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THE THERMAL TRANSFORMATION OF PYROPHYLLITE TO MULLITE

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The synthesis of mullite from pyrophyllite has been investigated by Bradley and Grim (1951), who found that the product was oriented with "three mullite c axis traces parallel with the original pyrophyllite poles (020), (110) and ($\bar{1}10$)." They concluded that the Al octahedra of the pyrophyllite structure remain undisturbed and that mullite strings are formed with moderate ease, unaccompanied by a sharp exothermic reaction.

In the present study the changes occurring on heating pyrophyllite powder and fibers at temperatures ranging from 970°–1200° C. were investigated. Material from Graves Mountain, Georgia was used. No precautions were taken to prevent rehydration after heating.

At 970° C. dehydroxylated pyrophyllite is obtained. The a , b and c axis spacings of this anhydride form are 5.23, 8.90 and 9.33 Å respectively. Fiber photographs taken about the fiber (a) axis show both arcs and spots, indicating varying degree of order in different crystallographic directions. On the zero layer spots correspond to (0k0), arcs to (00l) reflections. (0kl) reflections are very weak or absent and (0k0) reflections occur only for $k=2n$.

After heating the material at 1150° C. the photographs are much weaker. No change occurs in the a axis spacing, but the arcs corresponding to (00l) reflections are no longer present. The repeat distance in the b direction is reduced from 8.90 to 8.75 Å. Some reflections due to mullite appear, notably those of the (00l) series which are observed as spots on the zero layer. The unit cell dimensions of mullite are probably $a=2 \times 7.43$ Å, $b=2 \times 7.58$ Å, $c=5.74$ Å (Bragg 1937).

The following relatively sharp reflections can be detected on the zero layer:

4.37 Å	(020)	pyrophyllite anhydride, modified	$4.37 \times 2 = 8.74$
2.89 Å	(002)	mullite	$2.89 \times 3 = 8.67$
2.19 Å	(040)	pyrophyllite anhydride, modified	$2.19 \times 4 = 8.76$
1.44 Å	(004)	mullite	$1.44 \times 6 = 8.64$

Thus, since the ratio of the b axis spacing of the modified pyrophyllite anhydride to the c dimension of mullite is 3:2, the (0k0) reflection of the former give rise to an almost integral series with the (00l) reflections of the latter.

A photograph of the fiber mounted parallel to the b direction of the original pyrophyllite can be satisfactorily indexed on the assumption that the material is largely mullite, well oriented with c as rotation axis.

At 1200° C. photographs are obtained of preferentially oriented mullite and randomly oriented cristobalite. The morphology of the fibers is preserved. Mullite fibers are arranged with their [110], [010] and [120] directions parallel to the fiber axis. The (110) spacing of mullite is very similar to the *a* axis spacing of pyrophyllite anhydride. The first of the three arrangements thus appears to be favored. In all cases the mullite *c* axis, which corresponds to the direction of the chains of octahedral groups, is perpendicular to the fiber axis and the (001) reflections consequently appear as spots on the zero layer.

The photographs obtained are complex and have not, as yet, been completely interpreted. However, the salient point emerges that the thermal transformation of pyrophyllite to mullite involves an intermediate crystalline phase. It is evident that while extensive breakup of the structure occurs in the direction perpendicular to the sheets, within the sheets order is preserved and only relatively minor rearrangements take place, as envisaged by Bradley and Grim. Crystallization of mullite proceeds gradually over a range of temperature and the associated energy changes are small.

Studies of thermal transformations of clay and other minerals have shown that rearrangement of existing structures commonly occurs in preference to complete disintegration followed by crystallization of a new phase. Only rarely, however, can the successive changes be so readily observed. Detailed analysis of the structures of pyrophyllite anhydride, mullite and the intermediate phase would, no doubt, give valuable information on the type of mechanism involved.

REFERENCES

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LAZULITE FROM YUKON, CANADA

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LOCATION

Lazulite, a relatively rare phosphate mineral, was collected from along the Blow River near Mt. Fitton in the Yukon Territory. This location is 80 miles W.N.W. of Aklavik at 68°30' N. Lat., 138°45' W. Long. The mineral was discovered and collected by B. Cameron in the summer of 1959 while employed by Triad Oil Company.