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HYDROTHERMAL SYNTHESIS OF ANDALUSITE FROM KYANITE

D. F. WEILL,¹ *University of California, Berkeley, California.*

INTRODUCTION

Lacy (1951) reported the growth of andalusite in slates undergoing hydrothermal treatment at temperatures above 760° C., but the method of identifying the andalusite is not mentioned. Roy (1954) wrote of syntheses of andalusite from various mixtures in the system MgO-Al₂O₃-SiO₂-H₂O. The phase had a mean refractive index of $1.635 \pm .004$, and the x -ray diffraction pattern was reported as very similar to that of natural andalusite. All three Al₂SiO₅ polymorphs were reported synthesized by Coes (1955) at 700°-900° C. and 10,000 to 20,000 atmospheres, but the method of identification is not fully described. The hydrolysis of muscovite mica to pyrophyllite and andalusite at temperatures as low as 420° C. at 15,000 psi water pressure was recorded by Hemley (1959). The x -ray pattern given shows a correspondence with three of the strong reflections for andalusite. More recently, Aramaki and Roy (1959, 1962) have reported the growth of a new aluminosilicate, AS(H)-II, of probable composition Al₂SiO₅. This new phase is stated to be identical to that previously identified as andalusite by Roy (1954). Carr and Fyfe (1960) also noted the synthesis of an "andalusite-like" phase, x -andalusite, and found that whereas the x -ray powder diffraction pattern for andalusite and x -andalusite showed an approximate correspondence between some of the strongest lines, the differences left no doubt that they were dealing with a distinct phase. In the light of these later results it is apparent that previous reports of syntheses of andalusite are open to question.

During the course of some hydrothermal experiments in the system Al₂SiO₅-H₂O the author has observed some results which seem to represent stronger evidence for the synthesis of andalusite than has heretofore been reported.

EXPERIMENTAL RESULTS

The synthesis of andalusite is petrologically significant if it is achieved from other mineral phases under conditions which are not far removed

¹ Present address: University of California, La Jolla, California.

from those occurring during metamorphism. In this manner a meaningful order of thermodynamic stability under metamorphic conditions may be determined. Accordingly an attempt was made to synthesize andalusite from the other Al_2SiO_5 polymorphs at hydrothermal conditions not very different from those which have been inferred as its natural environment during metamorphism.

Two anhedral grains of andalusite from Minas Gerais, Brazil were used. Andalusite-K was sealed in a platinum capsule with 30 milligrams of powdered kyanite and 50 milligrams of distilled water, and andalu-

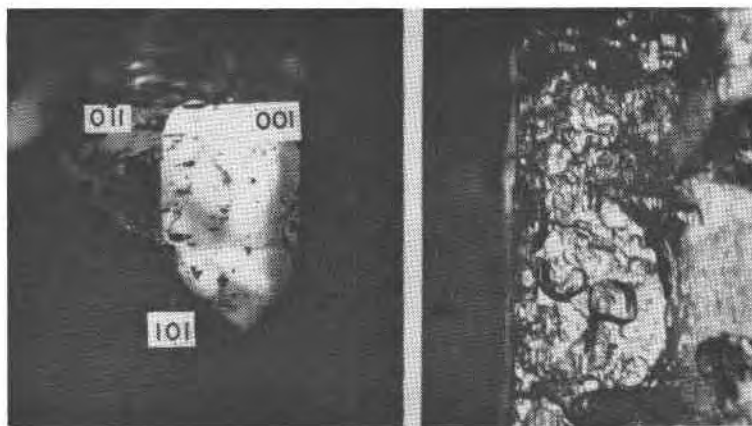


FIG. 1. A (left). New crystal faces developed on andalusite seed sealed with kyanite powder and water for 75 days at 700°C . and 3 kbars, $\sim 40\times$. B (right). Solution pits developed on andalusite crystal sealed with sillimanite powder and water under identical conditions, $\sim 20\times$.

site-S was sealed with the same amounts of powdered sillimanite and water. The kyanite came from Graphiteville, North Carolina, and the sillimanite from Benson Mines, New York. Both were ground to 140–300 mesh. The two capsules were taken to 3 kb at 700°C . for 75 days and quenched. Andalusite-K gained 3.2% weight and showed signs of growth. Three new crystal faces were developed at one end of the crystal (Fig. 1A), and small prismatic spines had grown on the seed crystal in apparent optical continuity with it. The three newly developed faces were measured on a two-circle reflecting goniometer and gave the following interfacial angles (the indices assigned are those for andalusite): $001 \wedge 101 = 35^\circ 36' \pm 15'$ and $001 \wedge 011 = 34^\circ 56' \pm 15'$. The axial ratios for andalusite fall well within the range calculated from these measurements ($c/a = 0.7158 \pm .0066$ and $c/b = 0.6985 \pm .0065$ compared to $c/a = 0.7124$ and $c/b = 0.7025$ for andalusite as given in Dana (1932)). The corresponding ratios for AS(H)-II taken from the cell dimensions given by Aramaki

and Roy (1962) are $c/a=0.7494$ and $c/b=0.6842$. Corundum prisms (ca. $15\ \mu$ long) were also identified in the powdered charge. The incongruent solution of the Al_2SiO_5 polymorphs in water to form corundum has already been observed by Weill (1962).

In contrast, andalusite-S lost 1.2% weight, and showed abundant evidence of having been appreciably dissolved (Fig. 1B). Again, small corundum prisms had developed in the powdered charge. The weight loss of andalusite-S is most likely accounted for by the growth of a supersaturated (*i.e.*, more stable) phase in the $\text{Al}_2\text{SiO}_5\text{-H}_2\text{O}$ system. Under the conditions of the experiment this could only be sillimanite. It is not thought likely that the loss can be explained simply by the solution of andalusite since presumably the fine grained sillimanite of the charge dissolved much more rapidly than the relatively large crystal of andalusite. Unfortunately, no unambiguous evidence of sillimanite growth could be detected in the powdered charge.

SUMMARY

Evidence has been presented in support of the reaction kyanite \rightarrow andalusite at $700^\circ\ \text{C.}$ and 3 kb. The author believes this to be the first unambiguous demonstration of the laboratory synthesis of andalusite. Somewhat less satisfactory data suggest that the reaction andalusite \rightarrow sillimanite may also take place. The order of stability thus indicated for these polymorphs under these conditions is compatible with the phase diagram tentatively suggested by Miyashiro (1949) on the basis of geological evidence.

It is believed that the use of carefully weighed single crystals may be of great value in experimental studies of petrologically significant solid-solid phase equilibria when the latter are prohibitively slow for study with the usual hydrothermal techniques involving only optical and x -ray identification of powdered charges. The weight changes of such crystals is much more sensitive to slow reactions and their use may enable a study of such reactions to be made in experiments of reasonably short duration. For example, small weight changes of a crystal involved in a reaction may be used to indicate the direction of the reaction. Such an approach is valid, of course, only if it can be firmly established that the weight change can be attributed to the reaction under consideration.

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OSARIZAWAITE FROM WESTERN AUSTRALIA—A CORRECTION¹

R. C. MORRIS, *Government Chemical Laboratories,
Perth, Western Australia.*

In a recent paper (Morris, 1962) the writer gave values for the unit cell of osarizawaite, $\text{Pb}(\text{CuAl})_3(\text{SO}_4)_2(\text{OH})_6$, from Western Australia as $a = 7.05 \text{ \AA}$ and $c = 17.025 \text{ \AA}$.

Y. Taguchi (pers. comm.) has pointed out that the value for c is more probably 17.25 \AA and that d-spacings calculated from this figure are in better agreement with the observed values, and in addition the pseudo-isometric nature of osarizawaite is made more apparent.

In support of this statement he argues that in the series between beaverite, $\text{Pb}(\text{CuFe})_3(\text{SO}_4)_2(\text{OH})_6$, and osarizawaite, the increase in the aluminium content is accompanied by an expansion of the c axis, a contraction of the a axis, and a decrease in density and mean refractive index, a trend borne out by the values reported for the series jarosite-alunite.

In view of this, Taguchi believes that it is unlikely that the virtually pure aluminium end member from Western Australia could have a value for the c axis smaller than that of the iron-rich Japanese osarizawaite (17.23 \AA) when the other physical properties are consistent with the trend.

The writer agrees that Taguchi's value is the more probable one. The amended values for the unit cell of the West Australian osarizawaite, with original values in parentheses, are, $a = 7.05 \text{ \AA}$ (7.05); $c = 17.25 \text{ \AA}$ (17.025); $c/a = 2.447$ (2.415); $a_{\text{rh}} = 7.045 \text{ \AA}$ (6.984); $\alpha = 60^\circ 3'$ ($60^\circ 38'$); calculated density 4.114 (4.167).

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