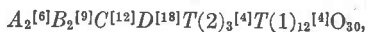


CRYSTAL CHEMISTRY OF MILARITE-TYPE MINERALS

WARREN C. FORBES, WERNER H. BAUR, AND AIJAZ A. KHAN,
*Department of Geological Sciences University of Illinois at Chicago
Chicago, Illinois 60680*

ABSTRACT

Milarite-type minerals have the general formula



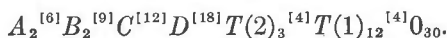
where the *A*-position can be occupied by Mg, Ca, Fe, Ti, Zr; the *B*-position by K, Na, Mg, H₂O (or H₃O); the *C*-position by K, Na, Ca, Ba; the *D*-position by H₂O; the *T*(2)-position by Al, Be, Mg, Fe, Li; and the *T*(1)-position by Si and Al; the coordination number is given in bracketed superscripts. The *B*-position which previously had not been recognized as being occupied in milarite-type minerals is 9-coordinated with three short and six longer bonds to the surrounding oxygen atoms. This position must be occupied in all milarite-type compounds in which the number of non-tetrahedral cations and/or H₂O, relative to the 15 tetrahedral cations, is greater than three. If this number is greater than five (as in armenite) the 18-coordinated sites (in 0, 0, 0) in the center of the Si₁₂O₃₀ double hexagonal ring has to be occupied. In yagiite, merrihueite, roedderite, and "Ca-cordierite" part of the Mg (or Fe²⁺) must be in tetrahedral coordination in the *T*(2)-sites. The different groups of milarite-type minerals can be formally derived from a proto-milarite, 2MgO·2Al₂O₃·11SiO₂ or Mg₂Al₃Si₁₁AlO₃₀, by simple cationic substitutions which alter the Al/Si ratio and which progressively fill the *B*- and *C*-positions.

INTRODUCTION

Milarite was found first in the Val Giuf, Graubünden, Switzerland and was described by Kennigott (1870). Since then several other minerals isostructural with milarite have been characterized (see Table 2). Milarite group minerals crystallize in space group *P6/m 2/c 2/c* with approximate cell parameters of $a \approx 10 \text{ \AA}$ and $c \approx 14 \text{ \AA}$. The crystal-chemical formula of the milarite-type minerals as deduced by Ito *et al.* (1952) and adopted by many subsequent workers is $A_2^{[6]}C^{[12]}T(2)_3^{[4]}T(1)_{12}^{[4]}O_{30} \cdot H_2O$, where the coordination numbers of the cations are indicated as superscripts in square brackets. The most prominent feature of the structure is a hexagonal double ring of silicate tetrahedra, of composition Si₁₂O₃₀. These rings are joined by tetrahedrally coordinated cations in sites *T*(2) into a three-dimensional tetrahedral framework and further connected by octahedrally coordinated cations in the *A*-sites. The double rings are stacked in the *c*-direction thus forming the 12-coordinated *C*-site between them. The water molecules found in some milarite-type minerals have been as-

sumed to be located inside the double hexagonal rings in large voids, with 18-coordination.

The synthetic compound $K_2Mg_5Si_{12}O_{30}$ (Roedder, 1951) contains twice as much potassium as can be accommodated in the *C*-sites. A structure determination of $K_2Mg_3Si_{12}O_{30}$ (Khan *et al.*, 1971) has shown that this excess potassium is located in 9-coordinated sites (with three short K-O bonds, and six longer ones) which previously had not been known to be occupied in milarite-type structures. The general formula for milarite group minerals is therefore:



Thus, the six different sites (see Table 1) may contain up to 21 cations and/or water molecules per 30 oxygen atoms in the formula unit. Since the radii of the cations which can be accommodated in these different sites range from 0.4 Å (for Si^{4+}) up to 1.7 Å (for K^+ and Ba^{2+}), and since the Mg and Fe atoms can enter into both tetrahedral and octahedral coordinations (Khan *et al.*, 1971) the chemical variability of milarite-type minerals is large.

CRYSTAL CHEMISTRY

In Table 2 we show all known species of the milarite group with the cations and water molecules distributed among the six positions described above. Included in this list are two "calcium-rich cordierites", mentioned by Miyashiro (1956) as being probably of the milarite-type. The assignment of the atomic species to the different

Table 1. Cation and/or water positions in milarite-type structures (C.N. = coordination number). The list of elements occupying the different positions includes those listed in Table 2, as well as trace elements identified by Černý (1968) in amounts larger than 0.01wt% in milarites from Věžná.

Symbol	Equipoint	C.N.	Site Symmetry	Possible Occupants
T(1)	24(m)	4	1 - C_1	Si, Al, Mg
T(2)	6(f)	4	222 - D_2	Al, Be, Mg, Fe, Li, Ti, B
A	4(c)	6	32 - D_3	Mg, Ca, Fe, Ti, Zr, Mn, Sr, Co, Nb
B	4(d)	9(=3+6)	$\bar{6}$ - S_3	K, Na, H_2O , (H_3O^+), Mg, Fe
C	2(a)	12	622 - D_6	K, Na, Ca, Ba, Y, Yb, Pb
D	2(b)	18(=12+6)	6/m C_6^h	(H_2O ?)

Table 2. Distribution of chemical elements over the different sites in milarite-type minerals. All atom fractions are multiplied by a factor of 100. The sum of atoms exceeds in several cases the possible maximum occupancy of a given site. Such deviations are believed to be within experimental error of the chemical analyses.

Mineral	Locality	A ₁₆	B ₉₁	C ₁₂₁	D ₁₁₈	T(2) ₃₀₀	T(1) ₁₂₀₀	Reference for Analysis
roodderite	Indarch	Mg ₁₈₁ Fe ₂₇ ²⁺	Na ₁₀₀	K ₆₉ Na ₃₀		Mg ₃₀₀	Si ₁₁₈₈ A ₇ Mg ₅	Fuchs et al., 1966
roodderite	Michita County	Mg ₁₈₀ Fe ₁₀ ²⁺	Na ₁₅₀	K ₈₀ Na ₂₀		Mg ₃₀₀	Si ₁₁₅₀ A ₅ O	Olsen, 1967
Na ₂ O·5MgO·12SiO ₂	synthetic	Mg ₂₀₀	Na ₁₀₀	Na ₁₀₀		Mg ₃₀₀	Si ₁₂₀₀	Seifert & Schreyer, 1969
merrihuicite	MezS-Madras	Fe ₂₀₀	Na ₇₆	K ₉₄ Ca ₁₂		Fe ₁₈₅ Mg ₁₂₇	Si ₁₂₀₀	Bodd et al., 1965
K ₂ O·5MgO·12SiO ₂	synthetic	Mg ₂₀₀	K ₁₀₀	K ₁₀₀		Mg ₃₀₀	Si ₁₂₀₀	Khan et al., 1971
sedgemite	Central Asia	Zr ₈₅ Fe ₄₂ ³⁺ Ti ₃ Fe ₂ ²⁺ ¹⁸	Na ₉₀ K ₁₀	K ₁₀₀		Li ₂₆₁ Al ₆ Fe ₃ ³⁺ Li ₂₆₁ Al ₆ Fe ₃ ³⁺	Si ₁₂₀₀	Dusmatov et al., 1968
milarite	Val Giuf	Ca ₂₀₉	(H ₂ O) ₅₇ Na ₁₅	K ₁₀₅		Be ₂₁₀ A ₆ ⁹²	Si ₁₁₉₈	Prinche, 1951
milarite	Piz Ault	Ca ₂₀₆	(H ₂ O) ₆₈ Na ₃₈ K ₁₅	K ₁₀₀		Be ₁₇₂ A ₈ ⁸⁶ Fe ₁₉	Si ₁₁₉₄	Hügi, 1956
milarite	Xoia Peninsula	Ca ₂₀₉	(H ₂ O) ₇₀ Na ₁₀	K ₁₀₃		Be ₁₄₅ A ₆ ¹⁵³	Si ₁₂₀₂	Sosedko & Telesheva, 1962
milarite	Kazakhstan	Ca ₁₉₂ Mn ₈ Fe ₂	(H ₂ O) ₇₇	K ₉₅ Na ₅		Be ₂₂₆ A ₆ ⁷³	Si ₁₁₆₆	Christyakova et al., 1964
milarite	Central Asia	Ca ₁₉₅ Sr ₂	(H ₂ O) ₇₆ Na ₂₈	K ₉₈		Be ₁₈₆ A ₆ ¹¹⁴	Si ₁₀₅₉ A ₂ Al ₁₄₁	Iovcheva et al., 1966
osumilite	Sakkabira	Mg ₉₃ Fe ₉₆ Mn ₁₁		K ₆₅ Na ₂₃ Ca ₂		Al ₂₆₃ Fe ₂ ²⁺	Si ₁₀₃₈ A ₁₆₂	Olsen & Bunch, 1970
osumilite	Monte Atci	Fe ₂ ²⁺ Mg ₇₂ Mn ₁₀		K ₆₆ Na ₁₂ Ca ₃		Al ₂₇₂ Fe ₂ ²⁺	Si ₁₀₃₄ A ₁₆₆	Olsen & Bunch, 1970
osumilite	Oregon	Fe ₂ ²⁺ Mg ₈₈ Mn ₁₄		K ₆₅ Na ₈ Ca ₂		Al ₂₇₅ Fe ₃ ³⁺	Si ₁₀₄₁ A ₁₅₉	Olsen & Bunch, 1970
K ₂ O·MgO·5Al ₂ O ₃ ·20SiO ₂	synthetic	Mg ₂₀₀		K ₁₀₀		Al ₃₀₀	Si ₁₀₀₀ A ₂₀₀	Schreyer & Seifert, 1967
yuqite	Colomera	Mg ₂₀₀	Na ₅₀	Na ₇₀ K ₃₀		Al ₁₉₆ Mg ₆₀ Fe ₂₄ Ti ₁₀	Si ₁₀₂₀ A ₁₈₀	Bunch & Ruchs, 1969
"Ca-cordierite"	Laacher See	Mg ₁₂₆ Fe ₆₄ Mn ₁₀ ²⁺	(H ₂ O) ₉₂ Fe ₇₉	Ca ₉₅		Al ₁₃₂ Mg ₁₁₀ Fe ₅₂	Si ₁₈₆₅ A ₂ 335	Miyashiro, 1956
"Ca-cordierite"	Celebes	Fe ₁₆₇ Fe ₃₈ ²⁺	Mg ₁₅ (H ₂ O) ₃	Ca ₈₀		Al ₃₀₆	Si ₁₈₅₅ A ₂ 345	Miyashiro, 1956
armenite	Kongsberg	Ca ₂₀₃	(H ₂ O) ₁₉₁ Na ₀ K ₃	Ba ₉₅	(H ₂ O) ₂₅	Al ₃₀₀	Si ₁₈₈₀ A ₂ 316	Neumann, 1941
4MgO·4Al ₂ O ₃ ·7SiO ₂ ⁷	synthetic	Mg ₂₀₀				Al ₃₀₀	Si ₁₇₀₀ A ₂ 500	Schreyer & Schairer, 1962

sites is based on the published chemical analyses and on the sizes of the ions in question. The water molecules are assumed by us to be located in the 9-coordinated *B*-sites. Our reasons for this assignment are:

1. The H₂O molecule is of approximately the same size as the potassium ion which in K₂Mg₅Si₁₂O₃₀ prefers the *B*-sites over the 18-coordinated *D*-sites (which would be much too large for either K or H₂O).
2. Upon heating to 1000°C, and after losing H₂O, the *c* cell constant of milarite increases by approximately 0.06Å, while the *a* cell constant decreases by about 0.04Å (Černý, private communication, 1971; Iovcheva *et al.*, 1966). If the water molecules were located in the large *D*-sites as suggested by Iovcheva *et al.*, their loss should not affect the cell constants in any measurable degree. If they are, however, in the *B*-positions the changes in cell parameters could be interpreted as the result of an adjustment of the oxygen atoms around the *B*-site taking place after the removal of the H₂O (compare the discussion of the cell parameters of K₂Mg₅Si₁₂O₃₀ and osumilite in Khan *et al.*, 1971).

This assignment is at variance with the findings of Ito *et al.* (1952). However, their structure determination is of low accuracy ($R = 0.30$ for 105 observed structure factors) and cannot be taken as solid proof. Bakakin and Soloveva (1966) report in an abstract that they found in milarite "water molecules on threefold axes near the face of the Ca flattening octahedron". Their statement may possibly refer to what we call the *B*-sites, but since they do not give any further details, its meaning is uncertain. The presence of water in milarite-type minerals has been the subject of much discussion. Water is well authenticated in milarite and armenite only. Miyashiro (1956) assumed water to be present in osumilite. However, Olsen and Bunch (1970), who carefully analyzed osumilite from its three known occurrences, including the type locality, concluded that water is not a constituent of osumilite. Likewise there is no clear indication for the presence of water in merrihueite, roedderite, yagiite, or in the synthetic forms (K₂Mg₅Si₁₂O₃₀ and synthetic osumilite; Schreyer and Seifert, 1967).

The milarite group minerals may be divided into four subgroups on the basis of their Al contents. The roedderite subgroup is virtually Al-free, milarite contains between one and three Al atoms per formula unit, osumilite has five, and armenite as well as the "Ca-cordierites" of Miyashiro (1956) have six Al atoms. In all four groups, the octahedrally coordinated *A*-position is occupied by a divalent cation. An

exception is sodgianite, where this site appears to be occupied by 3- and 4-valent cations (sodgianite has been recognized by Strunz (1970) to belong to the milarite-type minerals). The *B*-sites are usually populated by alkalis and water and occupancy of this position is very variable between the groups. It is most fully occupied in armenite. The roedderite group contains up to 1.5 atoms in this position. In the osumilites the *B*-site appears to be empty. Brown and Gibbs (1969), in their crystal structure determination, do not report to have found any atoms to be located there. The composition of the Ca-cordierites requires that divalent cations, either Mg or Fe, are in this position, thus making the coordination of these cations rather unusual. However, one should keep in mind that an eight-coordination for Mg is commonly found in garnets.

Because water molecules and alkali ions seem to replace each other in the *B*-sites of milarite we actually may not be dealing with water molecules but instead with hydronium ions (H_3O^+). If this is the case we would expect the oxygen atom of the hydronium ion not to be exactly in the *B*-position ($1/3 \ 2/3 \ 0$), but slightly displaced from it along the threefold axis ($1/3 \ 2/3 \ z$), so that H_3O could form three hydrogen bonds in a trigonally pyramidal configuration to the three neighboring O(1) oxygen atoms (Khan *et al.*, 1971).

The *C*-position is occupied by alkali ions in all minerals but armenite, where it is filled by barium. The *T*(2)-sites are marked by considerable variation. They may be occupied by Mg, Fe^{2+} , Fe^{3+} , Be, Al, or Li. Magnesium in tetrahedral coordination is very rare in silicates. In the system K_2O -MgO-SiO₂ however, there are two phases in which Mg is 4-coordinated: Mg-merrihucite, $K_2Mg_5Si_{12}O_{30}$ (Khan *et al.*, 1971), where the four-coordination has been substantiated by the structure determination (with a bond length Mg-O of 1.955Å), and in $K_2O \cdot MgO \cdot 3SiO_2$, which according to Roedder (1951) has an X-ray powder pattern strongly resembling the pattern of kalsilite ($KAlSiO_4$). Consequently, $K_2O \cdot MgO \cdot 3SiO_2$ may be formulated as $K(Mg_{0.5}Si_{0.5})SiO_4$, with Mg and Si substituting for Al.

Schreyer and Schairer (1962) have shown that alkali-free milarite-type phases can be synthesized within the system MgO - Al_2O_3 - SiO_2 . As these authors recognized, the milarite structure theoretically could form from the composition $2MgO \cdot 2Al_2O_3 \cdot 11SiO_2$, *i.e.*, only the *A*, *T*(2), and *T*(1) positions would be filled. If we consider 2:2:11 to represent a "proto"-milarite composition, the main groups of milarite minerals may be derived by simple cationic substitutions. This is shown schematically in Table 3. As can be seen, numerous variations are possible, simply by altering the Al/Si ratio and consequently the

Table 3. Formal derivation of milarite-type minerals from the proto-milarite-type.

Replacement	A ₂ ^[6]	B ₂ ^[9]	C ^[12]	D ^[18]	T(2)	T(1)	O ₃₀	Type
(starting composition)	M ₂ ²⁺				M ₃ ³⁺	Si ₁₁ Al	O ₃₀	proto-milarite
(Si+2M ¹⁺ +3M ²⁺) for 4M ³⁺	M ₂ ²⁺	M ¹⁺	M ¹⁺		M ₃ ²⁺	Si ₁₂	O ₃₀	roedderite
(Si+5M ¹⁺ +M ⁴⁺ +M ³⁺) for (2M ²⁺ +3M ³⁺ +Al ³⁺)	M ⁴⁺ +M ³⁺	M ¹⁺	M ¹⁺		M ₃ ¹⁺	Si ₁₂	O ₃₀	sodgianite
(Si+M ¹⁺ +2M ²⁺) for 3M ³⁺		M ₂ ²⁺	(H ₂ O)	M ¹⁺		M ₂ ²⁺ M ³⁺	Si ₁₂	O ₃₀
(Al+M ¹⁺) for Si	M ₂ ²⁺		M ¹⁺		M ₃ ³⁺	Si ₁₀ Al ₂	O ₃₀	osumilite
(2Al+M ²⁺) for 2Si	M ₂ ²⁺	(H ₂ O)	M ²⁺	(H ₂ O) _x	M ₃ ³⁺	Si ₉ Al ₃	O ₃₀	armenite

alkali content. It should be mentioned, however, that this starting composition does not produce a milarite-type phase under the experimental conditions investigated by Schreyer and Schairer (1962).

Olsen and Bunch (1970) studied the compositions of natural osumilites and found that the $K/(K + Na + Ca)$ ratio varies with the occupancy of the C-position. Actually their diagram can be extended to include the contents of the B-position as well, thus allowing the addition of yagiite (Fig. 1). In Figure 2, we have plotted total Al + Fe³⁺ against $\Sigma(K + Na + Ca)$. It appears that as occupancy of the B-

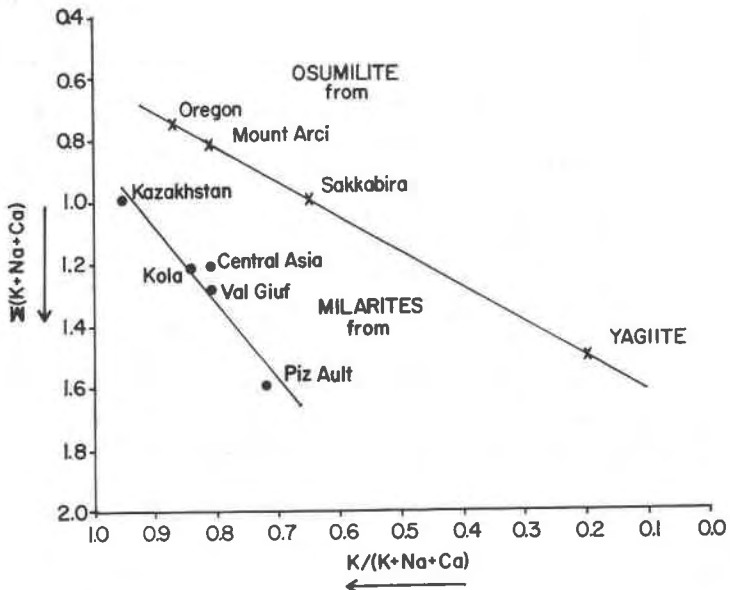


Fig. 1. Ratio $K/(K + Na + Ca)$ versus total $K + Na + Ca$ located in the B- and C-positions in osumilite and yagiite (crosses) and milarites (circles).

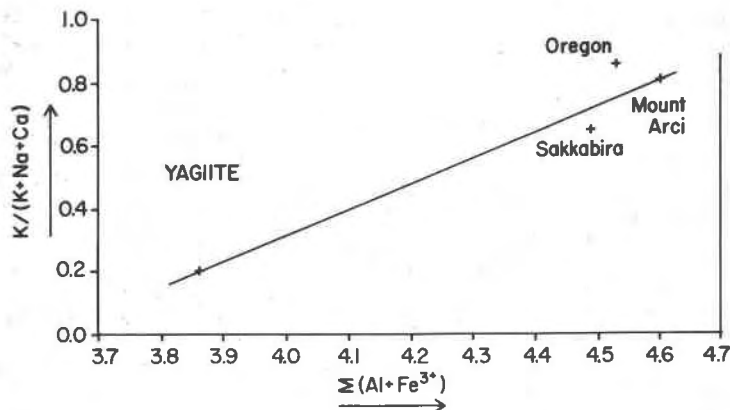


FIG. 2. Total Al + Fe³⁺ versus ratio K/(K + Na + Ca) in osumilite and yagiite.

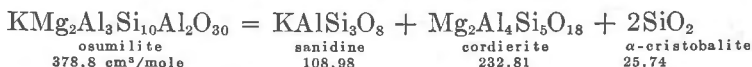
and C-sites increases, the Al + Fe³⁺ content decreases and the sodium content increases (see Fig. 1). Thus, natural osumilites and yagiite are members of a solid solution series, related by the substitution $\text{K} + \text{Al} \rightleftharpoons 2\text{Na} + \text{Mg}$. Idealized end members of this series are $\text{KMg}_2\text{Al}_3(\text{Si}_{10}\text{Al}_2)\text{O}_{30}$ and $\text{Na}_2\text{Mg}_2(\text{Al}_2\text{Mg})(\text{Si}_{10}\text{Al}_2)\text{O}_{30}$. The sodic end member may be related to roedderite through the substitution $4\text{Al} = 2\text{Si} + 2\text{Mg}$. However, intermediate members of the series are lacking. A similar relationship between the K/K + Na + Ca ratio and total cation occupancy of the B + C sites is found for milarites as well (Fig. 1). The Ca represents the excess over 2 in the A site. However, there are no corresponding changes in the abundances of other cations as was found for osumilites. Possibly this means that variation in K/K + Na + Ca ratio is correlated with the postulated hydronium content of the B-site. Unfortunately, there is no direct evidence bearing on this possibility.

OCCURRENCE

Milarite-type minerals are typically found in low pressure environments. Milarite itself as well as armenite occurs as a fissure-filling or pegmatitic mineral. Osumilite is found only in volcanic rocks, where it occurs within vesicles as well as in the groundmass. Milarite-type phases found in synthetic systems usually occur at low pressures. Roedderite, $\text{Na}_2\text{Mg}_5\text{Si}_{12}\text{O}_{30}$, was found as a decomposition product of richterite only at pressures below 50 bars $P(\text{H}_2\text{O})$ (Forbes, 1971). Iron-bearing roedderite, again the decomposition product of a sodic amphibole, occurs only at pressures below 500 bars $P(\text{H}_2\text{O})$ (Ernst,

1960). Seifert and Schreyer (1969) have synthesized Mg-merriheuite at pressures up to 32.5 Kbar; however, from consideration of the phase equilibria of this phase, they conclude that this compound is restricted to low pressure environments.

While field relations and experimental data indicate that milarite-minerals form under low-pressure conditions, it is surprising to note that the molar volume change for the breakdown of osumilite under volcanic conditions (Schreyer and Seifert, 1967)



is +14.5 cm³/mole, which suggests that osumilite in this instance would be stable at higher pressures. If orthoclase and α-quartz are used, instead of sanidine and cristobalite, ΔV is reduced to 8.4 cm³/mole but it is still positive. It is possible that changes in the phase compositions and/or nature of the phases might alter the sign of the volume change. The molar volume data for osumilite were obtained from Schreyer and Seifert (1967), for the other phases the compilation of Robie *et al.* (1966) was used.

As Miyashiro (1956) has recognized, milarite-type minerals, particularly osumilite, may be quite common in nature but are mistaken for cordierite. Several instances of optically anomalous cordierites were discussed by him. Two additional reports of optically positive "cordierite" (Rutherford, 1933; Conant, 1935) support the view that osumilite may be a fairly common mineral. It might be profitable to study such occurrences further.

ACKNOWLEDGEMENT

We thank Dr. P. Černý for communicating to us in advance of publication the results of his heating experiments on milarites and Dr. Olsen for a review of the manuscript.

REFERENCES

- BAKAKIN, V. V., AND L. P. SOLOVEVA (1966) The crystallochemical analysis of compounds with beryl and milarite structures. *Acta Crystallogr.* 21, A41.
- BROWN, G. E., AND G. V. GIBBS (1969) Refinement of the crystal structure of osumilite. *Amer. Mineral.* 54, 101-116.
- BUNCH, T. E., AND L. FUCHS (1969) Yagiite, a new sodium-magnesium analogue of osumilite. *Amer. Mineral.* 54, 14-18.
- ČERNÝ, P. (1968) Berylliumminerale in Pegmatiten von Vezná und ihre Umwandlungen. *Ber. Deutsch. Ges. Geol. Wiss.* B.13, 565-578.
- CHISTYAKOVA, M. B., G. A. SOLODKINA, AND Z. P. RAZMANOVA (1964) Milarite from central Kazakhstan. *Dokl. Akad. Nauk SSSR*, 159, 1305-1308. [transl. *Dokl. Acad. Sci. USSR, Earth Sci. Sect.* 170, 102-105].

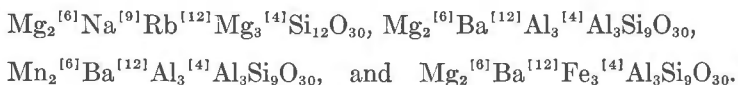
- CONANT, L. C. (1935) Optically positive cordierite from New Hampshire. *Amer. Mineral.* 20, 310-311
- DODD, R. T., W. R. VAN SCHMUS, AND U. B. MARVIN (1965) Merrihueite, a new alkali-ferromagnesian silicate from the Mezö-Madras chondrite. *Science*, 149, 972-974.
- DUSMATOV, V. D., A. F. YEFIMOV, Z. T. KATAYEVA, L. A. KHOROSHILOVA, AND K. P. YANULOV (1968) Sodginaite, a new mineral. *Dokl. Akad. Nauk SSSR*, 182, 1176-1177 [Transl. *Dokl. Acad. Sci. USSR Earth Sci. Sect.*, 182, 137-139].
- ERNST, W. G. (1960) Stability relations of magnesioriebeckite. *Geochem. Cosmochim. Acta*, 19, 10-40.
- FORBES, W. C. (1971) Synthesis and stability relations of richterite. *Amer. Mineral.* 56, 997-1004.
- FUCHS, L., C. FRONDEL, AND C. KLEIN (1966) Roedderite, a new mineral from the Indarch meteorite. *Amer. Mineral.* 51, 949-955.
- HÜGI, T. (1956) Verbreitung des Berylliums und der Beryllium-mineralien in den Schweizer Alpen. *Schweiz. Mineral. Petrogr. Mitt.* 36, 497-510.
- IOVCHEVA, E. I., I. I. KUPRIYANOVA, AND G. A. SIDORENKO (1966) Milarite from central Asia. *Dokl. Akad. Nauk SSSR*, 170, 1394-1397 [Transl. *Dokl. Acad. Sci. USSR Earth Sci. Sect.* 170, 160-163].
- ITO, T., N. MORIMOTO, AND R. SADANAGA (1952) The crystal structure of milarite. *Acta Crystallogr.* 5, 209-213.
- KENNGOTT, A. (1870) Mittheilungen an Professor G. Leonhard. *Neues Jahrb. Mineral. Geol.* 80-81.
- KHAN, A. A., W. H. BAUR, AND W. C. FORBES (1972) Synthetic magnesium merrihueite, dipotassium pentamagnesium dodecasilicate: A tetrahedral magnesian-silicate framework crystal structure. *Acta Crystallogr.* B28, 267-272.
- MIYASHIRO, A. (1956) Osumilite, a new silicate mineral and its crystal structure. *Amer. Mineral.* 41, 104-116.
- NEUMANN, H. (1941) Armenite, a water-bearing barium-calcium-alumosilicate. *Norsk Geol. Tidsskr.* 21, 19-24.
- OLSEN, E. (1967) A new occurrence of roedderite and its bearing on osumilite-type minerals. *Amer. Mineral.* 52, 1519-1523.
- , AND T. E. BUNCH (1970) Compositions of natural osumilites. *Amer. Mineral.* 55, 875-879.
- PALACHE, C. (1931) On the presence of beryllium in milarite. *Amer. Mineral.* 16, 469-470.
- ROBIE, R. A., P. M. BETHKE, M. S. TOULMIN, AND J. L. EDWARDS (1966) X-ray crystallographic data, densities, and molar volumes of minerals. *Geol. Soc. Amer. Mem.* 97, 27-73
- ROEDDER, E. (1951) The system K_2O - MgO - SiO_2 . *Amer. J. Sci.* 249, 81-130; 224-248.
- RUTHERFORD, R. L. (1933) Optically positive cordierite from the Northwest Territories, Canada. *Amer. Mineral.* 18, 216.
- SCHREYER, W., AND J. F. SCHAIRER (1962) Metastable osumilite-and petalite-type phases in the system MgO - Al_2O_3 - SiO_2 . *Amer. Mineral.* 47, 90-104.
- , AND F. SEIFERT (1967) Metastability of an osumilite end member in the system K_2O - MgO - Al_2O_3 - SiO_2 - H_2O and its possible bearing on the rarity of natural osumilites. *Contrib. Mineral. Petrology*, 14, 343-358.
- SEIFERT, F., AND W. SCHREYER (1969) Stability relations of $K_2Mg_5Si_2O_{20}$, an end member of the merrihueite-roedderite group of minerals. *Contrib. Mineral. Petrology*, 22, 190-207.

SOSEDKO, T. A., AND R. L. TELESHEVA (1962) Chemical composition of milarite. *Dokl. Akad. Nauk. SSSR*, 146, 437-439 [Transl. *Dokl. Acad. Sci. USSR. Earth Science Section*, 146, 112-114].

STRUNZ, H. (1970) *Mineralogische Tabellen*, 5th ed. Akademische Verlagsges., Leipzig.

Manuscript received, August 23, 1971; accepted for publication, October 28, 1971.

Note added in proof. Professor Schreyer called to our attention a paper (Borchert and Petzenhauser, 1966) in which the synthesis of four additional members of the milarite group is reported. These could be formulated in accord with the general formula proposed by us, as



The last compound could also be $\text{Fe}_2^{[6]} \text{Ba}^{[12]} (\text{Mg}_2 \text{Fe})^{[4]} \text{Al}_3 \text{Si}_9 \text{O}_{30}$. Of particular interest is the Na-Rb compound because the authors report that they were not able to synthesize the corresponding pure Rb compound. This may mean that Rb is too large to enter the nine coordinated *B* sites, which however can be occupied by potassium ions.

BORCHERT, W., AND I. PETZENHAUSER (1966) Osumilithbildung in verschiedenen silikatischen Systemen. *Ber. Deutsch. Keram. Ges.* 43, 572-576.