1) was obtained using a Perkin - Elmer model 683 infrared spectrophotometer. A pellet was prepared containing 200 mg of KBr and 0.76 mg of thornasite. The resolution of the spectrum as a whole is poor; however, the band at 1630 cm^{-1}, characteristic of

TABLE 2. CHEMICAL COMPOSITION OF THORNASITE (ELECTRON-MICROPROBE DATA)

	1	2	3	4	5	AVG
SiO_2	56.99	58.30	57.22	58.02	59.12	57.93
Al_2O_3	1.20	1.16	1.20	1.77	1.79	1.42
ThO_2	22.30	21.90	22.38	22.20	23.49	22.45
UO_2	0.56	0.58	0.69	1.22	1.37	0.88
Na_2O	2.06	1.67	1.66	1.17	1.17	1.55
K_2O	0.92	0.83	0.97	1.40	1.40	1.10
CaO	n.d.	n.d.	n.d.	0.17	0.17	0.17
F	0.20	0.20	0.14	0.75	0.73	0.40
Cl	0.03	0.01	0.02	0.11	0.10	0.05
*H_2O^-	-	-	-	-	-	8.62
*H_2O^+	-	-	-	-	-	5.14
-(F+Cl)=O	0.09	0.08	0.06	0.34	0.33	0.18
Total	84.17	84.58	84.22	86.47	89.01	99.53

* H_2O by simultaneous TG and EG analysis.

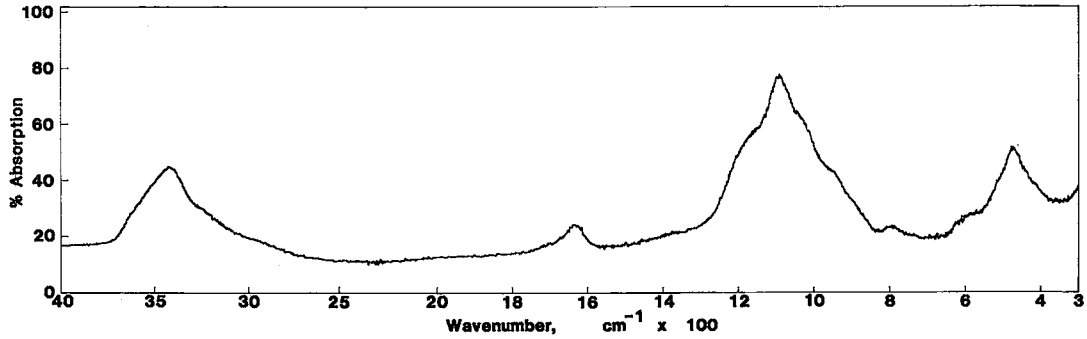

FIG. 1. Infrared-absorption spectrum of thornasite, from 300 to 4000 cm^{-1}.

H–O–H bending, confirms the presence of water in the mineral.

Simultaneous thermogravimetric (TGA) and evolved-gas analysis (EGA), carried out under high vacuum (10^{-6} Torr) on 5.16 mg of thornasite, showed an initial loss of 0.445 mg after 3.5 h of preliminary high-vacuum treatment. On heating at 10°C/minute, a steady rate of loss amounting to 0.26 mg was observed up to 1135°C. Three peaks on the pressure curve at 105, 275 and 600°C correspond to three stages of water evolution on the EGA curve. Aside from a weak trace of CO_2 detected at higher temperatures, no other volatiles were evolved. The total weight-loss was 0.71 mg, which is interpreted as a water content of 13.8(1.0) wt.%. The residue consists of thorianite plus huttonite, as identified using X-ray powder-diffraction methods.

The numbers of (Th+U), (Na+K+Ca) and (Si+Al) atoms in the unit cell, calculated using the measured density, the cell volume and the average results of five electron-microprobe analyses, are 17.66, 15.28 and 195.6, close to 1:0.9:11 for $Z = 18$. Therefore, the chemical formula of thornasite was calculated on the basis of 11(Si+Al) to: $(Na_{0.56}K_{0.26}Ca_{0.03})_{\Sigma 0.85}(Th_{0.94}U_{0.04})_{\Sigma 0.98}(Si_{10.69}Al_{0.31})_{\Sigma 11.00}O_{24.12}F_{0.23}Cl_{0.02}\cdot 8.5H_2O$. Since neither the structural role of water nor the ratio of H_2O to OH is known, all water is tentatively treated as H_2O. The formula may be simplified to $(Na,K)ThSi_{11}(O,H_2O,F,Cl)_{33}$.

The compatibility index of thornasite, based upon the Gladstone–Dale relationship (Mandarino 1976, 1979, 1981), is 0.096. The poor compatibility of the chemical and physical data is most likely due to the slightly metamict character of thornasite.

Thornasite is not related to any other known mineral.

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