

# X-RAY STUDIES OF THE ALTERATION OF SODA FELDSPAR<sup>1</sup>

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## ABSTRACT

Studies have been made of the alteration of pure albite single crystals and powders. No structural modification of the feldspar itself has been detected. No evidence has been obtained for any preferential orientation of an alteration product in relation to the initial feldspar. Under hydrothermal conditions at 280° C and 430° C, albite flakes and powders have been subjected to attack by 0.1 *N* HCl for periods ranging from a few hours to 52 days. The flakes changed mainly to boehmite, the powders to a variety of products, including a well-defined kaolinite. This difference of behavior is interpreted in terms of Correns' ideas on the weathering of silicates.

## INTRODUCTION

This paper reports exploratory work undertaken to elucidate the formation of clay minerals from feldspars. An attempt has been made to study the process at three stages of development, namely (i) the initial stage when the alteration of the feldspar commences, (ii) an intermediate stage, and (iii) the final stage when clays and/or other products are fully developed.

Feldspars are formed of three-dimensionally linked SiO<sub>4</sub> and AlO<sub>4</sub> tetrahedral groups. Clay minerals consist mainly of two-dimensionally linked SiO<sub>4</sub> and AlO<sub>4</sub> tetrahedra, together with octahedral groups containing Al and other cations. A considerable structural rearrangement is involved, therefore, in passing from feldspar to clay mineral and it is not obvious how the transformation takes place. We have therefore looked for evidence which may show how a feldspar alters in the initial stage of the transformation, and also for evidence of any crystallographic relation between an alteration product and the initial feldspar. Various x-ray diffraction techniques have been applied, involving both single crystals and powdered materials. The work began with a study of some naturally altered feldspars, but progressed towards laboratory-controlled alterations. Various physical and chemical environments have been used and the nature of the end products determined.

## EXAMINATION OF SOME NATURALLY ALTERED FELDSPARS

Eleven rock specimens, mainly granites, containing weathered feldspars from the surface and apparently unweathered feldspars from below the surface were first examined. They appeared to be well suited for the present investigation. Microscopic examination showed small surface cavities containing crumbly or

<sup>1</sup> Contribution no. 55-25 from the College of Mineral Industries.

clayey materials, commonly white in color. These were carefully hand picked and examined by x-ray powder methods. They revealed no obvious clay minerals and appeared to be essentially feldspar material broken down to small particle size.

Comparison of the surface feldspar with the interior feldspar by x-ray powder and single crystal methods gave disappointing results. The apparently fresh, interior feldspars seemed to be variable, probably even within single crystals, so that no reliance could be placed on comparisons between weathered surface material and fresh interior crystals. Since it was an essential condition for the present work that it should be possible to interpret the x-ray data unambiguously, attention was directed towards well-defined feldspars, the alteration of which could be followed under laboratory-controlled conditions.

### SINGLE-CRYSTAL TESTS FOR FELDSPAR ALTERATION

Soda feldspar (albite) was chosen as being the most suitable for preliminary experiments. It is more easily altered than potash feldspar and is more easily obtainable in good crystalline condition than lime feldspar. Experiments were carried out principally on two albites: (a) from Court House, Amelia Co., Virginia; and (b) a cleavelandite from Auburn, Maine. These gave identical x-ray powder diagrams in good agreement with the data of Goodyear and Duffin (1954), and from the lattice spacing-composition diagram<sup>1</sup> of Tuttle and Bowen (1950, fig. 5, p. 581) it appears that they are low-temperature forms with less than 3 percent of lime feldspar. Tuttle and Bowen state: "Low-temperature albite apparently cannot tolerate more than very small amounts of potash. Crystals which have formed side by side with potash feldspar usually contain only a few tenths of a percent of  $K_2O$ ." On the basis of this evidence, we consider that the materials used in the present experiments were pure or almost pure soda feldspar.

Under the microscope, the Amelia albite showed liquid inclusions and the cleavelandite a few solid inclusions and possibly a trace of muscovite. The specimens cleaved readily on (010) and cleavage flakes about  $6 \times 4 \times 0.5$  mm in size were easily obtained.

An x-ray examination of cleaved flakes was made before and after various treatments. It was expected that any marked extraction of alkali or aluminum ions by an alteration process would modify the relative intensities of the  $0k0$  reflections; a one-dimensional Fourier synthesis should then give an indication of the nature of the change taking place. Flakes were carefully mounted on goniometer arcs on a G.E. XRD3 Geiger counter diffractometer and the intensities of the  $0k0$  reflections were accurately measured. The experiments failed to show any changes in the *relative* intensities of the  $0k0$  reflections, even though these were practically destroyed by some of the treatments.

Additional experiments were carried out in which the  $0kl$  reflections of fresh and of partially altered albite crystals were recorded on Weissenberg photographs. No reflections became markedly weakened or diffused by the treatments applied.

The outcome of all these experiments is that no evidence was obtained for any

<sup>1</sup> In this diagram, the angular interval  $2\theta$  should be given as  $131-131\bar{1}$ .

systematic alteration of the feldspar lattice as a first step towards the breakdown of the mineral.

### ABSENCE OF ORIENTATION RELATIONSHIPS BETWEEN SODA FELDSPAR AND ITS ALTERATION PRODUCTS

The possibility was envisaged that alteration of a feldspar may proceed by chemical breakdown followed by an oriented development of the new products on the surface of the feldspar. Partially altered flakes were observed to develop a shell of altered material around a core of apparently unchanged feldspar. Glancing-angle photographs of slightly altered flakes were recorded with a flat plate camera and pinhole collimation. No positive evidence was obtained for an oriented development of an alteration product. However, as will be shown in the following section, it was only rarely that claylike products were formed in the experiments with albite flakes. We cannot therefore exclude the possibility that oriented growth may sometimes occur. It can only be stated that we have not so far detected any such effect.

### ALTERATION PRODUCTS FROM FELDSPARS TREATED HYDROTHERMALLY WITH 0.1 *N* HCl

Preliminary attempts to alter albite with acid and alkali treatments up to 100° C proved extremely slow. Hydrothermal treatments were therefore applied. Experiments with powdered albite and H<sub>2</sub>O + CO<sub>2</sub> in a steel bomb at around 400° C yielded a mica-like product, probably of biotite type, and a chlorite-like material, but the products were not well crystallized and there was contamination by reaction with the steel bomb.

All subsequent experiments were carried out with 20 cc gold-lined Morey bombs charged with 10 cc of 0.1 *N* HCl together with the specimen, either a (010) cleavage flake of albite weighing about 10 to 20 mg, or a similar amount of powdered material. In some experiments, the particle size was reduced to less than 5 microns. The bombs were maintained at about 280° C and saturation pressure, or at 420 to 435° C and about 10,000 psi for periods of 2 hours to 52 days. There was a large excess of HCl in these experiments; the initial pH was about 1 and the final pH about 1.2 after a run.

Table 1 summarizes the main experimental results. The product obtained from the flakes was most commonly boehmite, AlO(OH). Kaolinite was obtained in experiment 56 when <5 micron cleavelandite was kept at 285° C for 52 days. In experiment 40, finely powdered Amelia albite after 24 hours at 285° C yielded a rather doubtful kaolin-type mineral. Occasionally the product hydralsite was obtained; this has been described previously by Roy and Osborn (1952, 1954) and appears to have a composition approximating to 2Al<sub>2</sub>O<sub>3</sub> · 2SiO<sub>2</sub> · H<sub>2</sub>O. As these authors state, previous workers have probably obtained hydralsite, but have confused it with pyrophyllite. In addition, an unknown product *X* was obtained on several occasions. This yields a rather simple x-ray powder diagram of sharp lines (Table 2). The material has not been identified so far and may possibly be a new phase.

TABLE 1. — ALTERATION OF ALBITES BY HYDROTHERMAL TREATMENTS WITH 0.1 N HCl (A = ALBITE FROM AMELIA, VIRGINIA; C = CLEAVELANDITE FROM AUBURN, MAINE)

Expt. no.	Albite	Temp.	Time	Results
I. <i>Experiments with albite flakes</i>				
37	C	285°C	2½ days	Boehmite and residual albite.
28	A	285°	17 days	Boehmite, strong sharp x-ray diagram.
55	A	275°	52 days	Boehmite predominant, but diagram less sharp than no. 28. Component showing lines at 7.2, 3.6, 2.65, 1.57 Å may be a kaolin-type mineral.
56	C	285°	52 days	Flake disappeared completely; data for powder, see below.
22	A	420°	2 hours	Boehmite; weak unidentified x-ray lines which may be hydralsite; residual albite.
34	C	430°	1 day	Hydralsite, nearly pure; a few additional x-ray lines.
26	A	420°	2 days	Boehmite.
29	A	420°	13 days	Boehmite, strong sharp x-ray pattern.
II. <i>Experiments with powdered albites</i>				
40	A, <5μ	285°	1 day	Kaolin mineral, rather doubtful; unknown component, X.
55	A, <5μ	275°	52 days	Uncertain product resembling a disordered pyrophyllite.
56	C, <5μ	285°	52 days	Good kaolinite; small amount of boehmite; additional lines not identified including a sharp line at 12.5 Å. Also a glassy deposit giving no crystalline pattern.
30	A	435°	1 day	Boehmite and hydralsite in comparable proportions.
31	C	430°	2 days	Unknown X; a little boehmite.
38	A	435°	2½ days	Unknown X; boehmite; other lines not identified.

Boehmite,  $\text{AlO}(\text{OH})$ ; hydralsite,  $2\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot \text{H}_2\text{O}$  (Roy and Osborn, 1954); kaolinite,  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ; unknown product X, x-ray pattern given in Table 2.

## DISCUSSION OF THE HYDROTHERMAL RESULTS

The results obtained with the two kinds of albite from Amelia and from Maine are not quite consistent. Also, what is less surprising, the results obtained with flakes and powders are different. The present results may be compared with results obtained by Gruner (1944) who, in certain of his experiments, used conditions closely similar to those employed in the present work. Using a powdered soda feldspar containing 87 percent albite and 0.1 N HCl in gold-lined bombs, Gruner obtained after 17 days at 300° C pyrophyllite and some kaolinite and after 14 days at 400° C pyrophyllite and some unchanged feldspar. The pyrophyllite which he recorded may have been the hydralsite which we have observed. Boehmite was seldom observed by Gruner, but this may be attributed to his use of powdered material rather than flakes. It appears that in experi-

TABLE 2.—X-RAY POWDER DATA FOR UNKNOWN PRODUCT, *X*

d, A	I (est.)	d, A	I (est.)
5.95	vw	1.900	w
3.22	s	1.801	vw
3.06	vw	1.664	m
2.770	vs	1.595	m
2.350	m	1.450	vw
2.170	vw	1.425	vw
1.957	vs	1.380	wm

ments of this kind it is rather easy to obtain variable results, probably because all the factors influencing the reactions are not fully appreciated or fully under control.

With the data so far available, it appears that feldspar flakes are changed principally to boehmite, although if the experiment is continued sufficiently long (c.f. Expts. 55 and 56) the boehmite itself may change. Finely powdered materials tend to pass quickly through the boehmite stage, but the full course of the transformation is not yet clear. It appears that unknown *X*, hydralsite, and possibly other products may be intermediates before a clearly recognizable kaolinite is obtained.

A result not brought out in Table 1 is that throughout the experiments with flakes (with Expt. 56 as the only exception) the flakes retained their size and shape during the hydrothermal treatment. They became porous and chalky in appearance, and lost a considerable fraction of their weight. Except for the shortest treatments, the feldspars were completely altered; in cases of incomplete alteration, the inner core remained as unaltered feldspar so far as we could ascertain by x-ray tests (see section on single-crystal tests).

The alteration to boehmite is noteworthy. This mineral may not be the final reaction product but it appears to be a significant stage in the alteration. It is observed more clearly with flakes than with fine powders. The boehmite shows no detectable preferential orientation. It is probably not strictly valid to compare this result with the equilibrium studies of Ervin and Osborn (1951) on the system  $\text{Al}_2\text{O}_3\text{--H}_2\text{O}$ , according to which we would expect corundum or diaspore as the most likely product under the temperature-pressure conditions of our experiments. However, we do not have the simple  $\text{Al}_2\text{O}_3\text{--H}_2\text{O}$  system, since HCl,  $\text{SiO}_2$ , and  $\text{Na}_2\text{O}$  are also present, and in addition it is probable that equilibrium has not been reached in many of the present tests. However, the significant fact is that alumina rather than silica remains behind within the confines of the flake. This appears to rule out entirely any hypothesis which requires that the HCl shall attack the feldspar and form  $\text{AlCl}_3$  which then hydrolyzes to  $\text{Al}(\text{OH})_3$  and subsequently transforms to  $\text{AlO}(\text{OH})$ . In one experiment (no. 26 in Table 1), the flake lost 51 percent of its initial weight, and an approximate chemical analysis by Dr. R. C. Vanden Heuvel gave the following data:

	Before treatment	After treatment
$\text{Al}_2\text{O}_3$	19.5%	49%
$\text{SiO}_2$	68.7%	16%

This confirms the marked extraction of  $\text{SiO}_2$  from the flake by the acid treatment.

The different behavior of flakes and fine powders may be interpreted in terms of a mechanism similar to that considered by Correns (1940) and his co-workers. This has been summarized conveniently by Van Schuylenborgh and Sanger (1950). The outer layer of a weathering particle consists predominantly of the more slowly dissolving components. Dissolution of the components within a particle is then determined by their rates of diffusion through the surface layer. The thickness of the leached layer grows until there is an equilibrium between the rates of diffusion through it and its own rate of dissolution. Under the conditions of the present experiments, it is clear that silica dissolves more readily than alumina from the albite flakes, so that a leached layer deficient in silica, and a concentration of alumina within the flakes, are to be expected. We cannot, however, offer any explanation why boehmite rather than other forms of alumina is the product. In the case of fine powders, however, we may picture the bulk of the material as residing in the surface layer, so that no appreciable segregation of alumina takes place. The whole system then becomes reactive and the products approach equilibrium in considerably less time.

During the course of the experiments described here, Dr. G. W. Morey (1955) of the Geophysical Laboratory, Washington, gave an account (at a meeting of the Geophysical Union) of experiments on the decomposition of albite and microcline by a continuous flow of water at 350° C and 5000 psi for long periods. In the case of albite he recorded the formation of boehmite together with paragonite; analcite was formed at the exit tube from the bomb. The two latter products have been recognized in the present experiments.<sup>1</sup>

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<sup>1</sup> Since this was written, a detailed account of these experiments has been published; see Morey and Chen (1955).

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