

## **Glaucochroite (olivine, $\text{CaMnSiO}_4$ ) from Franklin, New Jersey: Its composition, occurrence, and formation**

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### **ABSTRACT**

Glaucochroite,  $\text{CaMnSiO}_4$ , is a member of the olivine group, known primarily from the Zn-Mn-Fe deposit at Franklin, New Jersey. Euhedral crystals occur in a number of assemblages with nasonite, willemite, clinohedrite, hardystonite, diopside, and cuspidine. Massive, coarse-grained, blue glaucochroite occurs with willemite, franklinite, calcite, hardystonite, leucophoenicite, and andradite. Massive, fine-grained brown glaucochroite occurs with esperite, hodgkinsonite, calcite, willemite, zincite, and franklinite, and as "calctephroite," an inhomogeneous, sheared or altered material. Glaucochroite was not observed in association with tephroite, rhodonite, or wollastonite, all of which occur at Franklin.

Fourteen glaucochroite analyses conform closely to the ideal formula, with little solid solution toward tephroite. Glaucochroite can form either by the heterogeneous reaction calcite + bustamite + tephroite = glaucochroite +  $\text{CO}_2$  or by gradual extension of olivine composition from tephroite to glaucochroite above the olivine solvus. At Franklin, glaucochroite is interpreted as forming instead of the commoner assemblage rhodonite + calcite in local areas with high activity of water and low activity of carbon dioxide, but under the same temperature and pressure conditions as the deposit as a whole.

Cuspidine,  $\text{Ca}_4\text{Si}_2\text{O}_7(\text{F},\text{OH})_2$ , is here confirmed as occurring at Franklin. It occurs with glaucochroite and hardystonite, and in solution vugs in willemite. Two microprobe analyses averaged  $\text{SiO}_2$  32.6, CaO 62.3, F 9.7, less O = F 4.1, total 100.5 wt%, conforming closely to the ideal composition.

### **INTRODUCTION**

Glaucochroite,  $\text{CaMnSiO}_4$ , was described by Penfield and Warren (1899) as minute blue crystals embedded in nasonite from the Franklin, Sussex County, New Jersey, Zn-Fe-Mn deposit. Palache (1935) described glaucochroite in "a coarse granular form of bluish color, intimately mixed with willemite, hardystonite, tephroite, and franklinite." More recently, glaucochroite has been found in a skarn formed at the contact between dolerite and marble in Anakit, Lower Tunguska, USSR (Pertsev and Laputina, 1974), and in calc-silicate rocks associated with manganese oxide ores in the Wessels mine, South Africa (R. D. Dixon, pers. comm.).

### **OCCURRENCE**

Frondel and Baum (1974) described the structure and mineralogy of the Franklin deposit, based on level maps of the mine prepared during mining operations. The deposit is a complex metasedimentary unit containing both Zn-rich ore units and Zn-poor calcium silicate skarns.

The deposit has been highly deformed and metamorphosed at temperatures of at least 650–750°C (Frondel and Klein, 1965) and pressures of several kilobars, based on the geology of the surrounding rocks (Hague et al., 1956).

The Franklin mine closed in 1954 and is now flooded. Assemblages can only be studied in hand specimens, most lacking reference to location or petrologic relationships. There are a number of references to glaucochroite on the mine-level maps, which suggest that it was widely distributed in the northern parts of the mine.

### **EUHEDRAL CRYSTALS OF GLAUCOCHROITE**

The initial discovery of glaucochroite (Penfield and Warren, 1899) was of euhedral crystals collected from the dump of the Parker shaft near the end of the last century, with other rare minerals, including nasonite and other lead silicates. Because the preserved samples of euhedral glaucochroite are few in number and their parageneses are only generally in agreement with the original descrip-

tion and that of Palache (1935), we gathered together the 15 available specimens of glaucochroite crystals, and they permitted the recognition of several distinct assemblages:

1. Glaucochroite occurs embedded in nasonite, Pb<sub>6</sub>Ca<sub>2</sub>Si<sub>6</sub>O<sub>21</sub>Cl<sub>2</sub>, with minor amounts of primary franklinite and andradite, and abundant light-yellow andradite, which surrounds the primary material. Present only in very minor amounts are barite, clinohedrite, CaZnSiO<sub>3</sub>(OH)<sub>2</sub>, willemite, and a late-stage Mn-chlorite (NMNH B21241, C2799).

2. Glaucochroite occurs embedded in massive green willemite that encrusts common, granular willemite-franklinite ore. Both andradite and hodgkinsonite, MnZn<sub>2</sub>SiO<sub>4</sub>(OH)<sub>2</sub>, form at the interface between the ore and the green willemite, suggesting a vein assemblage. Clinohedrite intergrown with the willemite has a vuggy texture suggesting later crystallization. Cuspidine occupies solution vugs in willemite. Barite is present in minor amounts. (NMNH B21402.)

3. Massive, white clinohedrite occurs associated with andradite and hardystonite, Ca<sub>2</sub>ZnSi<sub>3</sub>O<sub>7</sub>. Glaucochroite crystals occur within both hardystonite and clinohedrite. Andradite of several generations is present, encloses franklinite (sparse), and is older than the hardystonite + clinohedrite + glaucochroite assemblage. Some specimens have a breccia-like texture wherein garnet is cemented with the glaucochroite-bearing assemblage. Cuspidine is present on two specimens, in 2–3-mm crystals, and in apparent chemical equilibrium with the glaucochroite assemblage. (NMNH 93032, 113684, C2798.)

4. Massive willemite occurs with 1.0-cm white diopside crystals, and slightly pinkish-blue massive glaucochroite. Clinohedrite, nonfluorescent calcite, and stilpnomelane are minor phases. This assemblage is vuggy, and euhedral glaucochroite crystals line the vugs.

In most of these assemblages, glaucochroite forms bladed crystals up to 1 cm in length, frequently in columnar bundles. Assemblage 1 was that originally described by Penfield and Warren (1899). Assemblages 2 and 3 were apparently known to Palache (1935), but not described in detail. Assemblage 3 was found in the Yale University collection; the labels were in Penfield's handwriting, and the samples are undoubtedly from the turn-of-the-century finds. The axinite reported by previous investigators in this association is probably andradite.

The association of the high-temperature mineral glaucochroite with clinohedrite is anomalous, as clinohedrite is typical of altered and recrystallized assemblages. It frequently forms from hydrothermal alteration of hardystonite, a member of the melilite group. However, several samples show that the association of euhedral glaucochroite crystals with clinohedrite results from the alteration of hardystonite, the primary host for glaucochroite, to clinohedrite, particularly in assemblage 3.

#### CUSPIDINE

The presence of cuspidine in several of these assemblages is notable. Cuspidine, Ca<sub>4</sub>Si<sub>2</sub>O<sub>7</sub>(F,OH)<sub>2</sub>, was re-

ported from Franklin by Palache (1910, 1935) as a result of his interpretation of an 1899 analysis by C. H. Warren of colorless crystals associated with nasonite, but samples of this material have been unknown since then. Cuspidine forms colorless, clear, distorted crystals. The density is between 2.965 and 2.989 (Palache, 1910). Franklin cuspidine is readily recognized by its moderately strong yellow-to-light violet fluorescence in long-wavelength ultraviolet. The fluorescence in short-wavelength ultraviolet is similar, but weaker. Cuspidine, like glaucochroite, has not been found at Sterling Hill.

Microprobe analyses of Franklin cuspidine, which are in reasonably good agreement with that of Warren, yielded SiO<sub>2</sub> 32.3, 32.8; CaO 63.5, 61.1; F 9.8, 9.5; less O = F 4.1, 4.0; totals 101.5, 99.4 wt% and conform closely to the ideal composition. Substitution of (OH) for F is minimal in Franklin cuspidine. Cuspidine is in apparent chemical equilibrium with glaucochroite, which is younger, and hardystonite, which is altered in some specimens to clinohedrite. Cuspidine also occurs, within assemblages of glaucochroite crystals, as a late-stage mineral, forming druses with clinohedrite in solution vugs in willemite.

#### MASSIVE GLAUCOCHROITE

In the ores and calc-silicate rocks, several different massive glaucochroite-bearing assemblages may be distinguished. On the basis of textures of hand specimens, there are four assemblages containing coarse-grained blue glaucochroite and two containing fine-grained brown glaucochroite. The coarse-grained assemblages are as follows:

1. Bright-green willemite, blue glaucochroite, and franklinite, occur in octahedra up to 2 cm. All three species are always present; hardystonite and calcite are common but not always present. Leucophoenicite, (Mn,Zn)<sub>7</sub>Si<sub>3</sub>O<sub>12</sub>(OH)<sub>2</sub> and zincite are sparse. (NMNH 138406, 138407, 144684, R3494, and others in the Harvard and Delaware collections.)

2. Blue glaucochroite occurs with willemite and franklinite and leucophoenicite. Hardystonite and calcite are absent or present only in traces. One specimen of the assemblage (JEM 3107) shows in hand specimen blue glaucochroite grading into pink leucophoenicite over a distance of about 4 cm in a matrix of gneissic franklinite with minor willemite and sparse native copper. Average grain size is about 1–3 mm. In thin section, the specimen shows a granoblastic texture of the glaucochroite, leucophoenicite, and willemite, suggesting an equilibrium intergrowth of glaucochroite and leucophoenicite rather than replacement of one by the other. (NMNH 107374, 147244, C6172, R6602.)

3. Andradite, willemite, glaucochroite, and calcite occur in a coarse granular intergrowth. This assemblage was seen in three specimens, Harvard #114290 and two specimens in the Bostwick collection.

4. Andradite, bustamite, (Mn,Ca)<sub>3</sub>Si<sub>3</sub>O<sub>9</sub>, glaucochroite, and willemite make up the fourth assemblage; the an-

dradite and bustamite grains are up to 2 cm across. (Harvard 114190, Hauck 213, and two uncataloged specimens in the Delaware collection.)

In the fine-grained assemblages, glaucochroite is brown, with a fine granular texture. Other minerals may be coarser grained. There are essentially two fine-grained assemblages:

1. Brown glaucochroite occurs with esperite, (Ca,Pb)ZnSiO<sub>4</sub>, hodgkinsonite, calcite, willemite, zincite, and franklinite. Glaucochroite is frequently intimately associated with hodgkinsonite, and hodgkinsonite occurs at the esperite-glaucochroite interfaces. Esperite forms irregular segregations, and specimens frequently show shearing of the entire assemblage. In a few specimens, irregular, 3–5 mm, possibly remnant masses of blue glaucochroite are embedded in the brown glaucochroite.

2. "Calctephroite." Palache (1928) reported that "material known locally as 'calctephroite' was found abundantly at Franklin about 1924 . . . It is very dense and fine-grained, white when first taken from the mine but soon turning dark brown upon exposure to the light. It appears to be a very impure variety of glaucochroite." The analysis that he included conforms to glaucochroite with about 3% each of ZnO and MgO. Under the microscope, the "calctephroite" is fine grained and in microprobe analysis proved to be extremely inhomogeneous on a small scale. An X-ray powder-diffraction pattern of "calctephroite" shows it to be glaucochroite. The specimens in general give the impression of having been crushed or sheared; "calctephroite" is merely a sheared and/or altered glaucochroite.

#### CHEMICAL ANALYSIS

Fifteen samples of glaucochroite from the collections of the Smithsonian Institution were chemically analyzed using an ARL-SEM microprobe with an operating voltage of 15 kV and a beam current of 0.15 μA. The standards were rhodonite for Mn and Zn, wollastonite for Si and Ca, and hornblende for Mg and Fe. The data were corrected using a modified version of the MAGIC-4 computer program. The analyses are given in Table 1, along with molecular proportions calculated on the basis of four oxygens.

Brown (1982) found that glaucochroite from Franklin is highly ordered; he gives the structural formula (Ca<sub>0.98</sub>Mn<sub>0.02</sub>)(Mn<sub>0.85</sub>Mg<sub>0.10</sub>Zn<sub>0.05</sub>)SiO<sub>4</sub>, which is consistent with the findings of Francis and Ribbe (1980), who showed that Mg and Zn in tephroite strongly prefer the smaller M1 octahedral site in the olivine structure.

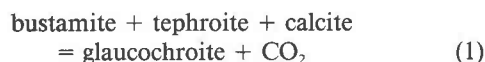
Although the samples include five of crystal occurrences and nine massive samples of varying colors and textures, they show little chemical variation. The maximum solid solution toward tephroite is 6 mol% shown by several glaucochroites, including C6172, associated with leucophoenicite. Although Francis (1985) found a complete range of tephroite-forsterite solid solutions, the maximum glaucochroite-monticellite solid solution is 12 mol% (R6602), and most samples are much lower. Zn

varies between 0.02 and 0.05 atoms and is probably buffered at a maximum value by coexisting willemite. No exsolution of willemite from glaucochroite was seen, although it has been observed in tephroite (Francis, 1985) and a number of other Franklin silicates (Fronzel, 1972).

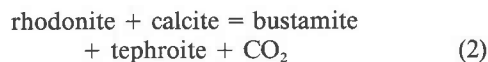
As a further attempt to find solid solution between tephroite and glaucochroite from Franklin, we surveyed a large number of samples of both minerals by optical oil-immersion methods. No samples showed intermediate optical properties that would indicate intermediate compositions.

#### FORMATION OF GLAUCOCHROITE

Tilley (1946) suggested that glaucochroite would form by the reaction



at a higher temperature than the reaction



These reactions were based on the mineralogy of the Treburland mine, Cornwall, England, where bustamite + tephroite is a typical assemblage but glaucochroite is absent. The stable assemblages predicted by Tilley's reaction are glaucochroite + bustamite + calcite, glaucochroite + tephroite + calcite, and glaucochroite + tephroite + bustamite.

Glaser (1961) studied the crystallization of melts in the system Ca<sub>2</sub>SiO<sub>4</sub>-Mn<sub>2</sub>SiO<sub>4</sub>; the system has a minimum in the liquidus curve about halfway between glaucochroite and tephroite compositions but shows complete solid solution between tephroite and an olivine even richer in Ca than glaucochroite composition. Glaser suggested that a complete range of compositions from tephroite to olivines more calcic than glaucochroite might be found in nature. From these data, Burt (1972) suggested that glaucochroite could form by the gradual extension of tephroite to more calcic composition at high grades of metamorphism at low to moderate pressures, but he suggested that at high oxygen fugacities "the solid solution is probably broken by tie lines between oxide phases [such as franklinite] and phases along the pyroxene join. This relationship would account for the separate occurrence of glaucochroite and tephroite at Franklin, New Jersey."

Mukhopadhyay and Lindsley (1983) investigated subsolidus relationships in the analogous system kirschsteinite (CaFeSiO<sub>4</sub>)-fayalite (Fe<sub>2</sub>SiO<sub>4</sub>), which also has a minimum in the liquidus curve about halfway between the two species. They inferred that this minimum "indicates that there should be a miscibility gap in the subsolidus region of the system," and they located and mapped such a gap by hydrothermal experiments. A similar gap presumably exists between glaucochroite and tephroite.

In the present study, the assemblage glaucochroite-bu-

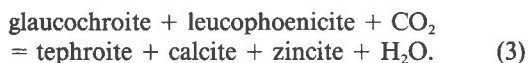
TABLE 1. Microprobe analyses of Franklin, New Jersey, glaucochroite with cations calculated on the basis of four oxygens

NMNH#	SiO <sub>2</sub>	MgO	CaO	ZnO	MnO	Total	Texture or assemblage
B21241	32.0 1.01	0.2 0.01	29.7 1.00	0.7 0.02	35.9 0.96	98.5	crystal 1
B21402	32.8 1.01	0.4 0.02	29.2 0.96	0.8 0.02	37.4 0.98	100.6	crystal 2
113685	32.1 1.01	0.3 0.01	29.4 0.99	0.8 0.02	36.6 0.97	99.2	crystal 3
C2798	32.2 1.00	0.2 0.01	29.5 0.98	0.8 0.02	37.5 0.99	100.2	crystal
93032	32.5 1.01	0.2 0.01	29.5 0.98	0.8 0.02	36.9 0.97	99.9	crystal
R3494	33.1 1.01	2.4 0.11	30.7 1.00	1.8 0.04	32.6 0.84	100.6	coarse 1
138406	32.8 1.01	0.6 0.03	29.5 0.97	1.7 0.04	36.8 0.96	101.4	coarse 1
138407	32.6 1.01	0.5 0.02	28.5 0.94	1.8 0.04	37.2 0.97	100.6	coarse 1
144684	32.9 1.01	0.8 0.04	28.8 0.95	1.6 0.04	37.1 0.96	101.2	coarse 1
C6172	32.5 1.01	0.5 0.02	28.3 0.84	1.0 0.02	37.6 0.99	99.9	coarse 2
R6602	33.4 1.00	2.7 0.12	30.2 0.97	1.6 0.04	33.8 0.86	101.7	coarse 2
107374	32.6 1.01	0.2 0.01	28.8 0.96	1.5 0.03	37.3 0.98	100.4	coarse 2
147244	32.3 1.01	0.4 0.02	28.5 0.95	1.9 0.04	37.0 0.98	100.1	coarse 2
JEM 3138	32.9 1.01	0.5 0.02	28.7 0.94	2.0 0.05	37.2 0.97	101.3	coarse
	32.13*		29.97*		37.91*	100.0*	

Note: Accuracy of data:  $\pm 3\%$  of the amount present for major elements.

\* Calculated CaMnSiO<sub>4</sub>.

stamite was seen three times. Palache's report (1935) of coexisting glaucochroite and tephroite was not confirmed; it may have been a mistaken identification of the samples in which fine-grained brown and coarse-grained blue glaucochroite occur together. The association rhodonite + calcite, which Tilley (1946) did not report from Treburland, is typical at Franklin (Palache, 1935; Fron-del, 1972). Glaucochroite was not found with rhodonite or with wollastonite, another Franklin species, in this study. Glaucochroite was much less abundant than rhodonite at Franklin, but notations on the mine maps showed that it occurred in a number of places in the northern part of the deposit and was not as restricted as Palache's (1935) report suggests. It does occur commonly with leucophoenicite, (Mn,Zn)<sub>7</sub>Si<sub>3</sub>O<sub>12</sub>(OH)<sub>2</sub>, and glaucochroite plus leucophoenicite (plus carbon dioxide) are equivalent to tephroite plus calcite plus zincite (plus water), a common mineral assemblage at Franklin:



The above observations indicate that the dominant mineral association at Franklin is one in which rhodonite + calcite is stable and glaucochroite is not. This range

of conditions might correspond to temperatures below (or CO<sub>2</sub> pressures above) those of Reaction 9 of Burt (1972, p. 430), as depicted to the upper left of Figure 1, a schematic *P-T* diagram of presumed decarbonation reactions involving glaucochroite in the system CaO-MnO-SiO<sub>2</sub>-CO<sub>2</sub>. (See Burt, 1972, for other possible decarbonation reactions in this system.) Locally, glaucochroite-bearing assemblages do occur, which suggests that calcite + rhodonite have reacted to form bustamite + tephroite (Reaction 9 of Burt, 1972, and Figure 1) and then that glaucochroite has formed by consumption of calcite, bustamite, and tephroite, as originally suggested by Tilley (1946). These equilibria imply that one would never expect to find glaucochroite with rhodonite, consistent with our observations.

There are two similar but distinct ways for glaucochroite to form during metamorphism, both of which are depicted on Figure 1. At temperatures below those of the tephroite-glaucochroite solvus, it can form via Tilley's heterogeneous reaction calcite + bustamite + tephroite = glaucochroite + CO<sub>2</sub> (univariant line 9a in Fig. 1). Burt (1972) pointed out that this reaction becomes divariant at temperatures above those of the olivine solvus, inasmuch as glaucochroite then forms by the gradual leftward extension of the olivine composition due to consumption

of calcite and bustamite. This divariant reaction cannot be depicted as a univariant line on  $P$ - $T$  diagrams such as Figure 1. The corresponding univariant reaction (line 9a) dies abruptly at a critical end point, which also marks the end of the near-vertical critical line demarking the Ca-Mn olivine solvus. The univariant line can have no dashed metastable extension, whereas the critical line does. (The continuation of the critical line becomes metastable because we are projecting through CO<sub>2</sub>, the composition of the vapor phase.)

At somewhat higher temperatures or lower pressures of CO<sub>2</sub>, the assemblage bustamite + calcite becomes unstable with regard to the assemblage wollastonite + glaucochroite (or olivine solid solution at higher temperatures). This is Reaction 10 of Burt (1972) and Figure 1. It occurs on both sides of the critical line and explains the assemblage glaucochroite + wollastonite observed at the Wessels mine in South Africa (Dixon, pers. comm.). The lack of glaucochroite-wollastonite assemblages at Franklin implies that the conditions needed for this reaction were not attained during metamorphism at Franklin.

Based on available evidence, it is unclear whether glaucochroite at Franklin formed abruptly, via a univariant reaction, or gradually, by extension of tephroite compositions, as suggested by Burt (1972). The limited solid solution of glaucochroite found in this study and the limited solid solution of tephroite and glaucochroite reported by Palache (1935) and Francis (1985) would imply that the olivine solvus occurs at temperatures above those encountered during metamorphism at Franklin. Nevertheless, tephroite and glaucochroite have not yet been found together nor exsolved from each other. Furthermore, glaucochroite from Franklin generally occurs with calcic phases such as calcite, bustamite, hardystonite, clinohedrite, diopside, and cuspidine, phases which would tend to "pull" its composition to the left, making it stoichiometric CaMnSiO<sub>4</sub>, no matter what the position of the solvus (cf. Fig. 1). However, glaucochroite of stoichiometric composition does occur commonly with the manganese-phase leucophoenicite. Experimental work on the glaucochroite-tephroite solvus or a find of coexisting glaucochroite and tephroite would be needed to resolve this problem.

Tie lines between franklinite, (Zn,Mn)Fe<sub>2</sub>O<sub>4</sub> (or other Mn-bearing oxides), and bustamite may generally prevent the coexistence of tephroite with glaucochroite (or their solid solution) at Franklin, as mentioned by Burt (1972). In any case, it is clear that glaucochroite occurrences at Franklin formed under metamorphic conditions between those of Reactions 9 and 10 of Burt (1972) as shown in Figure 1.

The occurrence of the different but equivalent mineral assemblages discussed above (e.g., rhodonite + calcite vs. glaucochroite; calcite + zincite + tephroite vs. glaucochroite + leucophoenicite) suggests that metamorphic conditions at Franklin varied locally. By analogy with metamorphosed dolomites, variations in the relative mole

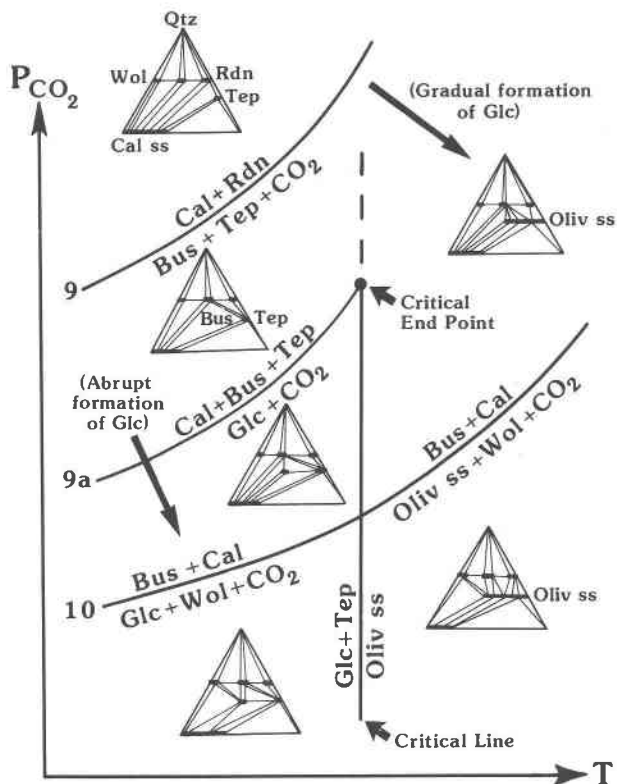


Fig. 1. Schematic  $P_{\text{CO}_2}$ - $T$  diagram showing possible reactions involving the formation of glaucochroite in the system CaO-MnO-SiO<sub>2</sub>-CO<sub>2</sub>. The system is projected through the CO<sub>2</sub>-rich vapor phase onto the CaO-MnO-SiO<sub>2</sub> plane. Reaction numbers 9 and 10 are those of Burt (1972). The diagram depicts a critical end point at the intersection of the univariant reaction Cal + Bus + Tep = Glc + Vap and the critical line marking the solvus at which olivine solid solution breaks down to Glc + Tep. At temperatures and pressures above those of this point, glaucochroite must form gradually, owing to extension of tephroite compositions to more calcic composition. The actual position of this point in  $P$ - $T$  space is not yet known. Cal = calcite, Wol = wollastonite, Qtz = quartz, Bus = bustamite, Rdn = rhodonite, Oliv = olivine, Tep = tephroite, Glc = glaucochroite, ss = solid solution.

fractions of CO<sub>2</sub> and H<sub>2</sub>O in the metamorphic pore fluid could have caused the observed assemblage variations. Inasmuch as glaucochroite is formed by reactions evolving CO<sub>2</sub>, and it is commonly associated with the hydrous species leucophoenicite, a locally higher mole fraction of H<sub>2</sub>O could have been responsible for the glaucochroite-bearing assemblages at the deposit.

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