THE AMERICAN MINERALOGIST, VOL. 52, SEPTEMBER-OCTOBER, 1967

A NEW OCCURRENCE OF ROEDDERITE AND ITS BEARING ON OSUMILITE-TYPE MINERALS

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Recently Fuchs, *et al.*, (1966) described a new mineral, roedderite, from the *Indarch* (enstatite chondrite) meteorite. Roedderite is essentially (K_2O, Na_2O):5MgO:12SiO_2, or $(K,Na)_2Mg_5Si_{12}O_{30}$. This was the first natural occurrence of a compound which Roedder (1951) encountered in his study of the synthetic system K_2O-MgO-SiO_2. Fuchs, *et al.*, presented six microprobe analyses of which one was arbitrarily chosen for comparison here (Table 2). In addition, synthetic K_2O:5MgO:12SiO_2, which was kindly supplied by Dr. E. Roedder, was analyzed by microprobe and the results are presented also. Prior to this the 1:5:12 proportions for this synthetic material were not the result of direct analysis but were based on a careful geometrical analysis of the phase diagram for this system (Roedder, 1951). This was the only method possible for microscopic grains prior to the ready availability of the electron microprobe.

In some recent work on silicate inclusions within graphite nodules in the octahedrite iron meteorite, *Wichita County* (USA), a new occurrence shows a major compositional variation which bears directly on the problem of osumilite-type phases, and indirectly on cordierite (Miyashiro, 1956; Schreyer and Schairer, 1962).

In Table 1 are given the X-ray powder diffraction patterns of the new roedderite, osumilite from Japan (both measured by the writer), the Indarch roedderite, Roedder's original pattern of K_2O:5MgO:12SiO_2, and the sodic analog Na_2O:5MgO:12SiO_2 (unpublished work of Schairer, Yoder, and Keene). The latter was obtained by measuring a diffractometer tracing which was reproduced in Schreyer and Schairer (1962). These patterns are strikingly alike. Films set adjacent to each other are even more striking and without careful scrutiny it is difficult to tell the Wichita County roedderite from osumilite in spite of the large differences in composition.

In addition to these there are the structurally similar minerals milarite, and armenite, as well as other possible phases in the system MgO-Al_2O_3-SiO_2, as discussed by Schreyer and Schairer (1962). They concluded that "there are thus at least five crystalline compounds of drastically different bulk compositions which nevertheless are presumably isostructural." The structure referred to here was determined by Miyashiro and is based on double hexagonal rings, $(Al,Si)_{12}O_{30}$.

The new roedderite was analyzed with an ARL electron microprobe

TABLE 1. X-RAY POWDER DIFFRACTION DATA

Roedderite, *Wichita Co.* meteorite		Osumilite, Sakkabira, Japan		Roedderite, Indarch meteorite (from Fuchs, 1966 and Fuchs et al., 1966)		Synthetic $K_2O:5MgO:12\ SiO_2$ (from Roedder, 1951)		Synthetic $Na_2O:5MgO:12\ SiO_2$ (from Schreyer and Schairer, 1962)	
I	*d*(Å)	*I*	*d*(Å)	*I*	*d*(Å)	*I*	*d*(Å)	*I*	*d*(Å)
10	8.827	30	8.827					w	8.84
50	7.126	70	7.126	60	7.15	s	7.12	m	7.14
10	5.637								
10	5.532	60	5.532	40	5.540	vw	5.54	w	5.51
80	5.062	70	5.091	20	5.084	s	5.13	s	5.04
90	4.383	60	4.392	30	4.391	vs	4.43	s	4.44
80	4.129	80	4.129	10	4.126	s	4.14	w	4.11
5	4.018	10	4.018						
90	3.729	80	3.745	70	3.747	vs	3.74	s	3.74
80	3.567	60	3.567	100	3.570	s	3.53	s	3.56
5	3.490			10B	3.35				
30	3.295	60	3.300	30	3.309			m	3.30
100	3.229	100	3.225	80	3.239	vvs	3.25	vs	3.22
10	3.006	40	3.006					w	3.01
10	2.940								
90	2.916	90	2.916	70	2.922	vs	2.90	s	2.91
10	2.829	10	2.838						
90	2.761	90	2.770	40	2.772	vs	2.76	m	2.76
30	2.717	30	2.721	10	2.720			w	2.71
		10	2.628						
30	2.566	30	2.569						
60	2.531	40	2.536	10	2.543	s	2.55	m	2.54
20	2.432	20	2.432	10	2.435	wB	2.456	vw	2.43
						wB	2.415		
10	2.393	40	2.388	10	2.375			vw	2.39
20	2.315	30	2.321						
10	2.292								
10	2.259					vwB	2.266		
20	2.201	10	2.201						
40	2.168	30	2.168	10	2.171	mB	2.175	w	2.16
30	2.099	10	2.096	10B	2.094	w	2.113	vw	2.10
10	2.067	10	2.069			vw	2.072		
50	2.012	50	2.014	10	2.019	mB	2.016	m	2.01
5	1.937			5B	1.936				
10	1.916	10	1.918	5	1.902	wB	1.914	w	1.90
40	1.897								
40	1.852	50	1.853	10	1.853	mB	1.852	w	1.85
+ 31 additional spacings to 0.7969		+56 additional spacings to 0.7793		+11 additional reported spacings		+33 additional spacings to 0.7795		No additional spacings could be measured (see text)	

All intensities are visual estimates. vs = very strong, s = strong, m = medium, w = weak, vw = very weak, B = broad or diffuse line. All measurements were made on a 114.59 mm. powder camera using Cu radiation.

and the results were corrected for detector deadtime, background, X-ray production efficiency, absorption, and fluorescence (where necessary) as outlined in Smith (1965). The standards used were: K-feldspar from Asbestos, Quebec (Smith and Ribbe, 1966); albite from Amelia Courthouse, Virginia (Ribbe and Smith, 1966); clinopyroxene H-1 from Webster, North Carolina (Smith, 1966). Because of the microgram quantities available it was impossible to analyze for the presence of H_2O by any method (chemical or infrared). Osumilite apparently has constituent water although Miyashiro says, "probably the water content is not essential to this mineral." This seems reasonable for the water occurs inside the double hexagonal ring cavities and could just as well be absent. Nevertheless, as noted below, this roedderite coexists with the amphibole, richterite, which contains approximately 1.1 wt.% water (Olsen, 1967). Thus it is entirely possible that water could be present in the associated roedderite. The analysis total is slightly low but within the analytical error, and cannot be used as evidence. This question cannot be answered at the present time. Fortunately it does not affect the results presented here.

TABLE 2. ELECTRON MICROPROBE ANALYSES

	1	2	3
SiO_2	68.0	70.8	70.7
Al_2O_3	2.5	0.4	—
FeO	0.4	1.0	—
MgO	19.0	19.6	19.8
Na_2O	5.3	4.2	—
K_2O	3.8	3.4	9.3
Total	99.0	99.4	99.8
Na/Na+K	0.68	0.65	—
Mg/Mg+Fe	0.99	0.97	—
Si/Si+Al	0.96	1.0	—

1. Roedderite from Wichita County meteorite; $(Na_{1.7}K_{.8})(Mg_{4.8}Fe_{.1})(Al_{.5}Si_{11.5})O_{30} = (Na, K)_{2.5}(Mg, Fe)_{4.9}(Al, Si)_{12}O_{30}$.
2. Roedderite from *Indarch* meteorite (from Fuchs, et al., 1966); $(Na_{1.4}K_{.7})(Mg_{4.9}Fe_{.1})Si_{11.9}O_{30} = (Na, K)_{2.1}(Mg, Fe)_5Si_{11.9}O_{30}$.
3. Synthetic $K_2O : 5\ MgO : 12\ SiO_2 = K_2Mg_5Si_{12}O_{30}$.

From the analyses it is clear that this new roedderite is more aluminum and alkali rich than the Indarch roedderite. The analysis was recalculated to give a total of 30 oxygens. This resulted in 0.5 atoms Al substituted for 0.5 atoms of Si and a concomitant excess of 0.5 alkali atoms in this empirical formula. The excess alkali is "stuffed" elsewhere in the structure and maintains over-all charge balance. This is the first natural example of significant solid solution in this structural type in the larger system (K_2O, Na_2O)-MgO-Al_2O_3-SiO_2, and bears out the prediction of Schreyer and Schairer (1962): "The finding of a ternary compound with an osumilite-like structure in the system MgO-Al_2O_3-SiO_2, although metastable, enhances the possibility of encountering osumilite-like compounds in the systems K_2O-MgO-Al_2O_3-SiO_2 and Na_2O-MgO-Al_2O_3-SiO_2." Similarly Miyashiro (1956) suggested variability in the alkali content of osumilite and this would, of course, require other adjustments to maintain charge balance.

In contrast to osumilite and the *Indarch* roedderite, both hexagonal (optically uniaxial) minerals, this mineral is slightly biaxial. The optical parameters are: $\alpha = 1.536 \pm 0.002, \beta \sim 1.536, \gamma = 1.543 \pm 0.002, (+)2V = 5°$–$8°$ (estimated), no dichroism observed. Schreyer and Schairer (1962), in discussing optical factors, note that biaxial optics do occur in the over-all osumilite-type group and say that for certain compositions ordering in the lattice may produce lower symmetry than hexagonal. Miyashiro (1965) mentions a report in the literature (Taneda, 1950) of a "cordierite" with a small positive optic axial angle. He concludes this may actually be an osumilite. For the roedderite in the *Wichita County* meteorite the biaxial character is small enough that it is not possible to measure the β index with the facilities available, nor is it possible to compute a value significantly different from α taking the optic angle at a maximum 8°.

In the case of the *Indarch* roedderite the associated nonopaque minerals are: roedderite, enstatite (En 100), plagioclase (Ab 90), and tridymite. In the *Wichita County* meteorite occurrence the coexisting nonopaque minerals are: roedderite, olivine (Fo 99), plagioclase (Ab 99), richterite, for which $Mg/(Mg+Fe) = 0.99$, and finally a new silicate mineral. This latter mineral is a Na-Mg-Cr silicate (apparently a sorosilicate) and it is currently being studied.

ACKNOWLEDGMENTS

The writer is grateful to Louis Fuchs, Chemistry Div., Argonne National Laboratory, for advice and criticism, and to Dr. Edwin Roedder for the use of his synthetic K_2O:5MgO:12SiO_2 crystals. This work was supported by National Science Foundation grant GA-307, and this support is gratefully acknowledged.

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THE AMERICAN MINERALOGIST, VOL. 52, SEPTEMBER-OCTOBER, 1967

SYNTHESIS OF CALCIOGADOLINITE

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Previous studies of synthetic gadolinite, $Fe^{+2}Y_2Be_2Si_2O_{10}$, by the writer (1965, 1966) have demonstrated that this mineral can form numerous solid solution series involving the replacement of Fe^{2+} and Y^{3+} by other divalent and trivalent metals respectively. The crystallographic properties and chemical stabilities of the resulting synthetic phases were also shown to vary systematically with the ionic radii of the elements involved in these substitutions. These investigations have now been extended to include the calciogadolinites.

Calciogadolinite, containing 11.9 weight percent CaO, was first described by T. Nakai (1938) from the pegmatite of Tadachi, Nagano, Japan. Nearly all other chemical analyses of natural gadolinite similarly report CaO in amounts that commonly range up to 3 weight percent (Dana, 1892), (Nagashima, 1960). The role of this calcium remained uncertain because of the possibility of admixed impurities in the samples analyzed and the unknown extent to which calcium could be incorporated into the gadolinite structure. The fact that the calcium reported in these analyses is frequently accompanied by ferric iron in an approximately 1:1 mole ratio, however, does suggest the presence of a solid solution series involving the partial replacement of Y^{3+} by Ca^{2+} with valence compensation being achieved by the concomitant substitution of Fe^{2+} for Fe^{2+} according to the general formula $(Fe_{1-X}^{2+}Fe_X^{3+})(Y_{2-X}Ca_X)Be_2Si_2O_{10}$. The calcium-rich end member of this series should contain a maximum of 13.38 weight percent CaO. Such calciogadolinite and several of its

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Mineralogical Contribution No. 529.

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