Metastability of an Osumilite End Member in the System K$_2$O-MgO-Al$_2$O$_3$-SiO$_2$-H$_2$O and its Possible Bearing on the Rarity of Natural Osumilites

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Abstract. A synthetic end member of the osumilite group with a composition KMg$_3$Al$_3$(Si$_2$Al$_2$-O$_{10}$)$_{1-x}$H$_2$O was obtained as a single phase through short-time hydrothermal treatment of a glass of the appropriate composition at 1 kb $P_{H_2O}$ and 650–700°C. Its optical and x-ray properties are close to those of natural osumilites. At temperatures outside the range 650 to 700°C, and at higher pressures osumilite could not be synthesized as a single phase, even in short runs. With experiment durations of 3 months and more the synthesized osumilite breaks down under the PT conditions of its formation, indicating that it is metastable under these conditions. The stable breakdown products, in the order of increasing temperatures, are: muscovite + amesitic chlorite + quartz; eastonitic phlogopite + cordierite + muscovite + quartz; and cordierite + K-feldspar + quartz. Natural osumilite of Sakkabira, Japan, treated under similar conditions also broke down. These results suggest that the rare mineral osumilite may represent a metastable phase due to rapid and non-equilibrium crystallization common in sanidine-facies environments. Natural myrmekite-like intergrowths of cordierite with quartz and K-feldspar occurring in metamorphic rocks may be the result of the breakdown of pre-existing osumilite phases in these rocks.

Introduction

Osumilite, a mineral akin to and occasionally mistaken for cordierite, was first recognized as a distinct species by Miyashiro (1953). Its general formula was given by Miyashiro (1956) as:

$$(K, Na, Ca) (Mg, Fe^{2+})_2 (Al, Fe^{3+}, Fe^{2+})_3 (Si, Al)_{12}O_{30} \cdot H_2O$$

The crystal structure of osumilite was found to be related to that of the Be-silicate milarite, KCa$_2$(Be, Al, Si)$_3$(Si, Be)$_{12}$O$_{30}$ · 0.5 H$_2$O (Ito, Morimoto and Sadanaga, 1932). Moreover Tennyson (1960) remarked that armenite, BaCa$_2$-Al$_2$Si$_4$Al$_2$O$_{10}$ · 2 H$_2$O (Neumann, 1941) is of the same structural type.

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MgO·Al₂O₃·4 SiO₂ (SCHREYER and SCHAIRER, 1962). On the basis of this knowledge
SCHREYER and SCHAIRER (1962) suggested that osumilite-type phases
might also form within the quaternary system K₂O-MgO-Al₂O₃-SiO₂. The present
paper deals with the hydrothermal synthesis and properties of such an osumilite-

type phase, which was encountered in the course of a more extensive study within
the hydrous system K₂O-MgO-Al₂O₃-SiO₂•H₂O.

According to MIYASHIRO (1956) the osumilite structure consists of six-membered
double rings of the composition (Si, Al)₁₂O₃₀ which are linked by one large 12-
coordinated cation (K, Na, Ca) per formula unit to form stacks. The connection
between these stacks of double rings is made by three cations in tetrahedral
coordination (Al, Fe⁺³, Fe⁺²) and two in octahedral coordination (Fe⁺², Mg) per
formula unit. Concerning the structural water MIYASHIRO (1956) suggested that
it fills the cavities inside the double rings. There would thus be a formal simi-
larly to cordierite, (Mg, Fe⁺²)₂Al₄Si₆O₁₈·xH₂O, which contains six-membered
single rings, (Al, Si)₆O₁₈, with molecular water partially filling the channels
formed by the stacking of these rings (SCHREYER and YODER, 1964). For the
anhydrous osumilite-type alkali magnesium silicates K₂O·5 MgO·12 SiO₂ and
Na₂O·5 MgO·12 SiO₂ SCHREYER and SCHAIRER (1962, Table 3) have pointed out
that the cavities inside the double rings probably accommodate the excess alkali
ions of these compounds.

With the cations available in the system K₂O-MgO-Al₂O₃-SiO₂•H₂O the simplest
theoretical osumilite-type compound to be derived from MIYASHIRO’s general
formula is

\[
\text{KMg}_2\text{Al}_3[\text{Si}_{10}\text{Al}_2\text{O}_{30}] \cdot x\text{H}_2\text{O}
\]

In this structure potassium would only be present between the double rings, and
water may or may not be a constituent.

One of the basic petrological problems concerning this theoretical end member as
well as the whole osumilite groups of minerals is that their bulk chemistry is by
no means unusual and can be represented by assemblages of common rock forming
minerals such as micas, cordierites, quartz and feldspars. Yet osumilite is un-
doubtedly a rare mineral in natural environments. Therefore, one of the goals of
this experimental study was to determine the stability relations of the osumilite
end member synthesized.

Investigations in the Dry System K₂O-MgO-Al₂O₃-SiO₂

The composition of the theoretical osumilite end member, \( \text{KMg}_2\text{Al}_3[\text{Si}_{10}\text{Al}_2\text{O}_{30}] \),
may be related to other phases in the quaternary system K₂O-MgO-Al₂O₃-SiO₂
through the following equation

\[
\text{KMg}_2\text{Al}_3[\text{Si}_{10}\text{Al}_2\text{O}_{30}] = \text{KAlSi}_9\text{O}_8 + \text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18} + 2 \text{SiO}_2
\]

This shows that this composition lies within the join K feldspar --- cordierite ---
SiO₂, which was studied by SCHAIRER (1954) as part of the important join leu-
cite --- cordierite --- SiO₂ in the quaternary system. Fig. 1 shows the position of
the theoretical osumilite end member in the liquidus diagram of this join as given
by SCHAIRER (1954). Although SCHAIRER (1954) working at 1 atmosphere does