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Eifelite, $KNa_3Mg_4Si_{12}O_{30}$, a New Mineral of the Osumilite Group with Octahedral Sodium

K. Abraham[1], W. Gebert[1], O. Medenbach[1], W. Schreyer[1], and G. Hentschel[2]

[1] Institut für Mineralogie, Ruhr-Universität, Postfach 102148, D-4630 Bochum 1,
[2] Hessisches Landesamt für Bodenforschung, Leberberg 9, D-6300 Wiesbaden, Federal Republic of Germany

Abstract. Eifelite of variable composition is uniaxial positive with n_0 near 1.543 and n_e near 1.544, a between 10.14 and 10.15 Å, and c about 14.22 Å, space group $P\frac{6}{m}\frac{2}{c}\frac{2}{c}$. There is a complete series of solid solution between the eifelite end member $KNa_3Mg_4Si_{12}O_{30}$ and roedderite, $KNaMg_5Si_{12}O_{30}$, following the $2\,Na \rightleftharpoons Mg$ substitution. Both eifelite and roedderite have milarite-type structures, but Na is always in six-coordinated sites: In roedderite Na occupies solely a newly defined $B'^{[6]}$-position which is slightly displaced along c from the ideal $B^{[9]}$-position lying on the (001/2)-mirror plane in $K_2Mg_5Si_{12}O_{30}$. In eifelite Na is located both in $B'^{[6]}$ and in the $A^{[6]}$-positions, where it partially replaces Mg. Eifelite has the highest cation occupancy of all osumilite group minerals known thus far.

Both eifelite and roedderite occur in vesicles of contact metamorphosed basement xenoliths ejected with the leucite tephrite lava of the Quaternary Bellerberg volcano in the Eifel, West Germany. They are considered to be precipitates from highly alkaline, MgSi-rich, but Al-deficient gas phases that originated through interaction of gaseous igneous differentiates with the xenoliths.

Introduction

The osumilite group as defined by Fleischer (1981) encompasses a series of silicate minerals characterized by the six-membered-double-ring milarite-type structure (Belov and Tarkhova 1951; Ito et al. 1952). In this chemically rather heterogeneous group osumilite itself (for recent compilation see Schreyer et al. 1983) as well as the minerals roedderite, merrihueite, and yagiite have the simplest major element chemistry involving only the cations K^+, Na^+, Mg^{2+}, Fe^{2+}, Fe^{3+}, and Al^{3+}, while the remaining ones carry essential amounts of some of the elements Li, Be, Ca, Mn, Zn, Zr, and Sn. For the former four minerals, which may also be regarded as an osumilite *subgroup*, a general formula can be given as

$$W_{1\text{-}2}X_2Y_3Z_{12}O_{30}$$

where W=K,Na; X=Mg,Fe^{2+}; Y=Mg,Fe^{2+},Al,Fe^{3+}; and Z=Si,Al.

In this subgroup, according to Bunch and Fuchs (1969), the Si/Al ratio of 7.0 is used as limit between the Al-rich members osumilite and yagiite on one side and the Al-poor or Al-free members roedderite and merrihueite on the other. The mutual substitutional relationships within the osumilite subgroup were discussed by Berg and Wheeler (1976). Previously, Forbes et al. (1972) had given a survey on the crystal chemistry of all milarite-type minerals known up to that time. Based on a structure determination of the synthetic phase $K_2Mg_5Si_{12}O_{30}$ by Khan et al. (1972) Forbes et al. (1972) proposed the following general formula for milarite-type minerals

$$A_2^{[6]}B_2^{[9]}C^{[12]}D^{[18]}T(2)_3^{[4]}T(1)_{12}^{[4]}O_{30}.$$

Figure 1 presents a simplified structural model for milarite-type minerals, in which most of the possible cationic positions given by the above formula are identified. Černý et al. (1980) have successfully applied this formula to the crystal chemistry of milarite sensu strictu.

For the mineral roedderite, $(Na,K)_2(Mg,Fe^{2+})_5Si_{12}O_{30}$, then only known from meteorites, Forbes et al. (1972) gave the following site allocations relative to their general formula

$$(Mg,Fe^{2+})_2^{[6]}Na^{[9]}(K,Na)^{[12]}Mg_3^{[4]}Si_{12}^{[4]}O_{30}.$$

This implied that in roedderite the positions A, C, T(2), and T(1) are completely filled, while B is only occupied by 50%, and D is vacant. In order to comply with the conventional sequence of elements used for silicate minerals we prefer to rewrite this roedderite formula here as

$$(K,Na)^{[12]}Na^{[9]}(Mg,Fe^{2+})_2^{[6]}Mg_3^{[4]}Si_{12}^{[4]}O_{30}.$$

More recently, Hentschel et al. (1977, 1980) have found the mineral roedderite as a rather common product of crystallization within cavities of certain basement xenoliths ejected by the tephritic Bellerberg volcano, Eifel, West Germany. Microprobe analyses of the euhedral crystals had indicated that these first terrestrial roedderites showed compositional variations from one xenolith to the other, mainly with regard to the Na/Mg and Fe/Mg ratios (Hentschel et al. 1980). In the course of continued analytical work on a greater number of xenoliths found by local mineral collectors, crystals with the typical roedderite morphology and physical properties were detected which, however, exhibited aberrant chemical compositions no longer fitting the roedderite formula so that the definition of a new mineral species seemed warranted. A first account on these new findings was published by Abraham et al. (1980). The name

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Offprint requests to: W. Schreyer

eifelite proposed by these authors was meanwhile approved by the IMA Commission on New Mineral Names. It stands for the region in which the new mineral occurs. Type material is deposited at Institut für Mineralogie, Ruhr-Universität, D-4630 Bochum, West Germany.

In the present paper the new mineral eifelite is described emphasizing its chemical variations and structural properties which are critical for a distinction against roedderite. From the chemical point of view eifelite carries as major elements only those included in the general formula of the osumilite subgroup. However, their amounts and ratios cannot be accommodated in this formula.

Occurrence

Eifelite occurs as beautiful euhedral crystals (Fig. 2) in vesicles of a contact metamorphosed basement gneiss xenolith that was found within the leucite tephrite lava quarried at Bellerberg, about 2 km north of Mayen in the eastern part of the Eifel, West-Germany. The xenolith consists mainly of quartz and sanidine; accessories occurring in the vesicles are pyroxene, amphibole, tridymite, hematite, and pseudobrookite. In contrast to the variably coloured hexagonal crystals of the osumilite group minerals mentioned by Hentschel et al. (1980) those of eifelite are always colourless or just very faintly yellow or green. However, we are hesitant to consider this as a critical distinction, because it may only apply to the limited amount of material studied.

Chemistry

Analytical work was performed with an electron microprobe MICROSCAN MKV from Cambridge Instruments Co. using the procedures as given by Abraham and Schreyer (1973). The results obtained on two eifelite crystals are listed in Table 1 (columns 1 and 2), but for comparison and subsequent discussion of crystal structure two analyses of roedderite (*A* and *B*) from the same locality as reported by Hentschel et al. (1980) are given as well. In addition, a new analysis of a blue roedderite crystal *D* is presented, because this crystal was also studied structurally; it is close in composition to roedderite sample C of Hentschel et al. (1980).

A microchemical Li analysis on one eifelite crystal from Bellerberg (sample 97) which, however, was not studied by microprobe, gave 0.1 weight % Li_2O. As this small amount does not change the relations appreciably, lithium will be largely disregarded in the discussion below.

The high totals for nearly all analyses of Table 1 leave no room for H_2O molecules, which, although well known for milarite, are apparently absent in minerals of the osumilite subgroup (Forbes et al. 1972; Berg and Wheeler 1976).

It can be seen from Table 1, especially from the recalculated formulae, that, with most element concentrations being rather constant, the main compositional differences between eifelite and roedderite lie in the amounts of sodium and magnesium present. There is a negative correlation between these two elements, essentially following a substitutional mechanism $2Na^+ \rightarrow Mg^{2+}$, which was already proposed by Hentschel et al. (1980) to explain compositional variations amongst their roedderite *A* and *B*.

These relationships are graphically depicted in Fig. 3 where Na is plotted against the sum of the divalent cations (Mg+Fe+Mn+Cu+Zn) per formula unit for analyses of Table 1. Except for the roedderite sample D, which has an alkali-deficiency possibly due to a partial substitution $Fe^{3+} \rightarrow Fe^{2+} + Na^+$ (Hentschel et al. 1980), all other analyses fall rather close to the substitution line $2Na^+ \rightarrow R^{2+}$, which connects the roedderite composition $KNaMg_5Si_{12}O_{30}$ with ideal eifelite $KNa_3Mg_4Si_{12}O_{30}$. Our eifelite sample 2 (Table 1) approaches the latter formula. Figure 3 thus provides evidence for a gradual transition of some of the Bellerberg osumilite group minerals from roedderite to the new highly sodic member eifelite by means of the 2Na for R^{2+} substitution. Note that the additional presence of Li assumed to accupy R^{2+}-positions would raise the plotted points in Fig. 3 vertically, so that analysis No. 1 might actually also lie closer to the ideal curve.

Table 1. Chemical compositions of two eifelite crystals 1 and 2, and of roedderite crystals *A*, *B*, *D*. (Total Fe as FeO)

	1	2	*A*	*B*	*D*
SiO_2	71.06	69.06	71.10	71.32	71.10
TiO_2	0.06	0.03	0.07	0.07	0.10
Al_2O_3	0.79	0.57	0.61	0.36	0.27
Cr_2O_3	0.06	0.00	0.00	0.00	0.00
FeO	0.48	0.19	0.97	0.49	4.45
MnO	0.46	0.53	0.37	0.31	0.26
CuO	0.08	0.16	0.13	0.00	0.00
ZnO	0.34	0.43	0.34	0.00	0.00
MgO	16.25	15.47	18.85	17.86	16.50
K_2O	4.18	4.16	4.33	4.16	4.28
Na_2O	6.48	8.07	3.32	5.29	2.24
	100.24	98.67	100.09	99.86	99.20
Numbers of ions on the basis of 30 oxygens:					
Si	11.98	11.92	11.92	11.99	12.10
Al	0.02	0.08	0.08	0.01	0.00
	12.00	12.00	12.00	12.00	12.00
Ti	0.01	0.00	0.01	0.01	0.01
Al	0.14	0.04	0.04	0.06	0.05
Cr	0.01	0.00	0.00	0.00	0.00
Fe^{2+}	0.07	0.03	0.14	0.07	0.63
Mn^{2+}	0.07	0.08	0.05	0.04	0.04
Cu	0.01	0.02	0.02	0.00	0.00
Zn	0.04	0.05	0.04	0.00	0.00
Mg	4.08	3.98	4.71	4.48	4.19
	4.43	4.20	5.01	4.66	4.92
K	0.90	0.92	0.92	0.89	0.93
Na	2.12	2.70	1.08	1.72	0.74
	3.02	3.62	2.00	2.61	1.67

The gradual character of the compositional change shown in Fig. 3 causes, of course, a nomenclature problem. We propose here to restrict the name eifelite to osumilite group minerals with (Na,K)≧3, Na>K, Mg>Fe, and Si/Al>7. The dividing line against roedderite is shown in Fig. 3. It lies at the composition $KNa_2Mg_{4.5}Si_{12}O_{30}$ which was first defined by Abraham et al. (1980) as the new mineral.

It is clear that the substitutional mechanism 2Na for R^{2+} leading from roedderite to eifelite raises also structural problems concerning the accommodation of the additional

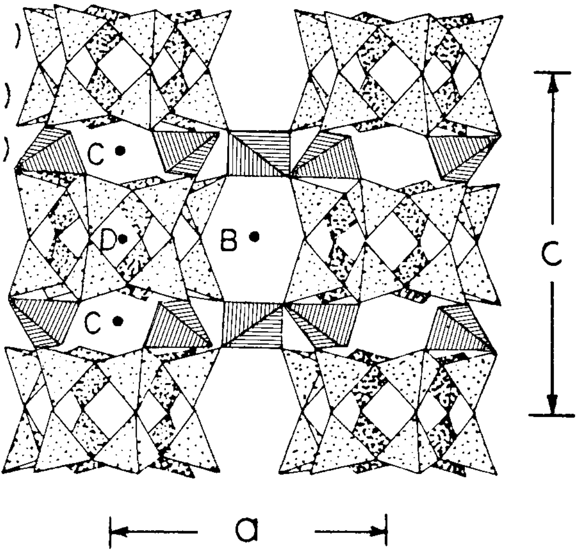

Fig. 1. Crystal structure of milarite as simplified by Černý et al. (1980) viewed parallel to [101]. Octahedral A sites located at the level of the T(2) tetrahedra are omitted for clarity

sodium ions in the milarite-type structure (Forbes et al. 1972). For this reason crystal structure analyses were performed on four crystals and will be discussed in a subsequent section.

Regarding its chemical behaviour eifelite is insoluble in HCl and H_2SO_4.

Physical Properties

The colourless to very light yellow or green, transparent crystals of eifelite attain diameters of nearly 1 mm and exhibit hexagonal morphology (Fig. 2). Their habit varies from platy through equidimensional to prismatic. The dominant faces are $\{10\bar{1}0\}$ and $\{0001\}$ with subordinate $\{11\bar{2}0\}$ and occasional $\{10\bar{1}2\}$, very much as it was pictured by Hentschel et al. (1980) for roedderite. The lustre of eifelite is vitreous, its streak white, and no cleavage can be observed.

Eifelite is uniaxial positive with $n_o = 1.5445(5)$ and $n_e = 1.5458(5)$ measured for crystal No. 1 and $n_o = 1.5430(5)$ and $n_e = 1.5443(5)$ for crystal No. 2 of Table 1. Hence the birefringence of eifelite is very low (0.001) and lies indeed decidedly below the values to be derived from the n_e and n_o figures for roedderites as given by Hentschel et al. (1980) in their Table 1, which range from 0.003 to 0.007. (Note that in Table 1 of Hentschel et al. the n_e figures for crystals *B* and *C* were erroneously exchanged.)

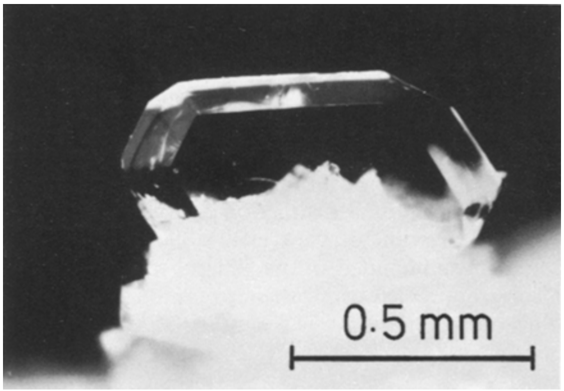

Fig. 2. Euhedral eifelite crystal with platy habit grown within a vesicle on top of finer grained felsic minerals. Bellerberg, sample No. 97 (Fot. Fischkandl)

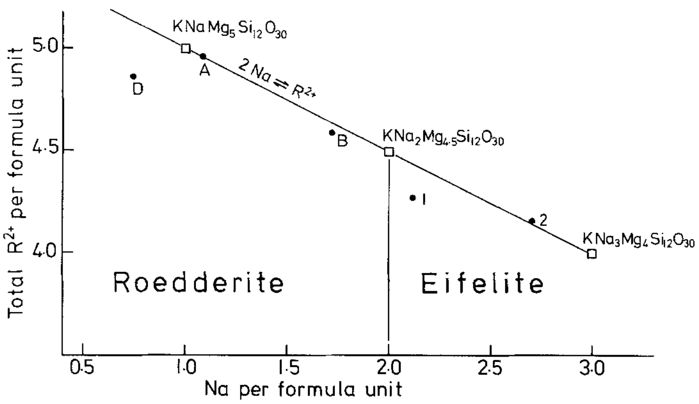

Fig. 3. Sodium versus R^{2+} plot of eifelite and roedderite analyses (solid dots) as taken from Table 1. Formulae and substitution line are idealized. Total Fe was taken as Fe^{2+}

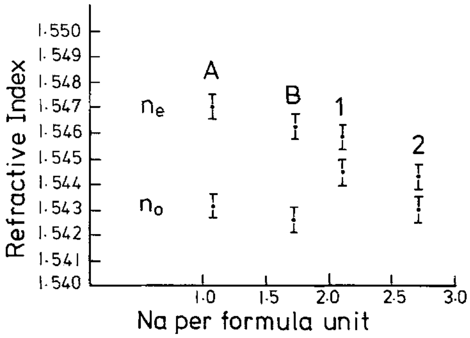

Fig. 4. Refractive indices of four eifelite and roedderite crystals (see Table 1) plotted against their sodium contents

For fair comparison between eifelite and roedderite the refractive indices of the two roedderite crystals *A* and *B* (Table 1) were also measured with the same technique as applied to eifelite, that is using the newly designed spindle stage with internal refractometer (Medenbach 1980). The results are as follows: Roedderite A has $n_o = 1.5432(5)$, $n_e = 1.5470(5)$ with a measured birefringence of 0.004; roedderite *B* has $n_o = 1.5426(5)$, $n_e = 1.5462(5)$, and measured birefringence 0.003. In Fig. 4 the refractive indices of all four crystals are plotted against sodium contents per formula unit as already used in Fig. 3. While it must be borne in mind that minor variations of the heavier cations Cr, Fe, Mn, Cu, and Zn will undoubtedly have a strong influence on the refractive indices, it does seem that – as a first approximation – there is a trend towards lower birefringence from roedderite to eifelite, that is as a function of the $2Na \rightarrow Mg$ substitution.

In the eifelite crystals no pleochroism was observed. The calculated density of crystal No. 2 is 2.67 g/cm^3.

The strongest lines in the powder X-ray diffraction pattern of eifelite obtained with a Gandolfi camera (CuK_α, Ni-filtered) are (*d*(*I*)*hkl*): 7.07(5)(002), 5.11(5)(110), 4.43(6)(200), 4.14(5)(112), 3.75(9)(202), 3.54(4)(004),

Table 2. Crystal data and R factors for the two eifelite (1–2) and two roedderite crystals (*A*, *D*) studied (compare Table 1)

	1	2	*A*	*D*
a [Å]	10.150 (5)	10.137 (5)	10.142 (5)	10.138 (5)
c [Å]	14.223 (6)	14.223 (6)	14.281 (6)	14.309 (6)
V [Å^3]	1268.98	1265.73	1272.14	1273.63
R	0.027	0.042	0.047	0.064

3.26(10)(211), 2.91(5)(114), 2.76(5)(204), 2.73(4)(213)(302), 2.56(3)(220), 2.21(3)(400), 2.03(4)(320), 1.914(3)(411), 1.864(5)(323)(412), 1.76(5)(008)(324). For practical purposes this pattern is identical with that of roedderite.

Thus, in summary, there is as yet no physical property which would allow a fully reliable distinction between eifelite and roedderite, and indeed only chemical analysis can lead to incontrovertible identification of these two osumilite group minerals.

Crystal Structure Analyses

Single crystal investigations were performed on the two eifelite crystals 1 and 2 as well as on two of the roedderite crystals (*A* and *D*) of Table 1. The intensity data were collected by the ω-scan method on a computer-controlled four-cycle diffractometer (SYNTEX R 3) using MoK_α radiation ($\lambda = 0.71069$ Å) and a graphite monochromator. The reflections up to $\sin\theta = 35°$ yielded data sets of about 3600 intensities each, which were reduced to about 850 unique data for each crystal. No absorption correction was applied as the crystal fragments used were nearly equidimensional.

The space group was found to be $P\frac{6}{m}\frac{2}{c}\frac{2}{c}$ for all crystals investigated, $Z=2$. The remaining crystal data and the non-weighted *R* factors attained using reflections with $I > 2.5\sigma$ are listed in Table 2. Refinement was achieved using the SHELX-TL computing system. Interestingly enough, the lowest cell volume was found for eifelite crystal 2, which is the most highly Na substituted one (see Tables 1 and 2). However, as for the optical data, other chemical variations may influence this property as well.

As the basis for our structure refinements the atomic positions for synthetic $K_2Mg_5Si_{12}O_{30}$ as determined by Khan et al. (1972) were used. It was expected that at least some of the additional Na of eifelite would be located in the 9-coordinated B-position (see Fig. 1 and formula in Introduction) of Forbes et al. (1972), which is only half occupied in the synthetic phase studied by Khan et al. (1972). Furthermore, as a result of the substitution 2 Na for Mg, significantly smaller site occupancy in the six-coordinated *A*-position, or even in the four-coordinated *T*(2) position, should occur. Neither of these expectations was fulfilled.

Refinements of the site occupancy factors for all four crystal structures showed that all *A*- and *T*(2)-positions are fully occupied, whereas in the B-position in fact no atom could be located. If in the calculations atoms were attributed to the *B*-position, the *R* value rose significantly and the anisotropic thermal vibration parameter U33 attained impossibly high values near 0.8. The possible atomic position $D^{[18]}$ of the Forbes et al. (1972) formula as well as a potential site $E^{[12]}$ in equipoint 6(g) as defined by Khan et al. (1972) were also found unoccupied.

On the positive side, the refinement yielded atomic positions (Table 3) and interatomic distances (Table 4) for the sites $A^{[6]}$, $C^{[12]}$, $T(2)^{[4]}$, and $T(1)^{[4]}$, but also for the oxygens O_1 through O_3. These parameters are principally similar to those found by Khan et al. (1972) for $K_2Mg_5Si_{12}O_{30}$. Therefore, undoubtedly the eifelite and roedderite crystals studied must have an atomic arrangement in essential agreement with a milarite-type structure as shown in Fig. 1. As in $K_2Mg_5Si_{12}O_{30}$ we always found $C^{[12]}$ occupied. In agreement with the remarkable constant cell contents of all our crystals in potassium (see Table 1) it is this relatively heavy atom which was located by the refinement in $C^{[12]}$.

The important finding during refinement was that a new structural position could be defined which accommodates part of the sodium. It occurs close to the vacant $B^{[9]}$-position, but, unlike this crystallographic 4(d) site, it does not lie within the mirror plane at (1/3, 2/3, 0) but slightly off that plane. By changing this site into an 8(h) position with (1/3, 2/3, *z*), which is also compatible with the space group $P\frac{6}{m}\frac{2}{c}\frac{2}{c}$, the Na atom could be fixed by the refinement at a z-parameter of about 0.02 for all four crystal structures investigated, and the temperature factors became acceptable (Table 3). As a result of this shift the coordination of the position is lowered from [9] to [6], and the mean interatomic distances for next neighbors are changed from M–O = 2.88 Å (Khan et al. 1972) to 2.63 Å (Table 4). Table 4 also shows quantitatively, how far away the remaining three oxygens of the former 9-coordination in B lie. The new site with reduced coordination is here called *B*′. A projection of the eifelite structure parallel **c** using the parameters for crystal No. 2 (Table 3) is given in Fig. 5, where the new octahedral *B*′ site is marked as Na.

It is important to emphasize here that the $B^{[9]}$-position is not only inapplicable to the Na-rich eifelites but also to the roedderite crystals studied here. Thus these roedder-

Table 3. Atomic positions, site occupancy factors, and anisotropic thermal parameters for eifelite crystal No. 2

Site	*x*	*y*	*z*	S.O.F.	U11	U22	U33	U23	U13	U12
C	0.000	0.000	0.250	0.074	0.027	0.027	0.030	0.000	0.000	0.013
B′	0.333	0.667	0.019	0.138	0.029	0.029	0.050	0.000	0.000	0.015
A	0.333	0.667	0.250	0.167	0.006	0.006	0.004	0.000	0.000	0.003
T(2)	0.500	0.000	0.250	0.250	0.014	0.007	0.008	0.000	0.000	0.004
T(1)	0.117	0.351	0.111	1.000	0.014	0.015	0.013	0.001	0.000	0.007
01	0.136	0.394	0.000	0.500	0.033	0.029	0.016	0.000	0.000	0.016
02	0.220	0.275	0.135	1.000	0.028	0.032	0.028	0.003	0.003	0.023
03	0.160	0.498	0.170	1.000	0.027	0.020	0.023	0.007	0.006	0.013

Table 4. Interatomic distances in eifelite crystal No. 2

Site	Atoms	Distances
C	$K^{[12]} - O_2$	12×3.035 (4) Å
B'	$Na^{[6]} - O_1$	3×2.495 (4) Å
	$- O_{3A}$	3×2.758 (4) Å
subsequent sphere of "coordination":		
	$Na - O_{3B}$	3×3.203 (4) Å
A	$(Mg,Na)^{[6]} - O_3$	6×2.077 (4) Å
T(2)	$Mg^{[4]} - O_3$	4×1.989 (3) Å
T(1)	$Si^{[4]} - O_1$	1.620 (2) Å
	$- O_2$	2×1.621 (2) Å
	$- O_3$	1.580 (2) Å

Values for oxygen-oxygen distances can be obtained from the authors upon request

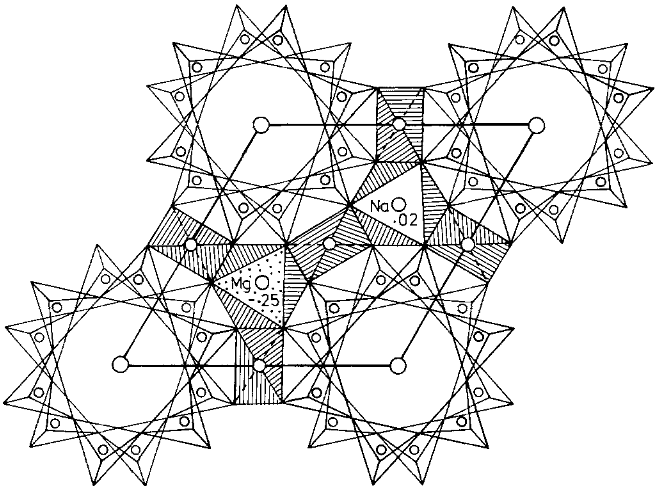

Fig. 5. Projection of the structure of eifelite crystal No. 2 along *c*. Na marks the new sixfold *B'*-site, Mg the *A*-site not shown in Fig. 1. Figures indicate *z*-parameters

Table 5. Theoretical and structurally derived alkali occupancy in *C*- and *B'*-sites of eifelite (1, 2) and roedderite (*A*, *D*) crystals as defined chemically in Table 1

	1	2	*A*	*D*
Theoretical occupancy (cf. Table 1):				
Na in $B'^{[6]}$	1.55	1.90	1.08	0.66
K in $C^{[12]}$	0.90	0.92	0.92	0.93
Occupancy derived from structure determination:				
Na in $B'^{[6]}$	1.56	1.65	1.05	0.41
K in $C^{[12]}$	0.88	0.89	0.87	0.96

ites are, strictly speaking, not isostructural with $K_2Mg_5Si_{12}O_{30}$ of Khan et al. (1972) but exhibit octahedral sodium as well.

The displacement of the Na atom in $B'^{[6]}$ from the mirror plane along c amounts to about 0.28 Å (0.02×14.2). As this value is small relative to the size of a sodium ion in sixfold coordination (near 1.1 Å), the new position $B'^{[6]}$ cannot be occupied according to its equipoint symbol 8(h) by 4 atoms per formula unit ($Z=2$), but only by 2 atoms just as the $B^{[9]}$ position in the general formula of Forbes et al. (1972). However, with $B'^{[6]}$ only partly occupied, the symmetry of the crystals should be reduced against that of the space group $P\frac{6}{m}\frac{2}{c}\frac{2}{c}$, provided the Na was incorporated in $B'^{[6]}$ in an ordered manner. In this respect it is important to note that the thermal ellipsoid of Na in $B'^{[6]}$ was found to be strongly elongated parallel to the *c*-axis, and this is interpreted to indicate a *statistical* distribution of sodium on both sides of the mirror plane which is again compatible with the above space group.

Interestingly enough, site deviations of the B position have been recorded previously for milarite-structure minerals. Bakakin et al. (1975) were first to discover that the H_2O molecules of both milarite and armenite are not situated in 4(d) = *B* but in 8(h) = *B'* with *z*-parameters of 0.06 and 0.07, respectively. For sodgianite, however, they found Na in $B(Z=0)$. According to Černý et al. (1980) H_2O molecules as well as Na, K, and Ca in milarite may occupy a *B'*-position in the present definition with a *z*-parameter of 0.02 like that found for eifelite and roedderite in our study.

With the $B'^{[6]}$-position available in eifelite for Na, and with the $A^{[6]}$- and $T(2)^{[4]}$-positions completely filled, the chemical requirements of the 2Na for Mg relationship between eifelite and roedderite (Table 1 and Fig. 3) can only be met, if the relevant number of Mg atoms are truly replaced by part of the Na. Because of the size of the Na ions this replacement can only take place in the $A^{[6]}$-, but not in the $T(2)^{[4]}$-position.

The structure determinations of eifelite crystals 1 and 2, and of roedderites *A* and *D*, were also used as an independent test on the validity of the substitution 2Na → Mg leading from roedderite to eifelite (Fig. 3). Therefore, the occupancy of sites *C* and *B* was determined from the crystal structures and compared with the values derived theoretically from the chemical analyses of Table 1. In the latter case, it was assumed that part of Na occupies the *A* sites to the extent that all $A+T(2)$-positions are filled to reach the total of 5.00 per formula unit. The result of this comparison is presented in Table 5 and shows clearly an increase of Na in *B'*[6] in the order *D*-*A*-1-2, that is in the same order as total Na increases from roedderites to eifelite in Fig. 3. In fact, the individual theoretical values and structural values are remarkably close, although obviously a summation of analytical errors and the neglect of Li will introduce uncertainties in the theoretical values and although there are also limits in the structural occupancy determination. Thus, Table 5 may confidently be taken as good proof for the 2Na → Mg substitutional relationship between roedderite and eifelite.

In roedderite crystal *D* a significant amount of iron was found analytically (Table 1). Nevertheless, the iron could not be located unequivocally in the structure. However, as the temperature vibration factor for the $A^{[6]}$ site in crystal *D* is slightly higher than for *T*(2), we presume that Fe substitutes for Mg in the former site. This can also be true, if Fe is partly trivalent as suggested by Hentschel et al. (1980) as a balance for the alkali deficiency in their roedderite *C*, which, strictly speaking, is not proper roedderite because of the prevalence of K over Na.

Summarizing the structural findings, the site occupancy for the eifelite-roedderite crystals studied can be expressed by the following formula

$$\underbrace{\mathrm{K}}_{\mathrm{C}^{[12]}}\quad \underbrace{\mathrm{Na}_{1+x}}_{B'^{[6]}}\quad \underbrace{(\mathrm{R}^{2+,3+}_{2-x}\mathrm{Na}_x)}_{A^{[6]}}\qquad \underbrace{\mathrm{Mg}_3}_{T(2)^{[4]}}\quad \underbrace{\mathrm{Si}_{12}}_{T(1)^{[4]}}\quad \mathrm{O}_{30}.$$

We emphasize again that even in the Na-poor roedderite crystals A and D sodium occupies $B'^{[6]}$ and not $B^{[9]}$ as K does in $K_2Mg_5Si_{12}O_{30}$ (Khan et al. 1972). This is obviously due to the smaller size of Na versus K.

The above site occupancy, which probably applies generally to roedderite-eifelite minerals, indicates that the composition $KNa_3Mg_4Si_{12}O_{30}$ (Fig. 3) represents an absolute end member of the series, because $B'^{[6]}$ can be filled by not more than 2 Na, that is x in the general formula must be equal to or smaller than 1.0.

With eight cations per formula unit outside the hexagonal double rings eifelite of the ideal end member composition has the highest cation occupancy of any osumilite group mineral. Osumilite itself contains only six, roedderite and merrihueite seven cations. In milarite the number of non-ring cations ranges between 6 and 6.5, but, in addition, molecular H_2O may partly occupy the $B'^{[6]}$ site (Černý et al. 1980), which is filled by Na in eifelite.

For the eifelite end member $KNa_3Mg_4Si_{12}O_{30}$ a corresponding pure sodium compound, $Na_2Mg_2Si_6O_{15}$ (1:2:6), exists in the system $Na_2O-MgO-SiO_2(-H_2O)$ (see Schairer et al. 1954; Witte 1975). However, a structural analysis by Cradwick and Taylor (1972) showed that this compound does not have a milarite-type structure but is isostructural with tuhualite, $(Na,K)_2Fe^{2+}_2Fe^{3+}_2Si_{12}O_{30}\cdot H_2O$, and that the Na ions have 9 to 10 oxygen neighbours within 3.1 Å. Thus it seems that the milarite structure for the roedderite-eifelite series is mainly stabilized due to the presence of one large K atom per formula unit (Table 1) which occupies the $C^{[12]}$-position. Nevertheless, there are striking similarities between the two structures with the six-membered double rings of the milarite-type opening to form a two-tetrahedra-wide corrugated ribbon or crankshaft with a six-membered identity period in tuhualite.

Genesis

The possible mode of formation of the roedderite crystals in vesicles of the basement xenoliths of the Bellerberg volcanics was discussed by Hentschel et al. (1980). In short, roedderite was considered to be a precipitate from highly alkaline, MgSi-rich, but Al-deficient gas phases percolating the xenoliths. The peralkaline chemistry of the new mineral eifelite relative to roedderite confirms and emphasizes this conclusion, and still higher alkali activities, especially of sodium, in the gas phase can be assumed. This is also in agreement with the common occurrence of acmitic pyroxenes in the xenoliths as well as in some altered lavas.

On the other hand, the origin of such strongly alkaline, but Al-deficient gas phases still poses a problem. Although the absence of roedderite-bearing vesicles within the tephrite lava was considered by Hentschel et al. (1980) to be more suggestive of the gas to have originated *within* the xenoliths, a close connection between gas formation and the process of igneous differentiation of the alkaline magma should also be envisaged. If so, the growth of the eifelite-roedderite crystals could be a new Al-deficient variant of the process of fenitization which is so common around alkaline igneous intrusives. Alkali metasomatism in general has long been known from "sanidinite" xenoliths of the Eifel (Brauns 1911; Frechen 1947; Wörner et al. 1982). Thus the unusual gas compositions may have evolved through interaction of magma-derived gases with siliceous xenoliths.

Regarding the magmatic gases that are likely to have contributed essentially alkalies and magnesium, it is interesting to note that supercritical hydrous fluids of similar compositions were encountered in experimental work by Seifert and Schreyer (1966, 1968) in the system $K_2O-MgO-SiO_2-H_2O$ at low pressures.

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