

Manganese humites and leucophoenicites from Franklin and Sterling Hill, New Jersey: parageneses, compositions, and implications for solid solution limits

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Abstract

The manganese humites, (alleganyite, manganhumite, and sonolite), together with some Mn-bearing samples of the Mg-humites, and the related phases leucophoenicite and jerrygibbsite, from the orebodies at Franklin and Sterling Hill, New Jersey, are described together with analytical data. Solid solution between humite and manganhumite is at least partially continuous. Expected Mn/Mg solid solutions between alleganyite and chondrodite, and between sonolite and clinohumite, are discontinuous; they are interrupted by apparently ordered phases. In all cases, the possible orderings involve Zn as well as Mn and Mg. There are no Mn end-members of the manganese humites at this locality. Manganese is apparently restricted in leucophoenicite (5.42–6.63 Mn per 7 octahedral cations) and in jerrygibbsite (7.79–8.02 Mn per 9 octahedral cations). Calcium is common to both leucophoenicite and jerrygibbsite, but among the Mn-humites, only sonolite accepts appreciable Ca (0.65 Ca per 9 octahedral cations). There is a “threshold” level of zinc in all studied samples; this “threshold” level is a constant for leucophoenicite (~0.3 Zn per 3 Si) and alleganyite (~0.2 Zn per 2 Si). No samples of leucophoenicite or jerrygibbsite were found to be Zn-free, suggesting either that Zn is required for their stability, or that these two phases might not be stable as end-members. Fluorine is present in all the Mn-humites and is proportional to the Mg-content, but is absent in leucophoenicite and jerrygibbsite.

Introduction

The magnesium humite species (norbergite, chondrodite, humite, and clinohumite) have been well-studied and recently summarized by Ribbe (1982). Their manganese analogues, however, have received less attention until recently. They are, in general, less common, found in metamorphosed Mn-deposits, and may have lower *P-T* stability ranges because of expansion of the structure by large Mn cations (Ribbe, 1982). Winter et al. (1983) have recently shown that the Mn-humites from Bald Knob, North Carolina, require water-rich conditions in silica undersaturated rocks and that the formation of particular species can be dependent upon complex phase relations involving $XH_2O/(XH_2O + XCO_2)$ and $a(SiO_2)$. In addition to the Mn isotypes of the humites, two other species, leucophoenicite and jerrygibbsite (Dunn et al., 1984) are known, but the factors which govern their formation remain obscure. Crystal structures have been determined for alleganyite, magnesian alleganyite, magnesian manganhumite, and sonolite, as cited below, but the degree of solid solution between the Mg- and Mn-humites is incompletely defined, although Winter et al. (1983) provided a partial plot of Mn/Mg miscibility, and Fukuoka (1981) provided compositional data for Japanese samples.

Franklin and Sterling Hill provide an ideal “laboratory” in which to investigate these solid solution relations for a number of reasons. First, *all* the known Mg- and Mn-

humite species occur there; the Mg-humites occur in the host Franklin Marble for the most part, and the Mn-humites in the orebodies themselves. Franklin is also host to the two anomalous, yet related, species, jerrygibbsite and leucophoenicite, the latter occurring in relative abundance. Second, Mn/Mg solid solution is common in the silicates at these deposits, and the activity of Fe is limited; in most cases it has been preferentially absorbed by andradite, which also may serve to isolate silicate reactions from the ubiquitous franklinite. Third, the Mn-humite minerals are moderately common at these deposits, occurring both within the primary ore and in secondary vein assemblages. Last, the Franklin and Sterling Hill assemblages offer a unique opportunity to study the behavior of zinc in the Mn-humites, an area of investigation not previously pursued and which, as is shown below, is worthy of continued investigation, particularly from a structural standpoint. The species discussed here are listed in Table 1, together with related phases. The Mg-humites from the Franklin Marble, having already been studied by numerous investigators, have been excluded from this study. Only samples with relations to the ore minerals are included.

Chemical composition

The samples studied herein were chemically analyzed using an ARL-SEM-Q electron microprobe utilizing an operating voltage of 15 kV and a sample current of 0.025 μ A,

Table 1. The humite and leucophoenicite groups.

$M^{2+}:Si$	Humite group	Mn-humite group	Leucophoenicite group
3:1	norbergite	unknown in nature	unknown
5:2	chondrodite	alleghanyite	unknown
7:3	humite	manganhumite	leucophoenicite
9:4	clinohumite	sonolite	jerrygibbsite

measured on brass. The standards used were synthetic tephroite (Mn,Si), synthetic ZnO (Zn), hornblende (Mg,Fe), and fluorapatite (F) for Mn-rich samples. Forsterite (Si,Mg) and rhodonite (Mn,Zn) were employed for Mg-dominant samples. The samples are homogeneous at the microprobe level. The resultant analyses are presented in Tables 2–6. All samples were individually verified by X-ray powder diffraction techniques prior to analysis.

Alleghanyite

Introduction

Alleghanyite was first described from the Bald Knob manganese prospect, Alleghany County, North Carolina, by Ross and Kerr (1932) who noted that it was an anhydrous manganese silicate related to tephroite. A subsequent study of Bald Knob material by Rogers (1935) found the mineral to contain H_2O , redefined the species as $Mn_5(SiO_4)_2(OH)_2$, and noted the apparent relationship to chondrodite. This isostructural relationship was further supported by Campbell Smith et al. (1944) in their description of alleghanyite from Rhiw, Wales. The crystal structure of alleghanyite was determined by Rentzeperis (1970) using Bald Knob material.

Alleghanyite, ideally $Mn_5(SiO_4)_2(OH)_2$, was first noted from Franklin and Sterling Hill by Cook (1969) who cleared up part of the confusion arising from earlier morphological studies. White and Hyde (1982) examined several Franklin samples using TEM techniques. Although they found several phases associated with alleghanyite (chiefly leucophoenicite and sonolite), these minerals were present as fragments, and not as intergrowths. Recently, a high-Mg alleghanyite from Sterling Hill was described by Petersen et al. (1984), and a crystal structure refinement of Sterling Hill material of similar composition by Francis (1985a) showed a high degree of cation ordering. Extensive analytical studies (present study) indicate that such ordering may be very common in samples from Sterling Hill.

Description

Alleghanyite occurs at both Franklin and Sterling Hill. At Franklin, there are few known assemblages. The best-preserved of these was identified by Palache (1928) as leucophoenicite. This material consists of veins up to 3 cm thick, associated with calcite, franklinite and sussexite. The bulk of this material is alleghanyite: it consists of massive alleghanyite which forms euhedral crystals on exposed surfaces. Leucophoenicite is also present, both as epitaxial

overgrowths on the underlying alleghanyite, and as apparently randomly oriented euhedra, perhaps of a subsequent growth period. Although alleghanyite has been found on a few Franklin specimens from other assemblages (e.g., NMNH C6884), most of the preserved pink-colored material from Franklin is leucophoenicite. However, it is not clear whether this is due to selective retention of the more attractive bright pink leucophoenicite by miners' casual collecting, or if this apparent predominance of leucophoenicite is due to geochemical conditions, a viewpoint here adopted.

At Sterling Hill, alleghanyite occurs as euhedral crystals and thin seams which crosscut the ore. These crystals frequently accompany arsenate species in hydrothermal veinlets and formed in apparent equilibrium with rare species such as kolicite, holdenite, magnussonite, adelite, kraisslite, chlorophoenicite, and others. Aside from these uncommon arsenates, other species associated with alleghanyite are franklinite, willemite, barite, and carbonates, all of secondary recrystallization. The observed species are all formed on willemite/franklinite ore which contains abundant calcite. The best samples have been found in recent years. The ubiquitous twinning observed by Winter et al., (1983) in end-member alleghanyite from Bald Knob was not observed either in these magnesian crystals, or in the more Mn-rich material from both Franklin and Sterling Hill.

Chemical composition

Microprobe analyses of alleghanyites are presented in Table 2. Examination of the data reveals several features. First is the absence of material with end-member composition. Although near end-member material is known from Bald Knob (Simmons et al., 1981; Winter et al., 1983), it is unknown at Franklin and Sterling Hill; most material from there is highly magnesian. Second, samples from Franklin are markedly lower in Mg than those from Sterling Hill, reflecting the higher general concentration of Mg in Sterling Hill silicates. Third, considering the first 6 analyses, there appears to be partial solid solution between the most Mn-rich material and that with an Mn:Mg ratio of 4:1 (the next five analyses in the table), but there is an evident break in solid solution between material with Mn_4Mg_1 and the rest of the samples. The possible ordering of some samples with a Mn:(Mg,Zn) ratio of 4:1 (8:2) might be permissible inasmuch as alleghanyite has space group $P2_1/b$, which has equipoint ranks of 4 and 2. Such material, however, has not yet been refined by crystal structure analysis.

The next 12 analyses in Table 2 represent the compositions of magnesian alleghanyite crystals from varied secondary fissures at Sterling Hill. In one sample which contained both massive material as a vein-filling, and euhedral crystals (NMNH 134638), the euhedral crystals have a higher Mg content. The ratios of Mn:(Mg,Zn,Fe) vary from 3.57:1.70 to 3.17:1.89. Material with the latter composition was described by Petersen et al. (1984), and has unit cell parameters, $a = 4.827$, $b = 10.613$, $c = 8.116\text{Å}$, $\alpha = 108.65^\circ$. A sample with very similar composition, i.e.: $Mn_{2.840}Mg_{1.948}Zn_{0.184}Fe_{0.023}Ca_{0.005}(SiO_4)_2(F,OH)_2$, was subjected to crystal structure analysis (Francis, 1985a). His

Table 2. Microprobe analyses of alleghanyite and chondrodite, in order of decreasing Mn content; octahedral cations are calculated on the basis of Si = 2.

Sample #	SiO ₂	FeO	MgO	ZnO	MnO	F	H ₂ O	O=F	Total	Fe	Mg	Zn	Mn	F	OH	ΣM ²⁺	Species	Locality
C6884	24.7	0.0	2.3	2.7	66.2	1.1	3.2	0.5	99.7	0.00	0.28	0.16	4.54	0.28	1.73	4.98	Alleghanyite	Franklin
147308	24.4	0.0	4.6	3.3	63.3	2.7	2.3	1.1	99.5	0.00	0.56	0.20	4.39	0.75	1.26	5.15	Alleghanyite	Sterling Hill
146909	25.0	0.2	2.8	3.1	64.0	0.2	3.7	0.1	98.9	0.01	0.33	0.18	4.34	0.05	1.97	4.86	Alleghanyite	Sterling Hill
R3878-2	25.1	0.0	4.8	3.4	64.2	1.2	3.2	0.5	101.4	0.00	0.57	0.20	4.33	0.30	1.70	5.10	Alleghanyite	Franklin
C6885	24.9	0.0	5.9	3.4	62.6	1.6	3.0	0.7	100.7	0.00	0.71	0.20	4.26	0.41	1.61	5.17	Alleghanyite	Franklin
93336	25.9	0.2	3.9	3.9	63.5	tr.	3.9	0.0	101.8	0.00	0.45	0.22	4.15	0.00	2.01	4.82	Alleghanyite	Franklin
137880	25.0	0.0	6.6	3.2	61.2	1.8	2.9	0.8	99.9	0.00	0.79	0.19	4.15	0.45	1.55	5.13	Alleghanyite	Sterling Hill
146201	25.0	0.3	6.2	3.2	60.4	2.2	2.7	0.9	99.1	0.02	0.74	0.19	4.09	0.56	1.44	5.04	Alleghanyite	Sterling Hill
147308	25.0	0.0	7.8	3.4	60.1	2.5	2.6	1.1	100.3	0.00	0.93	0.20	4.07	0.63	1.39	5.20	Alleghanyite	Sterling Hill
145958	25.1	0.3	6.7	3.6	59.3	2.4	2.6	1.0	99.0	0.02	0.80	0.21	4.00	0.60	1.38	5.03	Alleghanyite	Sterling Hill
148618	25.4	0.2	7.3	3.6	60.0	2.5	2.6	1.1	100.5	0.01	0.86	0.21	3.95	0.62	1.37	5.03	Alleghanyite	Sterling Hill
134638	25.5	0.2	12.5	3.9	53.7	3.6	2.1	1.5	100.0	0.01	1.46	0.23	3.57	0.89	1.10	5.27	Alleghanyite	Sterling Hill
JEM 1412	25.5	0.4	12.6	3.7	53.3	3.6	2.1	1.5	99.7	0.03	1.47	0.21	3.54	0.89	1.10	5.25	Alleghanyite	Sterling Hill
134866	25.7	0.2	12.2	3.6	53.7	2.3	2.8	1.0	99.5	0.01	1.42	0.21	3.54	0.56	1.45	5.18	Alleghanyite	Sterling Hill
JEM 1405	26.6	0.6	13.4	3.4	53.0	3.3	2.4	1.4	101.3	0.04	1.50	0.19	3.37	0.78	1.20	5.10	Alleghanyite	Sterling Hill
JEM 1404	26.4	0.6	13.1	3.7	51.9	3.3	2.4	1.4	100.0	0.04	1.48	0.21	3.33	0.79	1.21	5.06	Alleghanyite	Sterling Hill
134745	26.6	0.4	14.1	3.4	50.5	3.7	2.1	1.6	99.2	0.03	1.62	0.19	3.29	0.90	1.08	5.13	Alleghanyite	Sterling Hill
R8288	26.6	0.5	14.2	3.1	50.9	3.3	2.4	1.4	99.6	0.03	1.59	0.17	3.24	0.78	1.20	5.03	Alleghanyite	Sterling Hill
JEM 1408	26.9	0.5	14.7	3.3	50.9	2.9	2.7	1.2	100.7	0.03	1.63	0.18	3.21	0.68	1.34	5.05	Alleghanyite	Sterling Hill
JEM 1411	26.7	0.4	15.3	3.5	50.6	3.2	2.5	1.3	100.9	0.02	1.71	0.19	3.21	0.76	1.25	5.13	Alleghanyite	Sterling Hill
134638	26.4	0.3	14.5	4.1	50.0	3.8	2.2	1.6	99.7	0.02	1.64	0.23	3.21	0.91	1.11	5.10	Alleghanyite	Sterling Hill
146229	26.7	0.4	14.7	3.2	50.3	3.3	2.4	1.4	99.6	0.03	1.64	0.18	3.19	0.78	1.20	5.04	Alleghanyite	Sterling Hill
Petersen	26.7	0.4	15.0	3.5	50.3	3.1	2.5	1.3	99.9	0.02	1.68	0.19	3.17	0.73	1.25	5.06	Alleghanyite	Sterling Hill
144454	27.3	0.5	23.9	6.4	35.8	4.5	2.2	1.9	98.7	0.03	2.48	0.33	2.11	0.99	1.02	4.95	Chondrodite	Sterling Hill
JEM 2908	29.0	2.0	22.4	11.5	30.5	3.6	2.6	1.5	100.1	0.12	2.30	0.59	1.78	0.79	1.20	4.79	Chondrodite	Sterling Hill
Kolic-38	29.9	1.9	33.7	7.7	22.6	3.9	2.6	1.6	100.7	0.11	3.36	0.38	1.28	0.82	1.16	5.13	Chondrodite	Sterling Hill

determined lattice parameters, $a = 4.815(2)$, $b = 10.574(3)$, $c = 8.083(3)\text{\AA}$, $\alpha = 108.74^\circ$, are very similar to those of Petersen et al. (1984). Francis (1985a) found this alleghanyite to have ordered cations, with Zn and Mg in the smallest $M(3)$ octahedron, and Mn and Mg distributed such that the $M(1)$ and $M(2)_5$ octahedra are predominantly occupied by Mn with minor Mg. Of particular interest, in all of the analyses of alleghanyite presented here, is the invariance of Zn. Zinc is a common substituent in silicate phases at Franklin and Sterling Hill and its presence is expected. However, in no case is there an alleghanyite sample without Zn, nor is there one with Zn in excess of these relatively invariant amounts. Although it is tempting to dismiss relatively small amounts of Zn as a common octahedral substituent, such dismissal is not warranted here. Some of these alleghanyites co-exist with zincite (ZnO), but the majority have only willemite as an associated Zn-phase, and thus may be saturated with respect to Zn.

Because the amount of zinc is relatively invariant and because (as is shown below), the same relation holds for leucophoenicite, there is adequate reason to propose that Zn is limited in alleghanyite and other humite-related species. Fluorine is common in most alleghanyite, averaging 40 mol% of the (OH) site in magnesian material and generally decreasing with increasing Mn content.

The last three analyses in Table 2 are of manganooan chondrodites found in calcite-rich ore from Sterling Hill. The samples vary in texture and associated minerals.

Manganhumite

Manganhumite was originally described from the Brattfors Mine, Nordmark, Sweden, by Moore (1978), and the

crystal structure of this magnesian manganhumite was determined by Francis and Ribbe (1978) who found the material to be ordered with respect to Mn and Mg. End-member manganhumite was subsequently described by Simmons et al., (1981) and Winter et al. (1983) from the Bald Knob manganese prospect in North Carolina.

Manganhumite, formerly unknown from Franklin, is massive, medium brown in color, and occurs in cm-sized masses associated with abundant franklinite and zincite, the latter with much hetaerolite exsolution and traces of manganosite exsolution. Minor associated minerals are alleghanyite and calcite. Willemite is absent.

Manganooan humite was found at both Franklin and Sterling Hill. Some Franklin material consists of light brown anhedral blebs, associated with minor franklinite, willemite, and zincite in a rock which is predominantly calcite. The analysis labeled *Private* (Table 3) is that of a brown coating on what appears to be tephroite.

Manganooan humite from Sterling Hill is markedly different in texture. The samples are from one occurrence, are massive, medium brown in color, and associated with franklinite, calcite, and minor willemite. Chondrodite is intimately associated with the humite.

Microprobe analyses of manganhumite and manganooan humite are presented in Table 3. The analyses demonstrate much solid solution between Mg-rich material and that with approximately 3.75 atoms of Mg per 7 octahedral cations. Analyses of Franklin material show a gap between this composition and that of the magnesian manganhumite of Moore (1978). This gap, however, may be due to paucity of data points in this series and may not reflect compositional gaps in the natural system. Fluorine is found in all

Table 3. Microprobe analyses of manganhumite and humite, in order of decreasing Mn content; octahedral cations are calculated on the basis of Si = 3.

Sample #	SiO ₂	FeO	MgO	ZnO	MnO	CaO	F	H ₂ O	O=F	Total	Fe	Mg	Zn	Mn	Ca	F	OH	ΣM ²⁺	Species
R4105	26.2	0.0	0.4	2.7	65.6	1.8	1.2	2.0	0.5	99.4	0.00	0.07	0.23	6.36	0.33	0.43	1.53	6.88	Manganhumite
Private	30.1	1.2	25.2	10.8	29.7	0.2	2.4	1.9	1.0	100.5	0.10	3.75	0.79	2.51	0.02	0.76	1.26	7.17	Humite
Bostwick	30.9	1.8	29.5	13.4	21.6	0.0	3.2	1.6	1.3	100.7	0.15	4.27	0.96	1.78	0.00	0.98	1.04	7.16	Humite
RB1016	33.9	0.2	39.7	10.2	14.3	0.0	3.1	1.9	1.3	102.0	0.02	5.24	0.67	1.07	0.00	0.87	1.12	7.00	Humite
RB1015	35.0	0.2	44.5	8.3	10.8	0.0	2.5	2.3	1.1	102.5	0.02	5.69	0.52	0.78	0.00	0.68	1.32	7.01	Humite
S-U	34.7	0.9	44.3	9.8	9.6	0.0	3.0	2.0	1.3	102.1	0.06	5.57	0.62	0.70	0.00	0.82	1.15	6.95	Humite

samples and occupies approximately 22 to 49 mol% of the (OH) site.

Sonolite

Introduction

Sonolite, Mn₉(SiO₄)₄(OH)₂, was first described by Yoshinaga (1963) from eleven localities in Japan. It has subsequently been found elsewhere, including Sterling Hill and

Franklin (Cook, 1969) and Bald Knob (Winter et al., 1983). The crystal structure was determined by Kato (unpublished; discussed by Ribbe, 1982).

Description and composition

Microprobe analyses of sonolite are presented in Table 4. They are best discussed in three clusters, defined in part by apparent limitations in the Mg and Zn contents. The first group of analyses represents samples only from Frank-

Table 4. Microprobe analyses of sonolite and clinohumite, in order of decreasing Mn content; octahedral cations are calculated on the basis of Si = 4.

Sample #	SiO ₂	FeO	MgO	ZnO	MnO	CaO	F	H ₂ O	O=F	Total	Fe	Mg	Zn	Mn	Ca	F	OH	ΣM ²⁺	Species
149037	27.0	0.0	0.8	3.9	65.4	0.7	0.0	2.0	0.0	99.8	0.00	0.22	0.40	8.39	0.11	0.24	1.80	9.11	Sonolite
C6992	26.7	0.1	2.4	2.0	65.7	0.8	1.2	1.4	0.5	99.8	0.01	0.54	0.22	8.34	0.13	0.57	1.40	9.24	Sonolite
JEM 3068	26.3	0.0	0.5	2.9	64.4	2.0	1.0	1.5	0.4	98.2	0.00	0.11	0.31	8.30	0.33	0.48	1.52	9.05	Sonolite
C1412-1	26.9	0.0	0.9	2.2	65.5	2.4	0.0	2.0	0.0	99.9	0.00	0.20	0.24	8.25	0.38	0.00	1.98	9.07	Sonolite
149037	27.0	0.0	0.8	3.9	65.4	0.7	0.0	2.0	0.0	99.8	0.00	0.18	0.43	8.21	0.11	0.00	1.98	8.93	Sonolite
R1803	27.1	0.2	2.3	2.6	65.4	0.5	1.0	1.6	0.4	100.3	0.02	0.51	0.28	8.18	0.08	0.47	1.57	9.07	Sonolite
14152	26.7	0.3	1.9	3.1	64.2	1.0	1.2	1.4	0.5	99.3	0.04	0.42	0.34	8.14	0.16	0.57	1.40	9.10	Sonolite
JEM 3587	26.7	0.0	1.4	2.6	62.8	2.6	1.2	1.4	0.5	98.2	0.00	0.31	0.29	8.00	0.42	0.57	1.40	9.02	Sonolite
HAUCK.FMM	26.9	0.3	0.9	3.9	62.3	3.9	0.9	1.6	0.4	100.3	0.04	0.20	0.43	7.84	0.49	0.42	1.59	9.00	Sonolite
C2828-1	27.0	0.0	1.6	3.7	60.7	4.1	1.1	1.5	0.5	99.2	0.00	0.35	0.40	7.62	0.65	0.51	1.48	9.02	Sonolite
JEM 3080	27.3	0.0	4.6	2.7	63.0	0.0	0.9	1.6	0.3	99.8	0.00	1.01	0.29	7.82	0.00	0.42	1.56	9.12	Sonolite
JEM 1645	27.7	0.0	5.7	2.8	61.7	0.0	1.2	1.5	0.5	100.1	0.00	1.23	0.30	7.55	0.00	0.55	1.45	9.08	Sonolite
143755	27.6	0.2	6.0	2.7	61.4	0.0	1.0	1.6	0.4	100.1	0.02	1.30	0.29	7.54	0.00	0.46	1.55	9.15	Sonolite
JEM 1638	27.6	0.0	6.6	3.1	60.4	0.0	1.0	1.6	0.4	99.9	0.00	1.43	0.33	7.42	0.00	0.46	1.54	9.18	Sonolite
143587	27.5	0.2	7.6	3.1	58.8	0.0	0.9	1.6	0.4	99.3	0.02	1.65	0.33	7.24	0.00	0.41	1.55	9.24	Sonolite
143755	27.8	0.2	8.3	3.3	58.9	0.0	1.0	1.6	0.4	100.7	0.02	1.78	0.35	7.18	0.00	0.46	1.54	9.33	Sonolite
JEM 1946	27.8	0.0	8.1	3.4	58.4	0.0	1.2	1.5	0.5	99.9	0.00	1.74	0.36	7.12	0.00	0.55	1.44	9.22	Sonolite
13333	29.2	1.3	15.9	9.5	41.2	0.3	2.0	1.2	0.8	99.8	0.15	3.25	0.96	4.78	0.04	0.87	1.10	9.18	Sonolite
12965	29.3	1.4	16.4	10.0	40.7	0.2	1.8	1.3	0.8	100.3	0.16	3.34	1.01	4.71	0.03	0.78	1.18	9.25	Sonolite
R3522	29.2	1.5	16.9	10.1	40.1	0.3	2.0	1.3	0.8	100.6	0.17	3.45	1.02	4.65	0.04	0.87	1.19	9.33	Sonolite
S-K	29.4	1.0	17.4	8.8	39.9	0.1	1.7	1.4	0.7	99.0	0.11	3.53	0.88	4.60	0.01	0.73	1.27	9.13	Sonolite
RB1028	29.6	1.2	17.2	9.4	40.2	0.3	1.8	1.4	0.8	100.3	0.14	3.47	0.94	4.60	0.04	0.77	1.26	9.19	Sonolite
RB1040	30.0	1.1	18.9	7.4	39.8	0.2	1.6	1.5	0.6	99.9	0.12	3.76	0.73	4.50	0.03	0.67	1.33	9.14	Sonolite
JEM 2916	29.5	1.3	16.3	11.9	36.4	0.4	1.6	1.4	0.7	98.1	0.15	3.30	1.19	4.18	0.06	0.69	1.27	8.88	Sonolite
S-V	30.8	0.8	22.2	8.8	36.2	tr.	1.6	1.4	0.7	101.1	0.09	4.30	0.84	3.98	0.00	0.78	1.21	9.21	Clinohumite
S-A	30.6	1.6	22.4	7.4	35.6	0.1	2.1	1.3	0.9	100.2	0.17	4.37	0.71	3.94	0.01	0.87	1.13	9.20	Clinohumite
S-P	31.4	1.0	25.3	8.8	31.7	tr.	1.8	1.5	0.8	100.7	0.11	4.81	0.83	3.42	0.00	0.73	1.27	9.17	Clinohumite
S-R	31.6	1.2	25.5	9.9	30.1	tr.	1.8	1.5	0.8	100.8	0.13	4.82	0.93	3.23	0.00	0.72	1.27	9.11	Clinohumite
S-N	31.5	1.9	25.9	10.2	28.7	0.1	2.2	1.3	0.9	100.9	0.20	4.91	0.96	3.09	0.01	0.88	1.10	9.17	Clinohumite
149037	Franklin	Associated with jerrygibbsite, leucophoenicite, franklinite, zincite, and willemite.																	
C6992	Franklin	Associated with franklinite and zincite.																	
JEM 3068	Franklin	Associated with franklinite, zincite, and minor calcite and willemite.																	
R1803	Franklin	Associated with franklinite and zincite.																	
C1412-1	Franklin	Associated with franklinite, zincite, manganosite, and leucophoenicite.																	
14152	Franklin	Associated with franklinite, zincite, and willemite.																	
JEM 3587	Franklin	Associated with franklinite, manganosite, zincite, and leucophoenicite.																	
HAUCK.FMM	Franklin	Associated with willemite.																	
C2828-1	Franklin	Associated with willemite and minor franklinite in vein assemblage.																	
13333	Sterling Hill	Associated with zincite, franklinite, calcite, and tephroite (rimmed by sonolite).																	
12965	Franklin?	Associated with zincite, franklinite, calcite, and tephroite (rimmed by sonolite).																	
R3522	Franklin?	Associated with zincite, franklinite, calcite, and tephroite.																	
S-K	Sterling Hill	Associated with zincite and willemite in a calcite-dominant rock.																	

lin. The samples are characterized by primary, varied, relatively simple assemblages of several species which may include manganosite, willemite, sonolite, hetaerolite/franklinite exsolution intergrowths, jerrygibbsite, leucophoenicite, zincite (usually with abundant hetaerolite exsolution), and a relative scarcity of carbonates. Specific assemblages are noted in Table 4. These sonolites have near end-member compositions with minor solid solution of other octahedral cations for Mn. Of special interest is the relative constancy of zinc (averaging 0.33 Zn per 4 Si), a feature noted herein in leucophoenicite and alleghanyite. Also notable is the relatively high calcium content of several samples from a rather simple assemblage, in which approximately 0.5 of the 9 octahedral cations are calcium.

The second group of analyses represents euhedral crystals from secondary seams and varied vein assemblages at Sterling Hill. The crystals form on calcite-rich franklinite/willemite ore with no associated zincite. There is minor secondary sphalerite. The sonolite crystals are complexly formed, of prismatic habit, and relatively abundant inasmuch as several hundred specimens have been preserved. Representative SEM photomicrographs of several of these crystals are shown in Figures 1 and 2. Crystals from this assemblage were studied by White and Hyde (1982), and they found sample 143755 to consist of crystals which were either perfect or had a moderate amount of faulting.

The chemical composition of these crystals is remarkably similar in several respects to that of the ordered alleghanyite (Francis, 1985a) discussed above. The Mg and Zn contents per 4 Si compare closely with those given per 2 Si for alleghanyite, suggesting that this Mg:Zn:Mn ratio might also be ordered and responsive to P - T conditions inasmuch as both the ordered magnesian alleghanyite and these magnesian sonolites are found in secondary vein assemblages. Because the space groups of alleghanyite and sonolite are the same, such possible ordering in sonolite may be based on a similar scheme as that noted by Francis (1985a) for Sterling Hill alleghanyite. In addition, the rela-

tively invariant Zn content of these sonolites is consistent with that of the more Mn-rich samples found at Franklin.

The third group of analyses represents a very interesting assemblage initially reported by Cook (1969). None of the samples studied herein had zinc contents approaching that reported by Cook (17.6 wt.% ZnO), using XRF analysis. Sonolite occurs as dark brown reaction rims on zincian tephroite which has abundant willemite exsolution. The associated minerals are zincite, franklinite, willemite, and calcite. The composition of this tephroite (average for #13333 and #12965) is SiO_2 31.6, FeO 1.8, MgO 13.4, ZnO 7.9, MnO 45.7, CaO 0.3, sum = 100.7 wt.%, corresponding to an octahedral cation ratio of $\text{Mn}_{1.22}\text{Mg}_{0.63}\text{Zn}_{0.18}\text{Fe}_{0.05}\text{Ca}_{0.01}$, similar in zinc content to the analysis of similar material given by Francis (1985b). Not all material of this composition occurs as mantling on tephroite; such mantling was not observed on sample RB1040 or sample S-K. The mantling of tephroite by sonolite is interesting in that a similar reaction, in isostructural phases, was noted by Mitchell (1978) who found titanian clinohumite reaction rims on forsterite in kimberlites from the Jacupiranga carbonatite in Brazil. Because the ratios of octahedral cations in these sonolite rims are very similar to those of the underlying tephroite, the mantling may have occurred as a result of hydration of the primary tephroite (personal communication, Carl Francis).

Compositionally, these sonolite reaction rims are characterized by different Mg:Zn:Mn ratios than those of the previous two groups. Magnesium is more than doubled in content relative to the previous group (e.g.: an average of 3.44 Mg compared with an average of 1.45 Mg in group 2). A second feature of this cluster is the amount of Zn (0.96 atom average) per 9 octahedral cations. The constancy of composition of these rims from at least three varied parageneses suggests that these samples may represent, like the second group, a possible stable ordering of Mn, Mg, and Zn between sonolite and clinohumite.

The last cluster of analyses is of clinohumites. The sam-

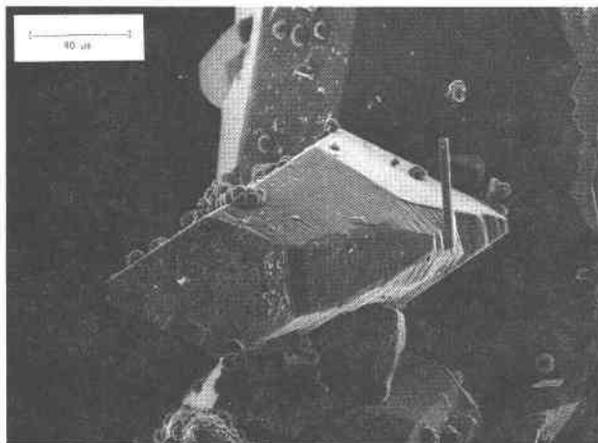


Fig. 1. Simple prismatic habit of magnesian sonolite from Sterling Hill, New Jersey. Scale bar is 40 μm .

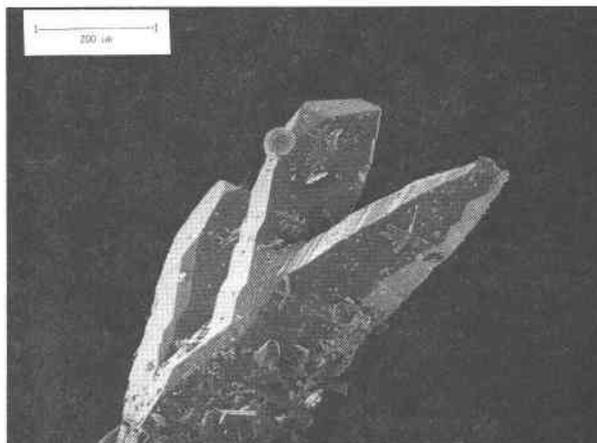


Fig. 2. Twinned magnesian sonolites from Sterling Hill, New Jersey. Scale bar is 200 μm .

ples come from one part of the Sterling Hill mine and are quite varied in texture, consisting of massive aggregates, veins, and disseminated blebs of brown clinohumite. Associated phases are zincite, franklinite, willemite, and abundant calcite. Mantling such as described above is present on several samples, but the mantled phase was not analyzed; it may be forsterite. Samples S-V and S-R are notable in that the clinohumite is surrounded by reaction rims of willemite which can be observed to have consumed clinohumite.

General observations on sonolite

In samples from Franklin and Sterling Hill, the octahedral cations occur in two apparent compositional clusters that may indicate special ordering schemes. These are: (a) with Mg:Zn:Mn approximately = 1.5:0.3:7.2, and (b) with Mg:Zn:Mn approximately = 3.4:1.0:4.6. Zinc is apparently limited to different degrees in all analyses. Iron is more abundant in clinohumite, and calcium is more abundant in sonolite; the latter likely due to cation radii requirements. Unlike the sonolite studied by Kato (*in Ribbe*, 1982), Franklin and Sterling Hill sonolites do have appreciable fluorine; it replaces up to 44 mol% of the possible (OH), increasing with the Mg content.

Leucophoenicite

Introduction

Leucophoenicite, $(\text{Mn,Zn})_7(\text{SiO}_4)_3(\text{OH})_2$, was originally described from Franklin, New Jersey, by Penfield and Warren (1899). Subsequent studies of its morphology were published by Palache (1928, 1935), and Moore (1967). The crystal structure was solved by Moore (1970) who noted that it has edge-sharing, half-occupied, silicate tetrahedra and is structurally distinct from the humite-group minerals, with which it has strong compositional similarity and is frequently associated. Recently, White and Hyde (1983a, 1983b), using TEM techniques, showed that leucophoenicite is a member of a family of structures (including some borates and germanates), and supported Moore's (1970) proposal of edge-sharing, half-occupied silicate tetrahedra. The recent discovery of jerrygibbsite, a possible polymorph of $\text{Mn}_9(\text{SiO}_4)_4(\text{OH})_2$, (Dunn et al., 1984) led those authors to speculate that there might be additional members of the leucophoenicite family, and that jerrygibbsite might be one of these.

Description

Until recently, leucophoenicite was known only from the zinc deposit at Franklin, New Jersey; it has never been found at the genetically related Sterling Hill deposit, just a few km distant, although bulk mineralogies are very similar at both localities. White and Hyde (1983b) have now shown that leucophoenicite occurs at Pajsberg, Sweden, and Winter et al. (1983) have noted that a calcian alleghanyite originally described by Dal Piaz et al. (1979) is leucophoenicite based on X-ray powder diffraction data. Because many of the uncommon minerals at Franklin

occur in restricted assemblages, several hundred leucophoenicite samples were examined. The results of this comparison supported the preliminary findings of Dunn et al. (1984) that leucophoenicite is indeed widespread at Franklin and occurs in a wide variety of assemblages, most of which have Ca-bearing associated minerals. A very small number of leucophoenicites occur without calcium-bearing species. These Ca-poor leucophoenicites are very uncommon; they occur in two types of assemblages:

1. With franklinite, willemite, and zincite in assemblages usually devoid of other associated phases. If other silicate phases are present, they are tephroite, sonolite, or jerrygibbsite.

2. With manganosite, zincite, and hetaerolite, in samples which are 80–95% manganosite (MnO) by bulk volume. This assemblage was examined in detail in search of the Mn-analogue of norbergite (MAN), which remains unknown in nature. The silicate phases found in this assemblage were tephroite, sonolite, and Ca-poor leucophoenicite. Leucophoenicite is the dominant silicate in this assemblage.

Chemical composition

The analyses shown in Table 5 were chosen to show the extent of compositional variation. Examination of these data permits a number of observations.

1. Most leucophoenicites are highly calcic. Of the 27 analyses given, 22 have Ca values in excess of 0.48 Ca per 3 Si. It should be emphasized, however, that this ratio of calcic samples is not reflective of the general ratio of calcic to non-calcic samples; calcic material is *much* more common. No samples contained the very high (up to 14 wt.%) CaO values reported by Cook (1969), using XRF analysis. The abundance of samples with values of 0.5–0.7 Ca per 7 octahedral cations suggests that this is a somewhat "stable" calcium content for samples which form in calcic assemblages.

2. Zn is a *constant* constituent of leucophoenicite. It is present in all analyses of leucophoenicite obtained by this writer, including many not published here. It is relatively invariant, amounting to approximately 0.3 Zn per 3 Si. In the last three analyses (Table 5), which are the most Mn-deficient, zinc is slightly higher. However, no Franklin leucophoenicite was found which was not Zn-bearing. Zinc was not reported in the Pajsberg, Sweden, leucophoenicite studied by White and Hyde (1983b). However, their analysis of sample C6800 from Franklin showed only a trace of zinc whereas several analyses performed as part of this study of several crystals of this same sample showed it to be homogeneous and contain 3.5–3.6 wt.% ZnO, suggesting perhaps there was undetected Zn in White and Hyde's Pajsberg material. The occurrence noted by Dal Piaz et al. (1979) from the Valsesia-Valtournanche area in the Italian western Alps, was shown by Winter et al. (1983) to be leucophoenicite, but it has uncertain composition. The constancy of Zn in all the samples studied herein suggests that either it may be essential to the species, or that there are limits on the amount of Zn permitted in leucophoenicite,

Table 5. Microprobe analyses of leucophoenicite, in order of decreasing Mn content; octahedral cations are calculated on the basis of Si = 3.

Sample #	SiO ₂	MgO	CaO	ZnO	MnO	H ₂ O*	Total	Si	Mg	Ca	Zn	Mn	ΣM ²⁺
JEM 3126	25.6	1.4	0.5	4.3	65.8	2.6	100.2	3.00	0.24	0.06	0.37	6.53	7.20
JEM 3126	25.9	2.1	0.4	4.1	65.6	2.6	100.7	3.00	0.36	0.05	0.35	6.44	7.20
47909	26.0	0.2	1.6	3.8	64.9	2.6	99.1	3.00	0.03	0.20	0.32	6.34	6.89
C6238	26.0	1.7	0.0	4.1	64.6	2.6	99.0	3.00	0.20	0.00	0.35	6.31	6.95
JEM 3136	26.0	2.6	1.2	4.1	64.1	2.6	100.6	3.00	0.45	0.15	0.35	6.27	7.22
C6882	26.4	1.0	4.9	2.9	63.4	2.6	101.2	3.00	0.17	0.60	0.24	6.10	7.11
C1415-7	25.9	0.3	3.9	4.1	63.3	2.6	100.1	3.00	0.05	0.48	0.35	6.21	7.09
144658	25.8	0.8	5.3	3.1	62.8	2.6	100.4	3.00	0.14	0.66	0.27	6.19	7.26
C6878	26.7	0.6	5.5	3.1	62.7	2.7	101.3	3.00	0.10	0.66	0.26	5.97	6.99
135739	26.2	1.2	4.6	3.3	62.4	2.6	100.3	3.00	0.20	0.56	0.28	6.05	7.09
JEM 3137	26.2	1.2	5.5	3.4	62.3	2.6	101.2	3.00	0.20	0.67	0.29	6.04	7.20
C6800	26.2	1.2	4.6	3.6	62.2	2.6	100.4	3.00	0.20	0.56	0.30	6.03	7.09
C6880	25.5	1.1	4.5	3.5	62.0	2.5	99.1	3.00	0.19	0.57	0.30	6.18	7.24
JEM 3134	26.6	2.2	4.2	3.9	61.5	2.7	101.1	3.00	0.37	0.51	0.32	5.88	7.08
95120	26.0	2.0	4.2	3.9	61.4	2.6	100.1	3.00	0.34	0.52	0.33	6.00	7.19
C6237	25.9	0.4	5.3	4.5	61.1	2.6	99.8	3.00	0.07	0.66	0.39	6.00	7.12
JEM 3113	26.0	2.2	4.2	3.9	61.0	2.6	99.9	3.00	0.38	0.52	0.33	5.96	7.19
JEM 3128	26.2	0.7	5.7	5.1	61.0	2.6	101.3	3.00	0.12	0.70	0.43	5.92	7.17
R3878-1	26.9	1.6	5.6	3.8	60.9	2.6	101.4	3.00	0.27	0.67	0.31	5.75	7.00
JEM 3098	26.3	0.6	6.2	3.7	60.5	2.6	99.9	3.00	0.10	0.76	0.31	5.84	7.01
C2920	26.1	0.5	7.0	2.7	60.4	2.6	99.3	3.00	0.09	0.86	0.23	5.88	7.06
116857	27.0	1.7	5.6	3.6	59.7	2.7	100.3	3.00	0.28	0.67	0.30	5.62	6.87
84964	27.0	3.3	4.7	4.3	59.5	2.7	101.5	3.00	0.55	0.56	0.35	5.60	7.06
JEM 3132	26.1	2.0	5.7	3.7	59.0	2.6	99.1	3.00	0.34	0.70	0.31	5.74	7.09
JEM 3135	26.5	2.4	5.3	5.2	58.4	2.6	100.4	3.00	0.41	0.64	0.43	5.60	7.08
149543	26.4	2.4	5.3	5.6	57.5	2.6	99.8	3.00	0.41	0.64	0.47	5.54	7.06
R6602-1	26.4	3.1	6.6	4.4	56.3	2.6	99.4	3.00	0.53	0.80	0.37	5.42	7.12

* - H₂O calculated based on 2(OH) per 3.00 Si.
Accuracy of data: ±3% of the amount present.

Fe and F present only as traces or absent.

or both. No samples contained the very high zinc content (up to 8 wt.% ZnO) reported by Cook (1969) using XRF analysis.

3. Fluorine is essentially absent in leucophoenicite. Some samples have traces of F, but such traces were well within the error of microprobe determinations.

The available evidence strongly suggests that Ca, Zn, and F/OH may play critical roles in the composition of leucophoenicite. Because most samples are highly calcic, the implication is that Ca might be a cation of preference for leucophoenicite. One might speculate that it should be ordered, inasmuch as Ca is ordered in all olivine-related structures investigated to date (Lumpkin et al., 1983; Ribbe, 1982). The affinity of Ca for leucophoenicite remains enigmatic.

The constancy of Zn, at least in Franklin samples, remains uninvestigated. The available evidence suggests that Zn may play some role in the formation of leucophoenicite and the stability of this phase relative to members of the humite group, where bulk rock compositions are zincian. Similarly, the absence of fluorine might indicate that OH/F ratios affect the selective formation of leucophoenicite relative to the F-bearing manganese humites. These lines of reasoning would apply equally to jerrygibbsite.

Jerrygibbsite

Jerrygibbsite, ideally (Mn,Zn)₉(SiO₂)₄(OH)₂, was first described from Franklin by Dunn et al. (1984). Subsequent to

the original description, several additional samples were found and studied. Their parageneses are similar in several respects to the original samples, i.e.: they are simple assemblages consisting of only zincite, willemite, tephroite, and franklinite, or leucophoenicite, and are notable for the lack of any Ca-bearing species. However, of the five known jerrygibbsite samples, four are texturally distinct from each other, suggesting they were to some extent spatially distributed in the Franklin orebody. Zincite is present in all samples.

Microprobe analyses of jerrygibbsite are presented in Table 6; the two analyses previously published by Dunn et al. (1984) are included for comparison. Like leucophoenicite, jerrygibbsite appears to be a phase which might have essential zinc. It is noteworthy that Mn does not exceed 32 of the possible 36 octahedral cations (full cell contents with Z = 4), and that Zn approximates 2 atoms in the full cell. The apparently low Mg content of both leucophoenicite and jerrygibbsite is noteworthy, especially because Mg is easily accommodated in the Mn-humites.

General observations on specific cations

Manganese

No end members are found among the Mn-humite species at Franklin or Sterling Hill, in large part due to ubiquitous Zn substitution and abundant Mg in many samples. Manganese is restricted in leucophoenicite (5.42–

Table 6. Microprobe analyses of jerrygibbsite, in order of decreasing Mn content; octahedral cations are calculated on the basis of Si = 4.

Sample #	SiO ₂	FeO	MgO	CaO	ZnO	MnO	H ₂ O	Total	Fe	Mg	Ca	Zn	Mn	H	ΣM^{2+}
C1417	26.9	0.2	1.1	2.0	5.3	62.3	2.0	99.8	0.02	0.24	0.32	0.58	7.85	1.98	9.01
C6991	26.9	0.2	3.0	1.6	3.2	61.8	2.0	98.7	0.02	0.67	0.25	0.35	7.79	1.98	9.08
R18772	27.1	0.3	1.4	0.4	3.9	64.1	2.13*	99.3	0.04	0.31	0.06	0.43	8.02	2.10	8.86
C3209	26.6	0.3	1.1	1.0	5.3	62.1	2.25*	98.6	0.04	0.25	0.16	0.59	7.91	2.26	8.95
Average/4									0.03	0.37	0.19	0.49	7.89		8.97

* analyses from Dunn et al., 1984; water by Penfield method.

6.53 Mn per 7 octahedral cations) and jerrygibbsite (7.79–8.02 Mn per 9 octahedral cations).

Calcium

Calcium is common to both jerrygibbsite and leucophoenicite, but among the humites only sonolite accepts appreciable Ca (up to 0.65 Ca per 9 octahedral cations).

Iron

The Mn-humites at these localities have minimal Fe substitution. Clinohumite accepts up to 0.2 Fe per 9 octahedral cations.

Magnesium

All the Mn-humites contain Mg, which apparently generates several ordered intermediate phases. Magnesium is apparently quite restricted in both leucophoenicite and jerrygibbsite.

Zinc

There is a "threshold" level of 0.2–0.4 Zn atoms per xSi in all the studied samples (x = 2 for alleghanyite/chondrodite, 3 for manganhumite/humite/leucophoenicite, and 4 for sonolite/clinohumite/jerrygibbsite). In both leucophoenicite and alleghanyite, this "threshold" level is relatively constant for all studied samples. Zn is apparently ordered in some Mg-bearing alleghanyites and sonolites. No samples are zinc-free.

Fluorine

Both jerrygibbsite and leucophoenicite are fluorine-free. Fluorine is common to the Mn-humites at Franklin and Sterling Hill, and is roughly proportional to the Mg-content.

It should be emphasized that many of these chemical features may be solely responsive to conditions at Franklin and Sterling Hill.

Unresolved matters

The crystal-chemical role of Zn in the Mn-humites is deserving of careful investigation. Not only do both sonolite and alleghanyite exhibit two apparent chemical clusters each, but it appears that these are related to not only

Mn:Mg ratios, but at Franklin, also to the Zn content. Although alleghanyite and sonolite are both known from other deposits as zinc-free phases, neither leucophoenicite nor jerrygibbsite has been shown to be zinc-free. Hence, leucophoenicite and jerrygibbsite may be characterized by essential zinc, or some limitation on the amount of Mn permitted, or other factors, such that these phases might not be stable in nature as manganese end-members.

In addition, the equilibrium relations which favor the formation of leucophoenicite or jerrygibbsite rather than the manganese humites remain unknown. Similarly, the effects of zinc on the phase relations for the Mn-humites remain unstudied, and very little is known of the partitioning of cations among the co-existing Mn-humites.

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