$\label{eq:constraint} \begin{array}{l} Mengxianminite \ (Ca_2Sn_2Mg_3Al_8[(BO_3)(BeO_4)O_6]_2) \ a \ new \ borate \ mineral \ from \\ Xianghualing \ skarn, \ Hunan \ Province, \ China, \ with \ a \ highly \ unusual \ chemical \ combination \\ \ (B + Be + Sn) \end{array}$

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ABSTRACT

Mengxianminite, ideally $Ca_2Sn_2Mg_3Al_8[(BO_3)(BeO_4)O_6]_2$, is a new borate mineral from Xianghualing skarn, Hunan Province, southern China. It occurs in the hsianghualite vein of Xianghualing skarn, associated with fluorite, phlogopite, hsianghualite, magnetite, dravite, magnesiotaaffeite-2N'2S, and calcite. Mengxianminite forms subhedral to euhedral green crystals from 20 to 200 µm long, translucent to transparent, with a vitreous luster. The crystals show perfect cleavage on {100} and good cleavage on $\{010\}$, and do not fluoresce in long- or short-wave ultraviolet light. The estimated Mohs hardness is 8, and the tenacity is brittle with irregular fracture. The calculated density is 4.170 g/cm^3 . Optically, mengxianminite is biaxial (-), with $\alpha = 1.80(2)$, $\beta = 1.83(2)$, $\gamma = 1.84(2)$ (589 nm). The mean chemical composition of mengxianminite is Al₂O₃ 40.00, SnO₂ 25.96, MgO 6.57, CaO 8.56, FeO 4.83, B₂O₃ 6.53, BeO 4.37, ZnO 1.81, MnO 1.23, Na₂O 1.13, TiO₂ 0.10, SiO₂ 0.04, sum 101.12 with a corresponding empirical formula calculated on the basis of 26 O atoms of (Ca1.64, Na0.39) 52.03 (Sn1.85, Zn0.24) 52.09 (Mg1.75, $Fe_{0.72}$, $Al_{0.42}$, $Mn_{0.19}$, $Ti_{0.01}$)_{53.09} $Al_8[(B_{1.01}O_3)(Be_{0.94}O_4)O_6]_2$. (Be and B were measured by secondary ion mass spectrometry, average of six electron microprobe analyze points and in wt%.) The strongest eight lines of the powder XRD pattern [d in Å (I)(hkl)] are: 3.000 (35)(16.2.0); 2.931 (100)(17.1.1); 2.475 (29)(022); 2.430 (30)(13.3.1); 2.375 (100)(14.0.2/640); 2.028 (52)(21.3.1); 1.807 (35)(913); 1.530 (98)(14.6.0/15.3.3). Mengxianminite is orthorhombic, space group Fdd2; unit-cell parameters refined from single-crystal X-ray diffraction data are: a = 60.699(4), b = 9.914(1), c = 5.745(1) Å, V = 3457.4(4) Å³, and Z = 8. The structure of mengxianminite is characterized by the alternating O-T1-O-T2-O'-T2 layers stacked along the a axis, which are equal to two alternating modules: the module A (O-T1-O) corresponding to the spinel module with an additional O layer (AlO₆ octahedra layer), and the module B (T2-O'-T2) showing the simplified formula CaSnAl(BeO₄)(BO₃), where SnO₆ octahedra are isolated in the T2 layers, connected via BeO_4 and CaO_{11} groups, and AlO_6 edge-sharing octahedra in the O' layer form chains running along the $\{011\}$ or $\{0\overline{1}1\}$ direction, connected in the c direction by the BO₃ triangular groups. Mengxianminite is the first borate mineral with both Sn and Be, likely crystallized under F-rich conditions at late stages of the Xianghualing skarn.

Keywords: Mengxianminite, new mineral, Xianghualing skarn, Hunan province, China

INTRODUCTION

A new mineral species, mengxianminite, ideally $Ca_2Sn_2Mg_3$ Al₈[(BO₃)(BeO₄)O₆]₂, has been found in the Xianghualing sharn, Linwu County, Hunan Province (southern China). We should point out that mengxianminite was first reported as a Sn-bearing oxide from the hsianghualite vein of the Xianghualing skarn, Hunan Province, southern China, by Huang et al. (1986, 1988). However, it was not properly characterized by these authors. Moreover, it was not listed either as a valid or an invalid unnamed mineral (Smith and Nickel 2007). Since the "mengxianminite" reported by Huang et al. (1986, 1988) is from the same locality and has a similar chemical composition and crystal-structure, we have retained the name "mengxianminite" for the Be-B-Sn mineral described here.

Optical microscopy, electron probe microanalysis, secondaryion mass spectrometry (SIMS), X-ray diffraction, and Raman and infrared spectroscopy were used to determine its petrographic features, chemical composition, and crystal structure. The new mineral is named after "Meng Xianmin" (1900–1969), a famous geologist in China, who devoted himself to work on mineral identification and theory of mineralization in deposits of non-ferrous metals and made significant contributions to the research of ore deposits in China. The species and the name have been approved by the International Mineralogical Association, Commission on New Minerals, Nomenclature and Classification

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(CNMNC) (IMA 2015-70) (Rao et al. 2016). The type sample of mengxianminite, used to collect the electron-microprobe data, is stored in the Geological Museum of China, No. 16, Yangrou Hutong, Xisi, Beijing 100031, People's Republic of China. Catalog number of material is M13293. The co-type sample used for the single-crystal structure and optical measurements is stored at the Laboratory of Mineralogy, University of Liège, Belgium (catalog number 20395). This paper describes the physical and chemical properties of mengxianminite and its crystal structure determined from the single-crystal X-ray diffraction data, and discusses the origin and implications.

OCCURRENCE

Mengxianminite was found in the Xianghualing skarn, Hunan Province, southern China, which is located at longitude E 112°34', latitude northeast of 25°26', about 20 km northeastern of Linwu County. In this area, the principal unit is the Laiziling granite, which intrudes sandstone and sandy shale of Tiaomajian Formation, and limestone and dolomitic limestone of Qiziqiao Formation. A vertical zonation is well developed from the deeper level upward to the top of the Laiziling mountain peak: protolithionite granite zone, leucogranite zone, albite granite zone, greisen zone, massive quartz zone, and pegmatoid stockscheider zone. The U-Pb dating of zircon of the protolithionite granite gave 155 Ma (Zhu et al. 2011). The Xianghualing skarn was typically distributed around the Laiziling granite, and/or along the related tectonic faults, formed a tin-polymetallic (Sn-W-Be-Li) deposit, which is the type locality of hsianghualite $Ca_3Li_2Be_3(SiO_4)_3F_2$ (Huang et al. 1958), liberite Li₂Be(SiO₄) (Chao 1964), and ferrotaaffeite-2N'2S BeFe₃Al₈O₁₆ (Yang et al. 2012). The Laiziling granite with high levels of Li, Be, Sn, W, Rb, Nb, and Ta, was generally regarded as the main source of the Xianghualing orebodies. The average concentrations of Be and Sn in the Laiziling granite are 16 and 65 ppm, respectively (Zhong 2014).

Mengxianminite occurs in the hsianghualite vein of the Xianghualing skarn, which is located in the exocontact zone of the Laiziling granite and in the Middle-Upper Devonian carbonate rocks of the Qiziqiao Formation. It forms subhedral to euhedral crystals ranging from 20 to 200 μ m in size (Fig. 1). In some cases, mengxianminite was replaced by late fluorite along cleavage planes, resulting in the local precipitation of small cassiterite crystals (Fig. 1a). Other associated minerals include phlogopite, hsianghualite, magnetite, dravite, magnesiotaaffeite-2*N*'2*S*, and calcite.

PHYSICAL AND OPTICAL PROPERTIES

Mengxianminite is green and shows well-developed or perfect cleavage on {100} and good cleavage on {010} (Fig. 1b). It does not show fluorescence in long- or short-wave ultraviolet light. The crystals are translucent to transparent with vitreous luster. The Mohs hardness is about 8 by comparison with the hardness of magnesiotaaffeite-2N2S, the tenacity is brittle with irregular fracture. Based on the empirical formula and singlecrystal unit-cell parameters, the calculated density is 4.170 g/cm³. Optically, mengxian minite is biaxial negative, with $\alpha = 1.80(2)$, $\beta = 1.83(2), \gamma = 1.84(2)$ (589 nm). The calculated 2V is 60°, and optical orientation: α , β , and γ are parallel to crystallographic a, b, and c, respectively. The pleochroism is X = light green, Y = light green, and Z = colorless. According to the calculated density and the measured indexes of refraction, the compatibility index $[1 - (K_P/K_C)]$ is -0.036, and corresponds to the "excellent" category (Mandarino 1981).

Raman and infrared spectroscopies

Raman spectra of mengxianminite were collected using a LabRAM HR evolution Laser Raman microprobe in the School of Earth Sciences at Zhejiang University. A 532 nm laser with a power of 50 mW at the sample surface was used for exciting the radiation. Silicon (520 cm⁻¹ Raman shift) was used as a standard. Raman spectra were acquired from 100 to 4000 cm⁻¹ and the accumulation time of each spectrum is 60 s. The Raman spectra were obtained on single crystals of mengxianminite on polished thin section chips. It is characterized by the strong sharp peak at 754 cm⁻¹, medium sharp peaks at 1009, 806, 722, 606, and 506 cm⁻¹, and weak sharp peaks at 553 and 156 cm⁻¹ (Fig. 2). The Raman shifts at 722 and 533 cm⁻¹ show the stretching and bend-

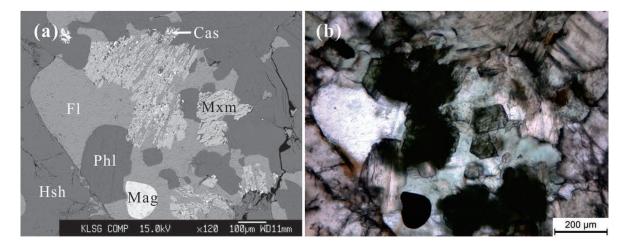


FIGURE 1. BSE image (a) and photomicrograph (b) showing the occurrence and mineral associations of mengxianminite in the Xianghualing skarn. Abbreviations: Mxm = mengxianminite, Fl = fluorite, Phl = phlogopite, Hsh = hsianghualite, Mag = magnetite, Cas = cassiterite.

ing modes of (BO_3) groups in the structure, and the Raman shift at 754 cm⁻¹ is related to the stretching mode of (AlO_6) groups. The Be-O vibration modes are probably at 1009 and 156 cm⁻¹, and the Raman shift at 606 cm⁻¹ probably corresponds to Sn-O vibration. The Ca-O and Mg-O vibration modes are probably at 806 and 506 cm⁻¹, respectively.

An infrared spectrum (400–5600 cm⁻¹) was obtained on a Nicolet iso50 FTIR spectrometer coupled with a Continuum microscope installed in the School of Earth Sciences at the Zhejiang University, using a KBr beam-splitter and a liquid-nitrogen cooled MCT-A detector. All measurements were carried out using reflection technique. The representative infrared spectrum of mengxianminite is given in Figure 3. The bands at 1426, 1328, 1155, 1050, and 918 cm⁻¹ correspond to the stretching vibrations of (BO₃) groups, the band at 711 cm⁻¹ to the vibrations of (BeO₄) tetrahedra, and the band at 672 cm⁻¹ to the bending of (AlO₆).

The infrared spectrum also gives no evidence for the presence of either H_2O or OH in the structure of mengxianminite.

Chemical composition

Quantitative elemental microanalyses of mengxianminite were conducted with a JEOL JXA-8100M electron microprobe (WDS mode, 15 kV, 20 nA, beam diameter 1 μ m) at the Second Institute of Oceanography, State Oceanic Administration (Hangzhou, China). Standards for the analysis were topaz (AlK α and SiK α), cassiterite (SnL α), forsterite (MgL α), fayalite (FeK α), synthetic MnTiO₃ (MnK α and TiK α), synthetic ZnO (ZnK α), albite (NaK α), K-feldspar (KK α), and synthetic CaSiO₃ (CaK α). The low analytical total is due to the presence of large amounts of BeO and B₂O₃ contents.

The BeO and B_2O_3 contents were measured using secondary ion mass spectrometry with a CAMECA NanoSIMS 50L at the Institute of Geology and Geophysics, China Academy of Sciences. A relatively high beam current (500 pA) was used initially to pre-sputter 12 × 12 µm of the matrix area. The measurements were undertaken in raster imaging mode by scanning a focused Cs⁺ primary ion beam (30 pA, 0.5 µm diameter) over 10 × 10 µm matrix area within the pre-sputtered regions. Negative secondary ions ⁹Be⁻, ¹⁰B⁻, ¹¹B⁻, ¹⁶OH⁻, and ¹⁸O⁻ were collected simultaneously, along with secondary electrons (SE). To be used as a standard for Be and B, the structure and composition of hambergite from the Xianghualing skarn was confirmed as the F-free end-member Be₂(BO₃)(OH) by single-crystal XRD in the Laboratory Mineralogy, Liège University (Belgium). The calibration factors for Be and B in the standard were obtained

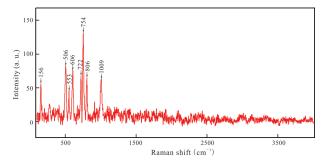


FIGURE 2. The Raman spectrum of mengxianminite.

through the calculation of the experimental Be and B ions yield, having chosen O as the inference element for the matrix. Thus we derived the ratios (Be/O) and (B/O), defined as $(Be^{-}/O^{-})/[(Be(at.)/O(at.)]]$ and $(B^{-}/O^{-})/[(B(at.)/O(at.)]]$, respectively, where Be⁻, B⁻, and O⁻ are the current intensities detected at the electron multiplier, and (at.) is the elemental atomic concentration. The ratios (Be/O) and (B/O) were then used to calculate the Be and B concentration in mengxianminite. Additionally, lithium was confirmed to be absent from the mineral.

The chemical composition of mengxianminite is given in Appendix¹ Table 1. The empirical formula, calculated on the basis of 26 O atoms per formula unit, is: $(Ca_{1.64}, Na_{0.39})_{\Sigma 2.03}(Sn_{1.85}, Zn_{0.24})_{\Sigma 2.09}$ $(Mg_{1.75}, Fe_{0.72}, Al_{0.42}, Mn_{0.19}, Ti_{0.01})_{\Sigma 3.09}Al_8[(B_{1.01}O_3)(Be_{0.94}O_4)O_6]_2$, yielding the simplified formula of $Ca_2Sn_2Mg_3Al_8[(BO_3)(BeO_4)O_6]_2$. The ideal formula requires Al_2O_3 38.41 wt%, SnO₂ 28.38 wt%, MgO 11.39 wt%, CaO 10.56 wt%, B₂O₃ 6.56 wt%, and BeO 4.71 wt%, total 100.00 wt%.

X-ray powder diffraction

The powder XRD pattern of mengxianminite was collected from micro-diffraction data of two crystals, on a RIGAKU D/max Rapid IIR micro-diffractometer (CuK α , $\lambda = 1.54056$ Å) at the School of Earth Sciences and Info-physics, Central South University, China. The micro-diffractometer was operated with a Gandolfi-like motion, under 48 kV and 25 mA, using a 0.05 mm diameter collimator; total exposure time was 2 h. The structural model of single crystals (see below) was used to index the powder XRD pattern of mengxianminite (Appendix¹ Table 2). Calculated intensities were obtained from the structural data with POWDER CELL (Krauz and Nolze 1996). The stronger eight lines of the powder XRD pattern [d in Å (I)(hkl)] are: 3.000 (35)(16.2.0); 2.931 (100)(17.1.1); 2.475 (29)(022); 2.430 (30)(13.3.1); 2.375 (100)(14.0.2/640); 2.028 (52)(21.3.1); 1.807 (35)(913); 1.530 (98)(14.6.0/15.3.3). Because of small size of the crystals and large beam diameter (100 m), some peaks of mengxianminite overlap with phlogopite and fluorite. Unit-cell parameters calculated from powder XRD data of mengxian minite are: a =60.83(14), b = 9.835(4), c = 5.742(1) Å, V = 3435(10) Å³, Z = 8, and space group is Fdd2.

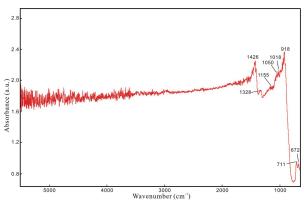


FIGURE 3. The infrared spectrum of mengxianminite.

¹Deposit item AM-17-105919, Appendix Tables 1–6 and CIF. Deposit items are free to all readers and found on the MSA website, via the specific issue's Table of Contents (go to http://www.minsocam.org/MSA/AmMin/TOC/2017/Oct2017_data/Oct2017_data.html).

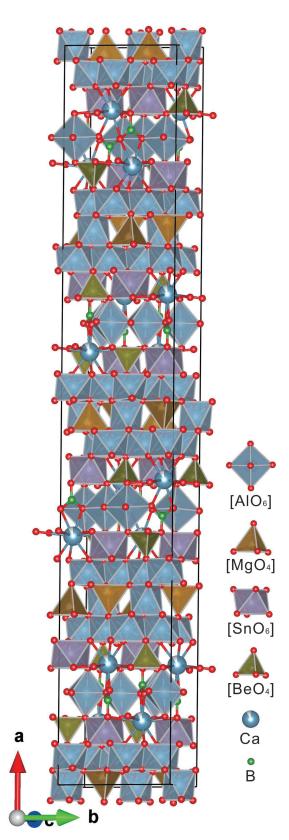


FIGURE 4. The crystal structure of mengxianminite, drawn using the VESTA 3 program (Momma and Izumi 2011).

CRYSTAL STRUCTURE DETERMINATION

Single-crystal X-ray diffraction data, aimed to perform a structure refinement of mengxianminite (CIF¹ available on deposit), were collected on an Agilent Technologies Xcalibur four-circle diffractometer, equipped with an EOS CCD area-detector (University of Liège, Belgium), on a crystal fragment measuring 0.139 $\times 0.108 \times 0.088$ mm. Some 420 frames with a spatial resolution of 1° were collected by the φ/ω scan technique, with a counting time of 90 s per frame, in the range $5.34^{\circ} < 2\theta < 53.31^{\circ}$. A total of 7315 reflections were extracted from these frames, corresponding to 2063 unique reflections. Unit-cell parameters refined from these reflections are a = 60.699(4), b = 9.914(1), c = 5.745(1)Å, $\alpha = \beta = \gamma = 90^{\circ}$, V = 3457.4(4) Å³, Z = 8, space group *Fdd*2. Crystal data, data collection information, and refinement details for mengxianminite are provided in Appendix¹ Table 3. Data were corrected for Lorentz, polarization and absorption effects, the last with an empirical method using the SCALE3 ABSPACK scaling algorithm included in the CrysAlisRED package (Oxford Diffraction 2007). Scattering curves for neutral atoms, together with anomalous dispersion corrections, were taken from International Tables for X-ray Crystallography (Wilson 1992). In the final refinement cycle, all atoms were refined anisotropically, leading to the R_1 ($F_0 > 2\sigma F_0$) value 0.0432. Final atomic coordinates, equivalent isotropic displacement parameters and bond valence sums, as well as anisotropic displacement parameters, are given in Appendix¹ Tables 4 and 5, respectively. Selected bond distances and angles are listed in Appendix1 Table 6.

The structure of mengxianminite is based on a closed-packed oxygen framework, with two different modules stacked along the *a* axis (Figs. 4 and 5). The first module (module A) consists of two O layers and one T1 layer. The O layer was made up of the edge-sharing octahedra, occupied by the cations of Al1, Al2, Al3, and Al4 (<Al-O> = 1.907–1.925 Å), each O layer contains six AlO₆ octahedra per layer in the unit cell. The T1 layer with Al3 in octahedra and Mg/Fe in tetrahedra occurs between two O layers;

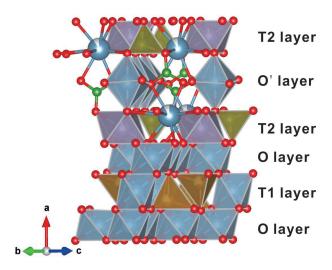


FIGURE 5. An enlarged view of a portion of the mengxianminite structure, showing the connectivity in the A module (O-T1-O) and B module (T2-O'-T2). Drawn using the VESTA 3 program (Momma and Izumi 2011). For key, see Figure 4.

it consists of two AlO₆ octahedra (<Al-O> = 1.907 Å) and four $(Mg,Fe)O_4$ tetrahedra (<Mg-O> = 1.943 Å) per layer in the unit cell, site population refinement shows that the tetrahedral site is occupied by 0.56 Mg, 0.36 Fe, and 0.08 Zn (Appendix¹ Table 4). The second module (module B) is more complex, consists of two T2 layers and one O' layer. The T2 layer is composed of two SnO₆ octahedra ($\langle Sn-O \rangle = 2.077$ Å), two BeO₄ tetrahedra ($\langle BeO \rangle =$ 1.668 Å), and two CaO₁₁ polyhedra (<Ca-O> = 2.71 Å), the Sn octahedral site was occupied by 0.92 Sn + 0.08 Mn, and 0.81 Ca + 0.19 Na in the CaO₁₁ polyhedral site (Appendix¹ Table 4); the O' layer was made up of four AlO₆ octahedra (<Al-O> = 1.975 Å) and four BO₃ groups ($\langle B-O \rangle = 1.364$ Å), Al5 occurs in the octahedral site, which was occupied by 0.69 Al + 0.31 Mg. As shown in Figure 5, the SnO₆ octahedra are isolated in the T2 layers; their connectivity is realized via BeO4 and CaO11 groups. In the O' layer, AlO_6 edge-sharing octahedra form chains running along the {011} or $\{0\overline{1}1\}$ direction; these chains are connected in the c direction by the BO₃ triangular groups (Fig. 5). The cation-layer sequence in mengxianminite can be described as $4 \times (O-T1-O-T2-O'-T2)$; the framework of mengxianminite is accordingly composed of four A modules and four B modules.

ORIGIN

Mengxianminite, ideally Ca₂Sn₂Mg₃Al₈[(BO₃)(BeO₄)O₆]₂, is a new Sn-bearing borate with additional O anions and without water (Strunz 06.AB.90, Dana 24.2.8.1), showing a novel structural type belonging to the Fdd2 space group. To consider the origin of mengxianminite, we must first define the context offered by the Xianghualing skarn. This skarn is characterized by the presences of large amounts of Sn minerals (cassiterite, hulsite, ferronigerite-2N1S), Be minerals (hsianghualite, liberite, chrysoberyl, hambergite, bertrandite, ferrotaaffeite-2N'2S), and B minerals (fluoborite, hambergite, dravite, hulsite), reflecting multiple Sn and Be mineralizations. Zhang and Wang (1986) indicated that the assemblages of chondrodite-phlogopitefluoborite-hulsite were formed by the replacement of forsterite, spinel, and and iron-poor augite ("fassaite"), together with cassiterite-magnetite mineralization during introduction of F- and B-rich solutions in the Xianghualing deposit, and that the Be mineralization of the Xianghualing skarn was attributed to the metasomatism and reformation processes of magnesium skarn by F- and B-rich fluids in the exocontact skarn zone.

Mengxianminite occurs as subhedral to euhedral crystals in the hsianghualite vein of the Xianghualing skarn (Fig. 1). The intimate intergrowths of mengxianminite with hsianghualite, phlogopite, and fluorite indicate that it crystallized under F-rich conditions during the late stages of mineralization in the Xianghialing skarn. To our knowledge, mengxianminite (or similar synthetic compounds) has not been synthesized. On the basis of fluid inclusions studies, Liu and Zeng (1998) suggested that hsianghualite formed from 270 to 290 °C and at 30-60 MPa in the Xianghualing skarn. Since mengxianminite occurs intergrown with hianghualite, these values must also represent the physical condition of mengxianminite formation. The presence of Sn and Be in the hydrothermal fluids responsible for the formation of mengxianminite is probably derived from the genetically related Laiziling granite, while B came from the wall-rocks (Tiaomajian Formation) with 284 ppm B (Zhong 2014).

IMPLICATIONS

Chemically, mengxianminite is close to minerals of the nigerite group, but has lower concentrations of Al₂O₃. Mengxianminite is the first borate mineral that contains both Sn and Be. The crystal structure of mengxianminite, shown in the Figure 4, is a new modular structure type with two different modules stacked along the *a* axis. The A module of mengxianminite consists of two O layers and one T1 layer, where one O layer and one T1 layer can be stoichiometrically identical to the spinel module of nigerite group minerals (Arakcheeva et al. 1995; Grey and Gatehouse 1979), as well as taaffeite group minerals (Armbruster 2002). The A module of mengxianminite might be thus considered as the spinel module with an additional O layer. The intergrowths of mengxianminite with magnesiotaaffeite-2N'2S, found in the Xianghualing skarn, suggest these minerals sharing the same structural unit may crystallize under similar Be-, Al-, and Mg- rich condition. The B module of mengxianminite is a new module, composed of two T2 layers and one O' layer (Fig. 5), showing the simplified formula CaSnAl(BeO₄)(BO₃). The Ca atoms are located in a 11-coordinated polyhedron occurring between T2 layer and O' layer; the bond valence sum for Ca is 1.62 (Appendix¹ Table 4), slightly lower than 2, indicating some light element such as Na in the 11-coordinated polyhedron site. The similar bond distance of Mg and Al in the octahedra, suggests the disordered distributions of Mg and Al in the octahedra of O' layer. However, the CaO₁₁ polyhedron and BO3 groups were never reported in the modules of nigerite group and taaffeite group minerals. The B module of CaSnAl(BeO₄)(BO₃) in crystal structure of mengxianminite is thus a new module found in the nature.

It should be noted that the modular structure of mengxianminite can be compared to the polysomatic structures of högbomite-24R(Hejny and Armbruster 2002) and the *c* unit-cell parameter of mengxianminite is 5.740 Å, which is very similar to the *a* unitcell parameter of phases belonging to the nigerite, högbomite, and taaffeite groups (Armbruster 2002; Hejny and Armbruster 2002). Like nigerite group minerals, mengxianminite may also be one of the polysomatic minerals. The chemical feature (Appendix¹ Table 1) and structural data show that Mg can be replaced by Fe at the Mg sites, and minor Ti can enter the Sn sites. Therefore, it may be imagined that the discovery of mengxianminite draws attention to a certain expectation that the Fe and Ti analog of mengxianminite will be found in the future. In addition, similar polysomatic minerals, made up of different ratios of mengxianminite modules, could also exist.

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