

NEW OCCURRENCES AND DATA FOR SPENCITE

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ABSTRACT

Spencite is described from two new occurrences in Ontario: a pyroxene-rich layer within a calcite-fluorite vein-dyke (Cardiff property) and a pyroxene — and amphibole — rich pegmatite (Faraday mine). All spencites are metamict but after heating to 900°C in air, are recrystallized to an apatite (britholite) phase and a fluorite (CeO_2) phase. The physical properties vary considerably but for convenience spencites have been divided into three groups: Spencite I (S.G. = 3.05-3.08, $n = 1.620$ -1.630), Spencite II (S.G. = 3.27-3.40, $n = 1.670$ -1.685) and Spencite III (S.G. = 3.27-3.81, $n = 1.711$ -1.713). These properties reflect characteristics in lanthanide composition with Spencite I being the most yttric (Y maximum), and Spencite III being the most ceric (Ce maximum) and forming a link with tritomite. A lanite is commonly associated with spencite and may, in part, be derived from it by addition of Si and Al and release of B. Because of leaching of constituents, spencite does not yield a rational formula but its origin from hellandite is considered possible.

INTRODUCTION

Spencite, a rare-earth borosilicate, was first described from a calcite-fluorite vein-dyke of the Haliburton-Bancroft region, Ontario by Frondel (1961). The following year it was reported from a granite pegmatite in New Jersey by Jaffe & Molinski (1962). Despite some substantial differences, these two described spencites did not appear to express the complete range of chemical composition and physical properties of this rare mineral. Spencite is also related to tritomite but their relationship remains vague, both minerals being metamict and of rather variable composition (Jaffe & Molinski 1962; Kupriyanova *et al.* 1966; Borneman-Starynkevitch 1968).

In an attempt to resolve these difficulties, the authors studied five specimens of rare earth borosilicates collected from occurrences in the Haliburton-Bancroft region, including spencites from two hitherto unreported occurrences, and compared the data with those for a specimen of type tritomite.

It is difficult to assign credits in a multi-authored paper of this nature. However, in general, the chemical data and nearly all physical and x-ray data were contributed by Semenov, Proshchenko, Kazakova and Kataeva. The collection of samples, information on occurrences and mineralogical associations are the work of Hogarth and Steacy. All authors contributed to the preparation of the paper and to the discussion.

NEW OCCURRENCES OF SPENCITE

The three known occurrences of spencite in the Haliburton-Bancroft region are shown in Figure 1. All lie within, or close to, a syenite horizon which is folded around the north end

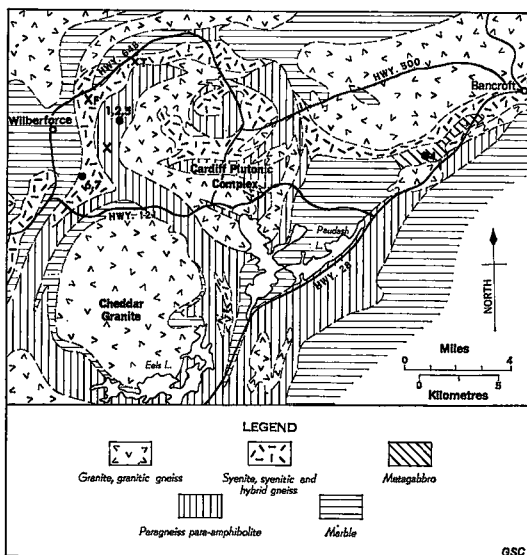


Fig. 1. Location of spencite occurrences near Bancroft, Ontario. Numbers correspond to the samples in this paper. Known occurrences are depicted by a solid circle (●). Possible occurrences are depicted by an (X).

of the Cardiff plutonic complex although there is no direct evidence of a genetic relationship with the syenite. The original locality is denoted by the occurrence symbol followed by the specimen numbers 1, 2, 5. The two new occurrences are the Faraday mine (specimen 4) and the Cardiff property (specimens 6, 7).

The Faraday mine operated from 1957 to 1964 and was one of the first uranium producers of the pegmatite class. Uranium orebodies at the Faraday mine have been described by Satterly (1957) and Bullis (1965). Spencite was found in pegmatite on the 613 drift, 900 foot level, in 1963. The mineral was collected from a small "tongue" or dyke of pegmatite intruding paragneiss, and situated under the main pegmatite dykes of the immediate area. Later, spencite was found in other pegmatites in the 613 drift.

The Cardiff property represents one of the unique calcite-fluorite-uraninite vein-dykes (nomenclature of Ellsworth 1932, p. 215) of the Haliburton-Bancroft region. The deposit was first explored for fluorite and later for uranium but operations ceased in 1953. The geology of the Cardiff property has been described by Satterly (1957). Spencite was collected in 1958 from the South Zone, some 350 feet north-northeast of the shaft and almost directly above the old adit. Here three layers of calc-silicate rock, intercalated with a calcite-fluorite vein-dyke, were exposed in a small pit. The central and widest layer of calc-silicate rock showed surface weathering to tengerite and contained considerable spencite. It was up to 1½ feet wide and was traced for about 25 feet. The other two layers contained abundant allanite but no visible spencite. A visit in 1968 proved disappointing: in 10 years the pit had largely filled with debris.

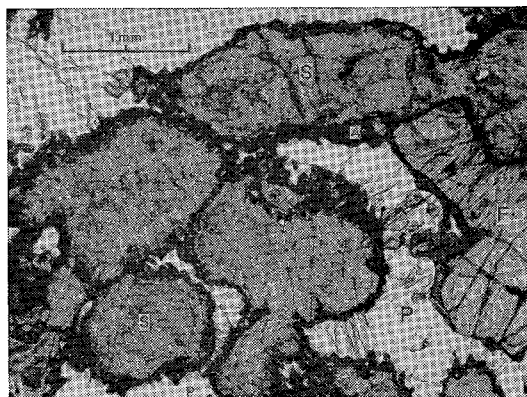


Fig. 2. Photomicrograph (plane-polarized light) of spencite from the Cardiff property. A rim of allanite and quartz (A) separates spencite (S) from microcline and oligoclase (P). Fluorite (F) is also rimmed by allanite and quartz.

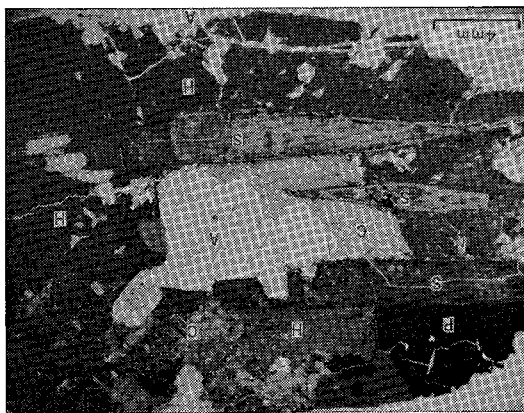


Fig. 3. Photomicrograph (plane-polarized light) of spencite from the Faraday mine. A: albite, C: calcite, H: arfvedsonite, P: pyrite, S: spencite.

Spencite at the Cardiff property occurs as rounded aggregates (Fig. 2) but spencite from the Faraday mine has a tabular to wedge-shaped habit (Fig. 3). In thin section these "crystals" are green inclining to yellow on their borders. All spencites are optically isotropic although portions of thin sections from the Cardiff mine are weakly anisotropic.

Mineral associations are shown in Table 1. All occur in coarse grained granoblastic rock, commonly rich in pyroxene and amphibole, although extremes of material poor in these minerals are also quoted (IIB and IIIB). The host rock from the Faraday mine is more alkaalic than that from the Cardiff property. Minor minerals identified from hand specimen by x-ray diffraction,

TABLE 1. MODAL ANALYSES OF SPENCITE-BEARING THIN SECTIONS

	Faraday mine, 613 drift, 900 ft. level (Granite pegmatite)			Cardiff property, south zone (Calcite-fluorite vein-dyke)		
	IIA	IIB	IIC	IIIA	IIIB	IIIC
Spencite	14.7	49.8	12.1	53.6	17.1	45.7
Allanite & clinozoisite	--	0.3	0.1	0.9	9.8	2.8
Opaque*	4.9	0.1	tr.	tr.	tr.	0.1
Titanite	0.2	3.1	2.0	0.3	tr.	0.1
Aegerine-augite	0.9	--	22.8	--	--	--
			As 40			
Augite	--	--	--	15.8	0.3	14.4
Arfvedsonite	53.4	--	8.4	--	--	--
Hornblende	--	--	--	--	tr.	--
Biotite	0.7	tr.	tr.	--	1.8	0.1
Calcite	13.3	3.8	35.3	4.0	tr.	7.7
Plagioclase	9.4	0.1	16.8	11.6	35.9	10.5
	An	5	4	26	23	23
Microcline (perthitic)	--	tr.	0.7	11.9	12.0	13.3
Quartz	2.3	25.3	1.5	1.0	7.2	1.8
Tourmaline	--	15.3	--	--	tr.	--
Fluorite	--	--	--	0.7	15.9	2.0
Serpentine	0.1	--	--	0.2	--	1.4
Others†	tr.	2.2	0.3	tr.	--	tr.
Total	99.9	100.0	100.0	100.0	100.0	99.9

*Opaque includes magnetite (IIA), pyrite and uraninite
 †Others include apatite and zircon (IIA), uranothorite (IIB), hematite (IIC), tengerite (IIIA), cenosite (IIIC)

though not seen in thin section were, from the Faraday mine: anhydrite, galena and chalcopryrite, and from the Cardiff mine: scapolite and apatite.

Minerals, possibly also belonging to the spencite group, have been found in other calcite-fluorite vein-dykes of the region. Thus on Lot 13, Con. XXII, Cardiff tp. (Topspar property) a radioactive mineral, which first appeared to be uranorhite, was shown to contain boron (Lang 1952, p. 138). The "greyish to reddish, heavy... material" from pit 14 at the Fission mine (Ellsworth 1932, p. 227) tentatively identified by Ellsworth as "chiefly titanite" and later by S.C. Robinson as "melanocerite" (Satterly 1957, p. 58) may be a mineral of this type. Specimens of these minerals could not be located in collections to which the writers had access but are plotted as possible occurrences in Figure 1. It should be mentioned that, in physical appearance, spencite is remarkably similar to allanite and the two minerals may have been confused with one another in the past.

PHYSICAL, X-RAY AND CHEMICAL DATA OF SPENCITES

Physical properties

Table 2 lists the physical properties of spencites from the Haliburton-Bancroft region (Nos. 2,4,5,6 and 7) together with data reported by Frondel (1961) for spencite from Haliburton and by Jaffe & Molinski (1962) for spencite from Cranberry Lake. These are compared in the table with the data for type tritomite (No. 8). Nos. 2,4,5 and 8 were sufficiently rich in massive spencite or tritomite for the mineral to be separable by hand. Nos. 6 and 7 were concentrated from a single specimen by heavy liquid and magnetic methods and the final separation of the two colour varieties was made by hand under a binocular microscope.

On the basis of specific gravity and refractive index (Table 2), the spencites can be conve-

niently divided into three groups that are referred to as Spencite I, Spencite II and Spencite III. Spencite I includes specimens from the original locality (Lot 7, Con. XX, Cardiff township) and is characterized by the lowest specific gravity (3.05-3.08) and refractive index (1.620-1.630). Spencite II comprises specimens from Cranberry Lake, New Jersey and the Faraday mine, together with a variety from the original locality. It is characterized by an intermediate specific gravity (3.27-3.40) and refractive index (1.670-1.685). Spencite III is from the Cardiff property and has the highest specific gravity (3.72-3.81) and refractive index (1.711-1.713).

Indentation hardness data are insufficient to categorize the three varieties. The great range in hardness for specimen 7 may be related to the presence of microfractures, a common feature in metamict minerals.

Differential thermal analysis (heating rate 12°C/min.; air-ignition) also suggests differences between Spencite I, Spencite II, and tritomite (Fig. 4, specimens 2,4 and 8 respectively). For Spencite II, the loss of weight at 175°C (T.G.A.) corresponds to 1.5% and at 495°C to 8.9% or to an amount approximately equal to the water in the unheated specimen. The 175°C endotherm may correspond to the release of molecular water and the 495°C endotherm to the disintegration of hydroxyl. The exothermic effect (730-860°C) in the three specimens is possibly the result of crystallization. A shortage of material prevented thermal investigation of Spencite III.

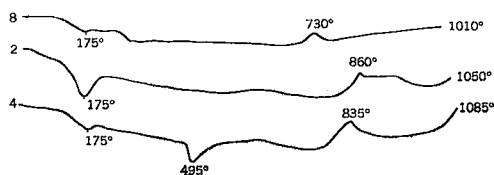


FIG. 4. Differential thermal analysis curves of spencite and tritomite. Exothermic peaks point upward.

TABLE 2. PHYSICAL PROPERTIES OF SPENCITE AND TRITOMITE

Type	Spencite I		Spencite II			Spencite III		Tritomite
Specimen no.	1*	2	3**	4	5	6	7	8
Locality	Lot 7, Con. XX, Cardiff Township, Ontario		Cranberry Lake, New Jersey	Faraday mine, Ont.	Lot 7, Con. XX, Cardiff Twp., Ont.	Cardiff property, Ontario		Langesundsfjord, Norway
Colour in hand specimen	blackish brown	blackish brown	black	blackish brown	blackish brown	amber	black	red brown
Colour in transmitted light	dark red brown	dark red	pale green	red brown	orange-red	amber	carmine red	red brown
Hardness } kg/mm ²	---	609	---	515	---	541	294-452	544
ness } Mohs'	(3.5)		(6.5)					
Specific gravity	3.05	3.08	3.40	3.39	3.27	3.81	3.72	4.20
Refractive index	~1.630	1.620	1.670	1.678	1.685	1.713	1.711	1.763

*Frondel (1961) **Jaffe & Molinski (1962)

•Determined by the indentation method by PMT-3 apparatus. Each value is an average of 6 to 13 measurements.

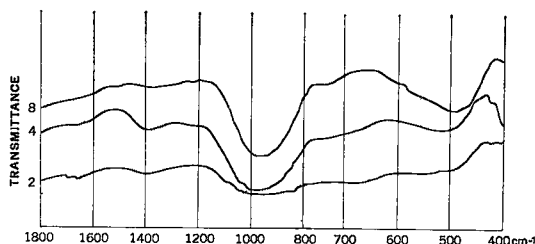


Fig. 5. Infrared curves of spencite and tritomite.

Infrared spectra (Fig. 5) are uniform and have the characteristics of metamict minerals: broad absorption bands in the range $900\text{--}1000\text{cm}^{-1}$, corresponding to valency fluctuations in the Si—O bond.

X-ray examination

X-ray data for spencites I, II and III are presented in Table 3. Photographs were taken with a 57.3 mm. diameter camera using Ni-filtered Cu radiation. Spencite is normally x-ray amorphous but one specimen (No. 2 from lot 7, Con.

XX, Cardiff township) yielded a few unidentifiable, diffuse lines with Cr radiation. In addition, a weakly anisotropic material from a specimen from the Cardiff property, which was close to No. 6 in specific gravity and refractive index (S.G. = 3.83, $n = 1.71$), gave with Cu radiation, a weak x-ray pattern doubtfully identified as monazite.

All spencites recrystallized after heating in air for 2 hours at 900°C , which is above the temperature at which the mineral begins to decompose ($450\text{--}550^\circ\text{C}$ according to Frondel 1961). Visual comparison of x-ray photographs suggests a single type of diffraction pattern, but on detailed examination differences are apparent in the number, quality and intensity of lines. After ignition, specimen 4 gave the best x-ray pattern, with the patterns of all other spencites notably inferior.

X-ray patterns of spencites ignited in air were generally similar, with the principal phases developed belonging to the apatite (britholite; type I of Kupriyanova *et al.* 1966) and fluorite (CeO_2) structures, but there are some differences. For example, the various specimens have characteristic interplanar spacings for the strongest line: Spencite I — $2.99\text{--}3.02\text{\AA}$, Spencite II — $2.77\text{--}2.83\text{\AA}$, Spencite III — 2.82\AA , and tritomite $3.11\text{--}3.13\text{\AA}$. Because the minerals were ignited under identical conditions, it may be assumed that the structural varieties of ignited products represent differences in the initial chemical compositions. After high-temperature ignition ($\sim 1000^\circ\text{C}$) the CeO_2 pattern becomes dominant.

Chemical composition

Chemical analyses of spencites and type tritomite are presented in Tables 4 and 5, the latter dealing specifically with the rare earth contents. Lanthanides in specimens 2 to 8 were determined by x-ray spectroscopy. Yttrium was derived by difference and the values shown are considered approximate. The analyses do not lead to unequivocal rational formulae.

Spencite I is notable for a high percentage of water and can be termed hydrated spencite. Spencite II has a high percentage of CaO but a rare-earth composition similar to that of Spencite I. The high temperature D.T.A. endotherm (Fig. 4, specimen 4) may represent hydroxyl water but infrared analysis (Fig. 5, specimen 4) did not prove its existence. Spencite III contains a similar amount of calcium as Spencite I but is set apart from Spencites I and II by its low silica and high rare earth contents. There is a predominance of light lanthanides in the rare earth distribution, with a maximum at cerium. In the main aspects of chemical composition as

TABLE 3. X-RAY DATA OF SPENCITES AFTER HEATING IN AIR

Spencite I		Spencite II		Spencite III	
1	2	3*	4	5	6
920°C	900°C	1000°C	900°C	900°C	900°C
1	2	3	4	5	6
$d(\text{\AA})$	$d(\text{\AA})$	$d(\text{\AA})$	$d(\text{\AA})$	$d(\text{\AA})$	$d(\text{\AA})$
-	-	50 4.02	3 4.11	-	-
-	-	40 3.63	3 3.94	-	-
-	-	30 3.42	3 3.46	-	-
-	-	-	-	-	1 3.44
6 3.13	3 3.12	50 3.13	5b 3.16	2 3.11	8b 3.13
-	-	40 3.05	-	-	10b 3.26
10 3.02	10 2.99	-	-	-	8b 3.11
4 2.91	2 2.89	-	-	-	-
4 2.80	7 2.80	100 2.78	10 2.83	10 2.81	10b 2.82
-	-	40 2.76	5 2.73	-	6b 2.87
6 2.71	4 2.71	30 2.69	-	-	5b 2.74
7 2.52	-	-	-	-	-
-	-	10 2.25	1 2.26	1 2.24	-
2 2.21	1b 2.19	-	-	-	-
2 2.12	1b 2.12	5 2.14	1 2.14	-	-
-	-	30 2.04	2 2.06	1 2.05	3b 2.16
-	-	-	4 1.96	-	-
-	-	40 1.930	4 1.93	4 1.94	5b 1.93
3 1.91	6b 1.91	30 1.874	2 1.90	3 1.89	-
-	-	-	7 1.85	6 1.84	5b 1.88
-	-	40 1.824	3 1.83	-	-
-	2 1.81	20 1.794	3 1.79	-	4 1.83
-	-	10 1.770	5 1.76	4 1.76	-
-	4 1.76	30 1.740	3 1.73	2 1.73	2b 1.74
-	-	30 1.709	-	-	-
5b 1.63	3 1.64	5 1.629	4b 1.65	1 1.64	7b 1.64
2 1.60	2 1.61	-	-	-	6b 1.64
-	-	5 1.571	-	-	-
-	-	5 1.530	3 1.55	2 1.54	1 1.54
-	-	20 1.498	2 1.51	3 1.52	-
-	-	20 1.468	2 1.48	2 1.48	-
1 1.46	3 1.46	30 1.448	4 1.46	1 1.46	1 1.46
-	-	20 1.423	2 1.44	-	-
1 1.34	-	20 1.311	-	-	2 1.36
-	-	20 1.278	-	-	-
-	-	30 1.251	3b 1.25	2b 1.24	3b 1.24
-	-	30 1.230	3b 1.23	-	-
-	-	40 1.217	-	-	-
-	-	10 1.174	2b 1.18	-	-
-	-	20 1.143	1b 1.14	-	-
-	-	30 1.130	-	-	-
-	-	50 1.111	5b 1.11	4b 1.11	-
-	2 1.10	50 1.103	-	-	-
-	-	10 1.091	-	-	-
-	-	5 1.077	-	-	-
-	-	5 1.029	-	-	3b 1.04
-	-	5 1.020	-	-	-
-	-	10 1.006	-	-	-
-	-	5 0.997	-	-	-
-	-	-	1b 0.92	-	3b 0.92

b=broad

*data of Jaffe & Molinski (1962)

well as physical properties, Spencite III, lies between Spencite II and tritomite.

Spencite is normally associated with uraninite and titanite, and impurities of uraninite may account for some of the uranium in the quoted analyses (Table 4). Spencite specimens 6 and 7

were comparatively pure (except for possible allanite rims) whereas those from the Faraday mine commonly contained titanite crystals. This could explain the relatively high content of TiO_2 (0.47%) in specimen 4.

DISCUSSION

Metamict spencite is derived from an unknown crystalline phase. It has been regarded as the yttrium analogue of tritomite (Jaffe & Molinski 1962) and as a member of the britholite group (Kupriyanova *et al.* 1966). The principal basis for these classifications is the presence of an apatite phase that appears only after heating to about 900°C. The rather obscure relationship of the chemical formulae of spencite and britholite, as previously questioned by Borneman-Starynkevitch (1968) would, by itself, hardly warrant grouping these minerals together in the apatite structural type.

In Figure 6 compositions of spencite are plotted with respect to X (the sum of cations with radii 0.7 to 1.3 Å), Z (the sum of cations with radii 0.2 to 0.5 Å) and O (the sum of O, F, Cl, OH and H_2O). Iron has been included with X and aluminum with Z. Also plotted are tritomite (I, II, 8), caryocerite (III) and melanocerite (IV).

It can be seen readily that compositions of the similar minerals spencite, tritomite, caryocerite and melanocerite are separated from apatite (A) and allanite (E). Spencites are clustered

TABLE 4. CHEMICAL COMPOSITION OF SPENCITE AND TRITOMITE

	Spencite I		Spencite II			Spencite III		Tritomite
	1	2	3	4	5	6	7	
P_2O_5	0.02	2.40	---	---	1.84	---	---	---
Nb_2O_5	---	---	---	1.44	---	---	---	1.70
TiO_2	0.27	0.45	0.22	0.47	0.65	trace	0.06	0.28
ZrO_2	---	<0.1	---	---	---	---	---	2.31
SiO_2	24.89	26.40	24.12	24.40	23.44	17.62	16.82	14.40
ThO_2	2.44	1.90	2.78	2.30	1.35	2.32	9.50	15.00
X_2O_3	33.80	36.84	27.99	35.11	33.72	50.68	44.13	41.44
Al_2O_3	3.87	2.82	6.08	1.46	6.97	1.20	3.19	0.85
B_2O_3	10.04	2.37	10.88	7.00	10.46	10.53	---	2.00
Fe_2O_3	3.22	7.15	---	3.57	2.22	1.80	1.15	1.71
FeO	---	---	3.39	---	---	---	---	---
CaO	7.81	5.48	17.32	12.90	13.90	9.66	5.45	7.52
SrO	0.05	---	---	0.10	---	---	---	0.62
MgO	0.50	1.08	0.20	0.40	0.77	0.15	trace	0.60
MnO	0.60	0.35	0.44	0.54	0.46	---	0.56	0.30
Na_2O	0.11	---	0.08	0.68	---	nf	---	---
H_2O	11.75	12.28	3.60	8.96	3.84	4.98	12.72	8.44
F	0.44	0.89	1.00	0.40	0.30	1.05	---	4.00
Others*	0.46	0.09	2.07	---	---	0.46	---	---
-O=F ₂	0.28	0.37	0.42	0.17	0.12	0.44	---	1.69
Total	99.99	100.13	99.75	99.56	99.80	100.06	---	99.48

*Others include: 1. K_2O 0.01, Cl 0.45 2. U_3O_8 0.09

3. UO_3 0.90, CO_2 0.84, PbO 0.11, BeO 0.22 6. U_3O_8 0.46

Analysis 1 is from Frondel (1961); analysis 3 is from Jaffe & Molinski (1962); other analyses are those of this study.

TABLE 5. THE RARE EARTH COMPOSITIONS OF SPENCITE AND TRITOMITE ($\Sigma\text{R.E.} = 100\%$)

	Spencite I		Spencite II		Spencite III		Tritomite
	1*	2	4	5	6	7	
La	2.2	2.2	3.8	2.0	15.7	15.5	25.0
Ce	7.1	7.6	21.0	7.8	35.0	38.0	44.0
Pr	1.6	2.1	3.2	1.9	3.0	2.9	2.5
Nd	5.6	8.7	15.4	7.5	11.9	12.5	10.0
Sm	3.2	4.2	4.4	2.6	1.8	1.5	0.6
Eu	0.4	0.8	0.8	0.4	0.3	0.2	0.1
Gd	5.0	5.6	7.0	4.0	2.7	2.3	0.4
Tb	1.0	1.2	0.9	0.7	0.3	0.2	0.0
Dy	5.9	8.0	6.2	5.3	2.7	1.9	0.5
Ho	1.6	2.2	1.2	1.4	0.5	0.2	0.0
Er	6.2	7.4	4.2	4.5	1.7	0.9	0.1
Tu	1.0	2.0	0.6	1.2	0.2	0.1	0.0
Yb	8.9	8.4	3.8	4.5	1.4	0.5	0.0
Lu	0.8	1.1	0.6	0.5	0.2	0.1	0.0
Y	49.5	[38.5]	[27]	[55.7]	[23]	[23]	[16.8]

*Joensuu & Ingamells (1966); data for 2 to 7 and tritomite are those of this study.

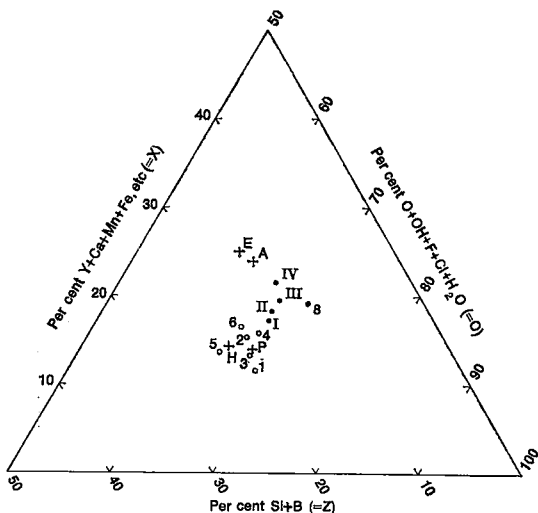
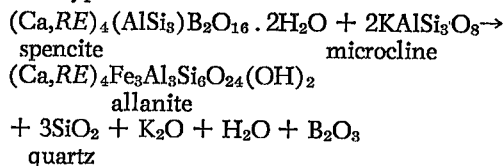


FIG. 6. Compositional diagram. Numbers 1 to 6 and 8 correspond to analyses of this paper; I, II are tritomites, III caryocerite, IV melanocerite (analyses from Borneman — Starynkevitch 1968); A is apatite-britholite ($\text{X}_5\text{Z}_3\text{O}_{18}$); E is allanite ($\text{X}_3\text{Z}_5\text{O}_{13}$); H is hellandite ($\text{X}_2\text{Z}_3\text{O}_9$) and P is a hypothetical compound ($\text{X}_5\text{Z}_7\text{O}_{24}$).

around H-hellandite and around P-($X_5Z_7O_{24}$). Specimen 5 is chemically similar to hellandite- $X_2Z_3O_8 \cdot H_2O$ (formula of Hogarth, Chao & Harris 1972) giving a formula $X_{1.85}Z_{3.09}O_{7.96} \cdot 0.77H_2O$.

Whether minerals of the hellandite group should be regarded as the ancestors of spencite is an entertaining hypothesis. The textures of the two are microscopically similar, both showing poorly-developed orthogonal cleavages. Hellandite has a tabular habit (Brøgger 1903; Hogarth, Chao & Harris 1972), as also does spencite at the Faraday mine. In addition, hellandite (monoclinic) at about 900°C, inverts to an apatite (hexagonal) structure, producing an x-ray pattern indistinguishable from that of ignited Spencite II (specimen 5).

Allanite, which is commonly associated with spencite, may be partly derived from it. The fact that, at the Cardiff mine, allanite and quartz occur along the contact of microcline (or plagioclase) and spencite (Fig. 4), suggests a reaction of the type:



where, under certain *T-P* conditions, the borosilicate became chemically unstable in the presence of feldspar and exchanged B_2O_3 with Al_2O_3 and SiO_2 to produce allanite. Fluorite appears to have preferentially replaced spencite.

Spencite varies widely in its physical properties and chemical composition but the data presented here will contribute to an understanding of its evolution in systematic mineralogy.

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