# On the existence of monoclinic $NaAlSi_3O_8$ at elevated temperatures

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Summary. Measurements from powder diffraction patterns recorded at temperatures up to  $950^{\circ}$  C for the supposed equilibrium forms of albite synthesized at  $800^{\circ}$ ,  $900^{\circ}$ , and  $1000^{\circ}$  C by MacKenzie (1957) indicate that only the form synthesized at  $1000^{\circ}$  C becomes monoclinic. Single-crystal measurements on heated pure natural albite ( $Ab_{99\cdot26}$ ) show that it becomes truly monoclinic, both in geometry and intensity distribution, at  $930^{\circ}$  C, the same temperature as for the highest-temperature synthetic form. These experiments were carried out in conditions that exclude possible contamination by potassium.

THE evidence presented by Brown (1967) on potassium contamination of 'monalbite' suggests the necessity for a re-examination of high-albite to see whether pure NaAlSi<sub>3</sub>O<sub>8</sub> can become monoclinic before melting or not. Data available in the literature and new data presented here, both carried out under conditions that exclude potassium contamination, show convincingly that the highest temperature form of albite does become monoclinic before melting. A revised definition of high-albite is given below.

After X-raying a number of synthetic plagioclases and alkali feldspars at elevated temperatures, MacKenzie (1952) found there was a discrepancy between the results obtained from plagioclase feldspars synthesized dry and those synthesized hydrothermally. Further investigation revealed that the temperature of synthesis and subsequent heat treatment were the main causes of these differences. Fig. 1 summarizes the results of some of the experiments on synthetic albite and some plagioclase compositions. The effect of dry heating at 1030° C and 1080° C on an albite crystallized at 800° C is to lower the extrapolated value of the temperature of its supposed triclinic-monoclinic inversion from 1180° C to 980° C, the temperature approaching that estimated for pure albite (viz 920±20° C) by extrapolation of the curve for the dry-crystallized plagioclases.

In the same paper (MacKenzie, 1952, p. 332) it was stated that there were slight differences in the X-ray powder patterns of hydrothermally

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crystallized plagioclase feldspars depending on the temperature of crystallization; this observation was subsequently more thoroughly investigated in the case of albite (MacKenzie, 1957) where it was found that in addition to the temperature of synthesis, time was also an



FIG. 1. Plot of the extrapolated temperature of the supposed monoclinic-triclinic transformation against composition for dry and hydrothermally synthesized albite-rich plagioclases.

important factor. As a result of this later work a number of supposed equilibrium forms of  $NaAlSi_3O_8$  intermediate between high and lowalbite were obtained and each of these related to a temperature of synthesis; in addition, a large number of non-equilibrium forms were synthesized. From this it is clear that the supposed equilibrium form of albite synthesized at say 900° C is not the highest temperature form of albite and presumably is slightly more ordered than the highest temperature form of albite.

A high-temperature X-ray powder diffraction study (using a diffractometer furnace with a Ni specimen holder) of the supposed equilibrium forms of NaAlSi<sub>3</sub>O<sub>8</sub> synthesized by MacKenzie (1957) has now been carried out at a series of temperatures up to 950° C and complete lattice parameters obtained (to be published). Data for the forms synthesized at 800°, 900°, and 1000° C (MacKenzie, 1957, table III, nos. 225, 329, and 307) are given in figs. 2 and 3. As can be seen from fig. 2 the albite synthesized at 1000° C becomes dimensionally monoclinic at a temperature of about  $930^{\circ}$  C. The albite synthesized at  $900^{\circ}$  C does not appear to become dimensionally monoclinic, and this is even clearer for albite synthesized at  $800^{\circ}$  C (fig. 3).



FIG. 2. Plot of  $\alpha^*$  and  $\gamma^*$  against temperature for synthetic and heat-treated natural albite.

Because the highest temperature attainable with the diffractometer furnace used was 950° C, a single-crystal precession study up to 1050° C has been made of heated pure natural albite (S404,  $Ab_{99\cdot26}$  from an analysis by Weibel in Brown, 1960). The material was sealed in a platinum tube and heated for 31 days at 1060° C. No signs of melting were observed. A fragment of this material was mounted without cement by sintering to a Pt/Pt-13 %Rh thermocouple for 30 minutes at  $1100^{\circ}$  C and then heated in a gas flame and photographed using ultrahigh-speed Polaroid film (exposure times 5–10 minutes). The results are shown in figs. 2 and 3, from which the great similarity to the albite



Fig. 3. Plot of  $\alpha^*$  against  $\gamma^*$  at a series of temperatures read from curves of the type in fig. 2. Same materials as in fig. 2 with, in addition, data for albite synthesized at 800° C, indicated by crosses  $\times$ . Other symbols are as in fig. 2.

synthesized at 1000° C is apparent. From fig. 2 it can be seen that the curves for  $\alpha^*$  and  $\gamma^*$  against temperature can be extrapolated to 90° at about 930° C. Precession photographs taken at two higher temperatures show that the crystal has indeed become dimensionally monoclinic. Pairs of reflections that differ markedly in intensity at room temperature become equal at about 930° C and remain so on photo-

graphs at higher temperatures (fig. 4). Thus not only is the crystal dimensionally monoclinic above 930° C, but the intensity distribution is also monoclinic. In addition, the crystal developed weak albite twin-



FIG. 4. Plot against temperature of intensity ratios hkl: hkl from precession photographs (hk0 reflections  $\times$ , 0kl reflections  $\bullet$ ) for a crystal of albite heated for 31 days at 1060° C.

ning during heating in the gas flame; no pericline twinning was seen on these short exposure photographs.

Thus for the heated natural albite, and by analogy for the albite synthesized at  $1000^{\circ}$  C, the evidence is conclusive that at temperatures above  $930^{\circ}$  C both materials become truly monoclinic.

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The term high-albite may have to be more rigorously defined in order to avoid confusion in the literature—we suggest restricting the usage to materials obtained by synthesizing albite at temperatures close to the melting point or by prolonged heating of natural or synthetic albites at temperatures close to the melting point. Materials synthesized at lower temperatures, even those having lattice parameters at room temperature identical to those of high-albite defined above, do not conform to this definition of high-albite: high-albite cannot be defined on the basis of its lattice parameters at room temperature alone (Grundy, in preparation).

#### References

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