

Zeitschrift: Schweizerische mineralogische und petrographische Mitteilungen = Bulletin suisse de minéralogie et pétrographie
Band: 47 (1967)
Heft: 1: Feldspäte

Artikel: On x-ray properties of "adularia", (K, Na) AlSi_3O_8
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DOI: <https://doi.org/10.5169/seals-36941>

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On X-ray Properties of "Adularia", (K, Na) AlSi_3O_8

By *R. Gubser* and *F. Laves* (Zürich)*)

With 5 figures in the text

Abstract

The lattice constants a^* (resp. d_{100}) and the triclinicities (Δ) of 105 specimens of "adularia" selected at random and chemically analysed with respect to their Na-content were measured by the powder method. 36 of these specimens were further investigated as to their b^* and c^* values. 6 specimens were chosen for Na-K exchange experiments. The effects of this exchange on the lattice constants were studied. Correlations between a^* , b^* , c^* , and Na-content are discussed with respect to the structural state of "adularia". The main result obtained is that all (alpine) adularias investigated have properties intermediate between those of sanidine and microcline. On an average these properties resemble those of microcline more than those of sanidine.

Zusammenfassung

Von 105 bezüglich Na-Gehalt analysierten „Adular“-Proben („zufällig“ gewählt) wurde a^* (bzw. Netzebenenabstand 100) und Δ (Triklinität) mit der Pulvermethode gemessen. Von diesen Proben wurden 36 ausgewählt, um zusätzlich b^* und c^* zu messen. Korrelationen zwischen a^* , b^* , c^* und Na-Gehalt werden im Hinblick auf „strukturelle“ Adular-Zustände diskutiert. Von diesen 36 Proben wurden 6 Proben ausgewählt, um den Effekt eines Na-K-Austausches auf die Gitterkonstanten a^* , b^* , c^* zu studieren. Der Vergleich mit entsprechenden Sanidin- und Mikroklin-Werten ergibt, dass alle untersuchten Adulare Eigenschaften haben, welche zwischen denen von Sanidin und Mikroklin liegen. Ferner ergab sich, dass im Durchschnitt die untersuchten (alpinen) Adulare Eigenschaften haben, welche eher denen des Mikroklin entsprechen als denen des Sanidin.

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INTRODUCTION

The term "adularia" is very commonly applied by mineralogists to certain types of feldspar, though a generally acceptable definition of the same is hard to find in the textbooks. Optical and morphological properties have to be taken into account as well as the manner of occurrence in nature. Thus most mineralogists, though having a fairly clear conception of what they mean by the word "adularia" would find it difficult to give a definition in precise terms. The only point sure of general acceptance is that "adularia" is a K-rich alkali feldspar.

It might be thought that X-rays would help to render the term more precise. Actually the reverse is the case and this for the following reasons: Whereas specimens of "adularia" frequently appear *optically* to be single crystals, X-ray photographs may show certain effects inconsistent with a *structurally* true monoclinic symmetry. Such effects may be:

a) Diffuse streaks starting from "monoclinic" spots and characteristic of "microclitic" domains oriented in twinned positions corresponding to the albite and pericline laws.

b) In addition, rather strong diffuse reflections may be present in positions that violate the extinction rules assumed to be characteristic for a truly monoclinic K-feldspar.

c) Specimens of adularia are encountered with regions which are optically not monoclinic but triclinic.

d) Single crystal X-ray photographs of these regions may show distinctly triclinic structural behaviour of a kind intermediate between that of a truly monoclinic sanidine and a triclinic maximum microcline.

The above observations a), b) and d) were first published and discussed by LAVES (1950), whereas the fact mentioned under c) was already known to MALLARD (1876). The properties of "adularia" are thus shown to be rather perplexing. There are indications that most adularias must be considered as "unstable" states intermediate between the "stable" states of the K-feldspars sanidine and microcline. This is held to be true both structurally and texturally as far as microscopical domain development is concerned. For a definition of "stable" and "unstable" states in relation to K-feldspars see the paper by LAVES (1960).

The existence of optically triclinic adularia was confirmed by CHAISSON (1950) and one of the present authors (LAVES 1950) correlated this "optically" triclinic behaviour with a structural counterpart. Furthermore, BAMBAUER and LAVES (1960) reported on the optical and struc-

tural behaviour of "domains" within a *single* morphological unit of adularia. Their results show that the "domains" exhibit a continuous variation from monoclinic to triclinic behaviour. In the one case the plane of the optic axes is \parallel (010), in the other nearly \perp (010). There is a continuous change of Ω (the angle between the optic axial plane and (010)) from 0° to approximately 90° . Concurrently $2V$ changes from approx $50^\circ \parallel$ (010) to approx 50° for positions about \perp (010). The minimum value of $2V$ is about 22° and occurs when Ω is near 45° .

The value $2V = 0$ was not observed during the series of measurements. The most triclinic parts showed a Δ -value¹⁾ of about 0.25. Such behaviour is not to be expected in material formed under conditions of equilibrium. Rather must it be attributed to states formed under non-equilibrium conditions as defined by LAVES (1960).

In order to obtain further information regarding the structural state of adularia, measurements of a^* , b^* , c^* and Δ ¹⁾ were made on 105 samples of natural adularia. As the Na-content of the natural material may influence these magnitudes, the amount of Na present was determined in all specimens investigated by means of flame-photometry²⁾.

II. EXPERIMENTAL

105 samples taken from 57 specimens of adularia from the Alps (mainly from the Swiss Alps) were investigated in respect to their Na-content²⁾ and the values of a^* , $d_{(100)}$ and "triclinicity" (Δ)¹⁾ 36 of these samples were selected for treatment in the following way:

¹⁾ The value $\Delta = 12.5 (d_{131} - d_{\bar{1}\bar{3}1})$ was defined by GOLDSMITH and LAVES (1954) as a characteristic expression of the "triclinicity" of K-feldspars. Several authors objected to this term and now call it "obliquity". However, such a change in terms does not appear justified for the following reasons: "Obliquity" is a general term equally applicable to monoclinic and triclinic crystals. On the other hand, the expression "triclinicity" was coined specifically to indicate a triclinic geometry of K-feldspars. The word "triclinicity" should perhaps be reserved for the more particular concept of the *obliquity* of triclinic substances. Then, however, another and more specific term "microcliniticity" might be introduced but will not be used at this stage.

²⁾ We gratefully acknowledge our indebtedness to Prof. Dr. Max Weibel of this Institute for providing these flame-photometric analyses. The limit of error is estimated by him to be smaller than ± 0.03 weight % Na_2O . (Cp. WEIBEL, 1957, WEIBEL and MEYER, 1957.)

a) Short-time heating (about 5 hours at approx 1050°C). The purpose of the treatment was to homogenize any cryptoperthitic unmixing possibly present in the material.

b) Long-time heating (from about 200 h to as much as 3000 h in some cases at 1100°C) for "sanidization".

c) Short-time heating (2 to 4 hours at 1050°C in KCl melts). This was done to produce Na-free phases without changing the Al/Si distribution. That this can be done was shown by LAVES (1951) and by GOLDSMITH and LAVES (1961), using glasses of alkali-feldspar composition as an exchanging medium. In these experiments "feldspar-glasses" were used to avoid any influence of flux additions. HAFNER and LAVES (1957) produced such exchanges by heating finely powdered microcline and albite. WYART and SABATIER (1956) and ORVILLE (1963) showed that alkali halides can also be used as an exchanging medium without any observable influence on the Al/Si distribution.

In the present instance powders with a grain size of between ~ 0.5 and $5\ \mu$ were used. The powders were prepared by centrifugation of aqueous suspensions. For the heat treatment the proportion of powder to KCl was about 1 : 500.

d) Samples (a), (b), and (c) were used to determine the d-spacings of 400, 060, and 002. This was done with a Guinier-type camera (after Jagodzinski) using $\text{CuK}_{\alpha 1}$ -radiation ($\lambda = 1.54051\ \text{\AA}$). Spinel ($a = 8.0831_2\ \text{\AA}$) was used for calibrating the photographs. The lattice constant of the spinel was determined by calibration with silicon ($a = 5.4305_4\ \text{\AA}$). All the photographs were taken with the same camera and with the same amount of powder (8 mg feldspar plus 2 mg spinel). The position of the lines was measured with an accuracy superior to $\pm 0.01\ \text{mm}$. Hence the errors in d_{100} , d_{010} , and d_{001} (or a^* , b^* , c^*) are smaller than $\pm 0.1^{\circ}/_{00}$, $0.1^{\circ}/_{00}$ and $0.2^{\circ}/_{00}$.

In addition to the samples (a), (b), and (c) mentioned above some pegmatitic and authigenic microclines were submitted to similar treatment and examination for purposes of comparison.

III. RESULTS

A. Figure 1 shows the triclinicity (Δ), Na-content and a^* of the 105 samples mentioned in the first paragraph of section II. This figure was previously published by LAVES and GOLDSMITH (1961) but without the additional data indicated by arrows. The diagram is reproduced here

for the purpose of comparison with the new results obtained in the present investigation. It is noteworthy that the Na-poor feldspars showed a more triclinic behaviour than the Na-rich ones and that the latter show a greater scattering in the plot than the Na-poor members.

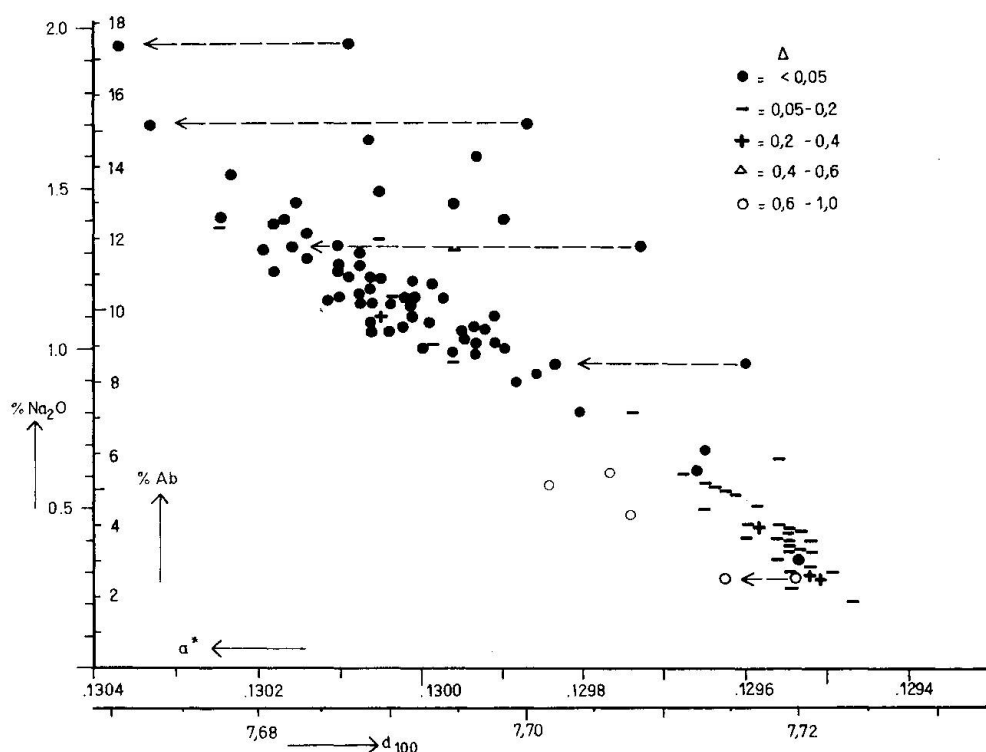


Fig. 1. $d_{(100)}$ (a^*) against Na-content of random specimens of adularia from the Alps (mostly of Switzerland). Specimens indicated by arrows were given short-time heat treatment for homogenization (see text).

B. As it was suspected that the scattering in question might be caused by varying degrees of cryptoperthitic unmixing in the different samples, the heat treatments described in section IIa were applied. Figure 1 shows some results obtained in extreme cases. It thus appears that after the described short-time heat treatment a rather good correlation between a^* and Na-content emerges.

C. The empty circles in figure 2a depict the a^*/Na correlations of all the 36 samples that were given short-time heat treatment as described — section IIa. In addition, the filled circles in Figure 2a show the results of the long-time heat treatment described in section IIb. The filled squares refer to the values obtained on maximum microclines that had been submitted to short-time heat treatment. The figure demonstrates

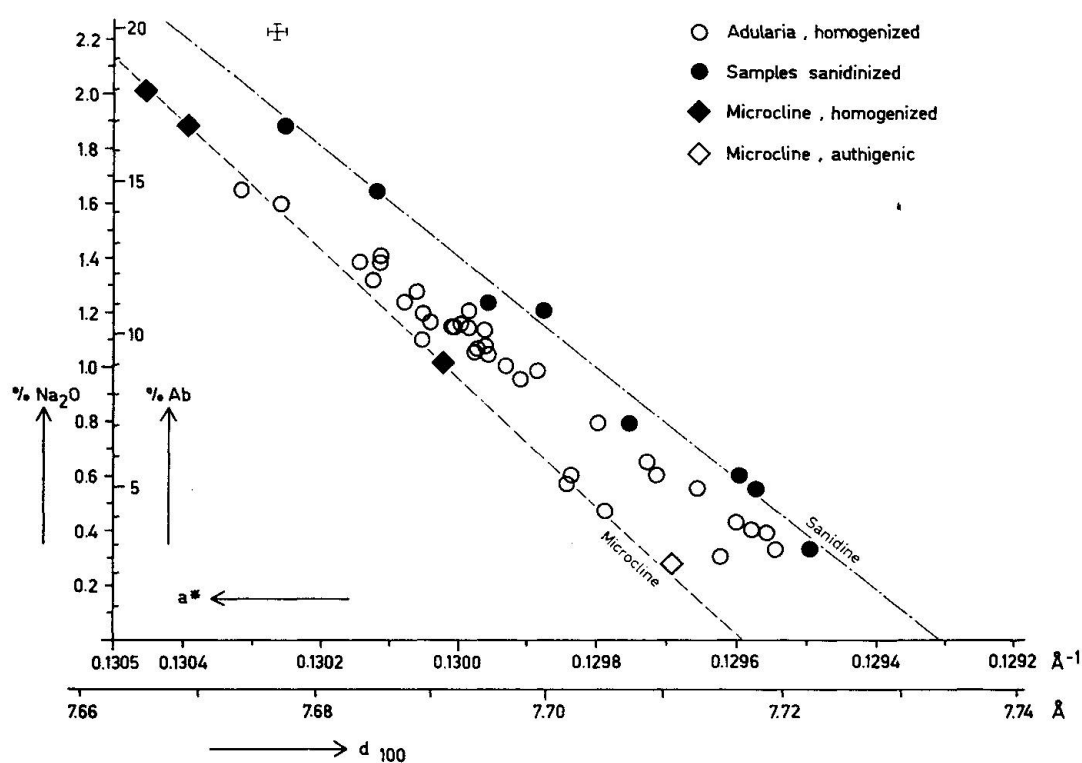


Fig. 2a. a^* against Na-content of alkali feldspars. Limits of error are indicated by the cross.

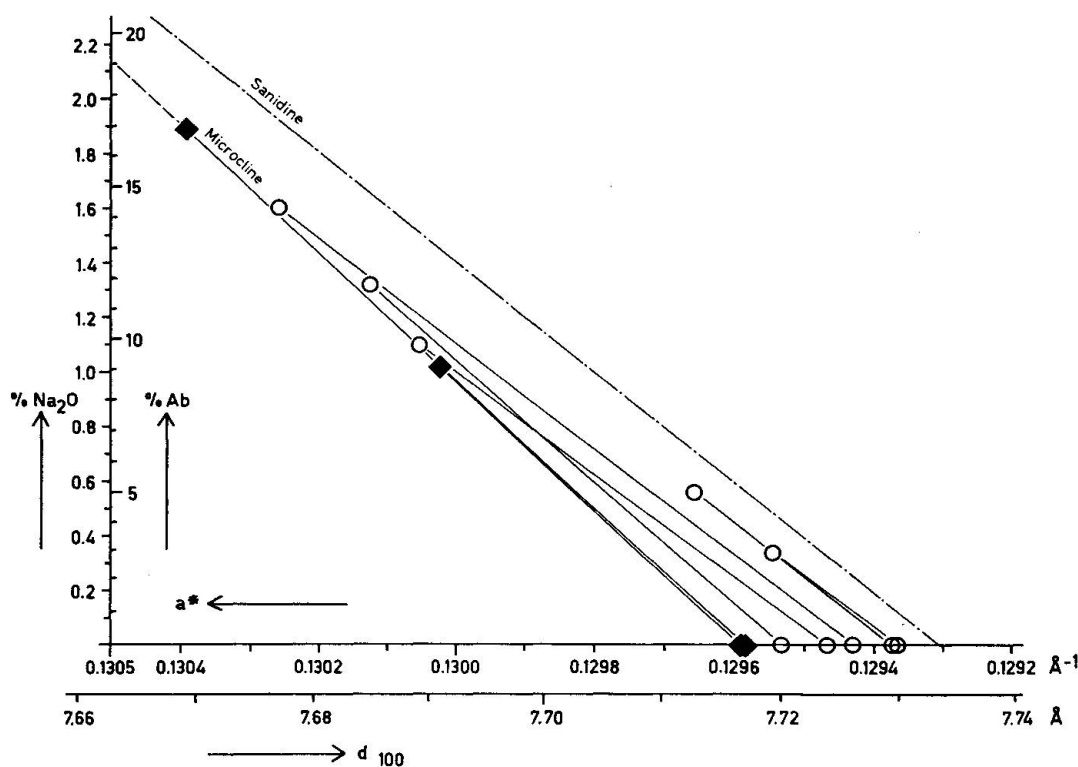


Fig. 2b. a^* against Na-content of selected specimens before and after heating in KCl (corresponding values are connected by lines). See Fig. 2a for the meaning of the symbols and the limits of error.

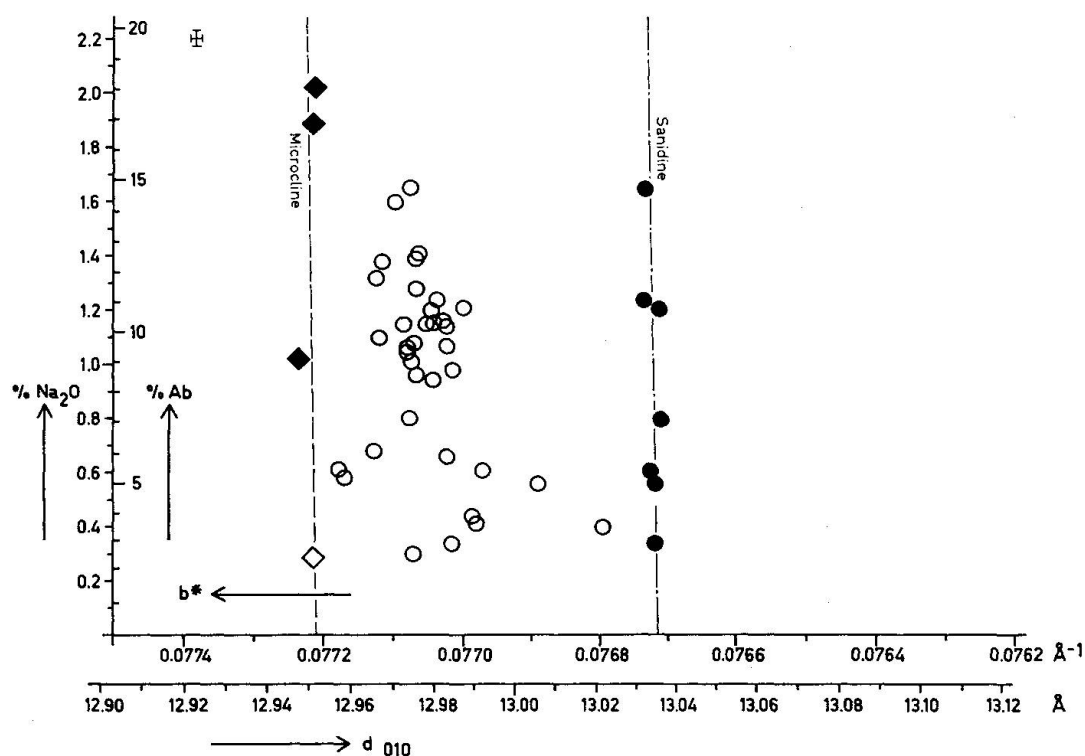


Fig. 3a. b^* against Na-content of alkali feldspars. Limits of error are indicated by the cross. See Fig. 2a for the meaning of the symbols.

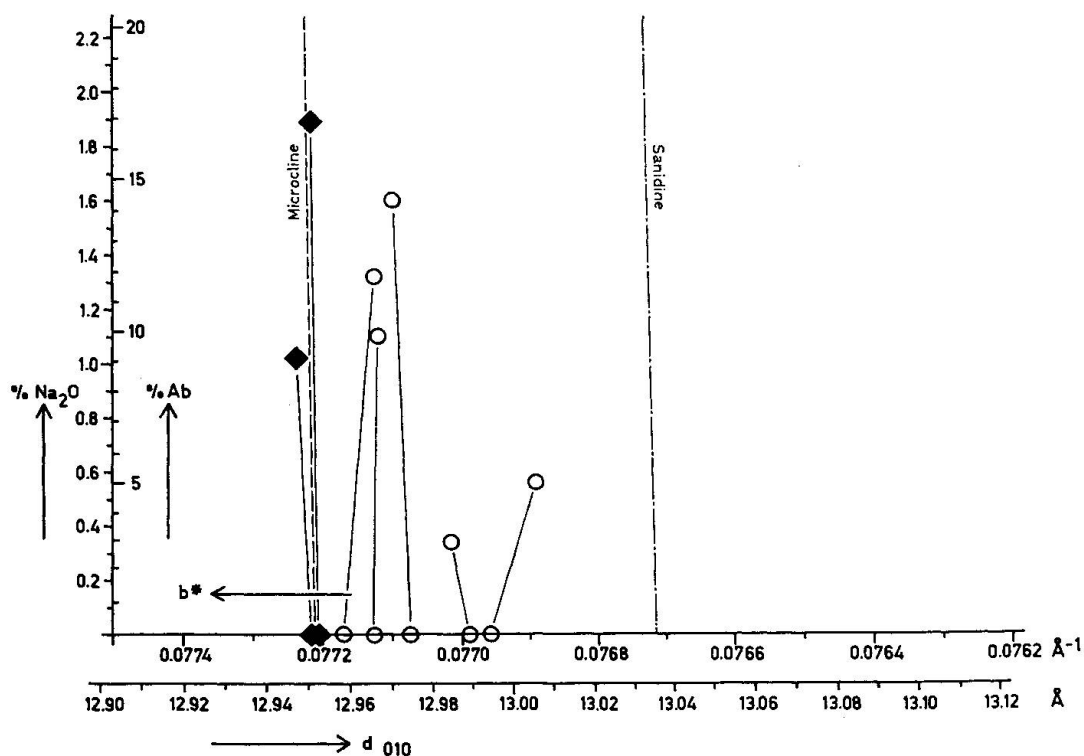


Fig. 3b. b^* against Na-content of selected specimens before and after heating in KCl (corresponding values are connected by lines). See Fig. 2a for the meaning of the symbols and Fig. 3a for the limits of error.

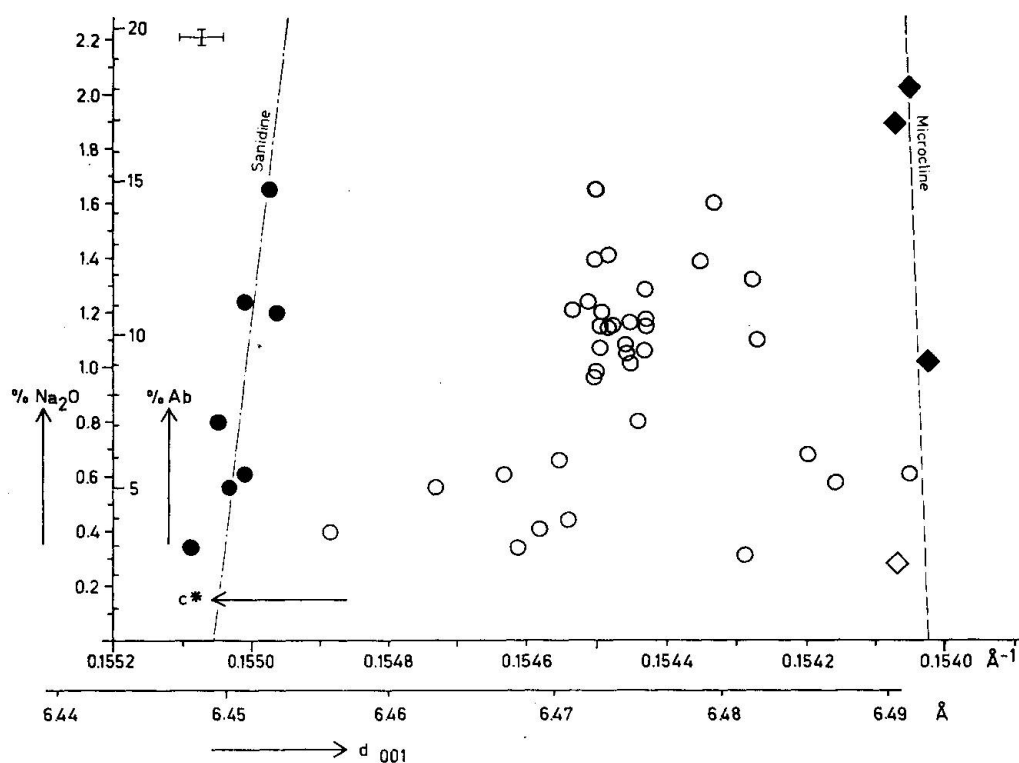


Fig. 4a. c^* against Na-content of alkali feldspars. Limits of error are indicated by the cross. See Fig. 2a for the meaning of the symbols.

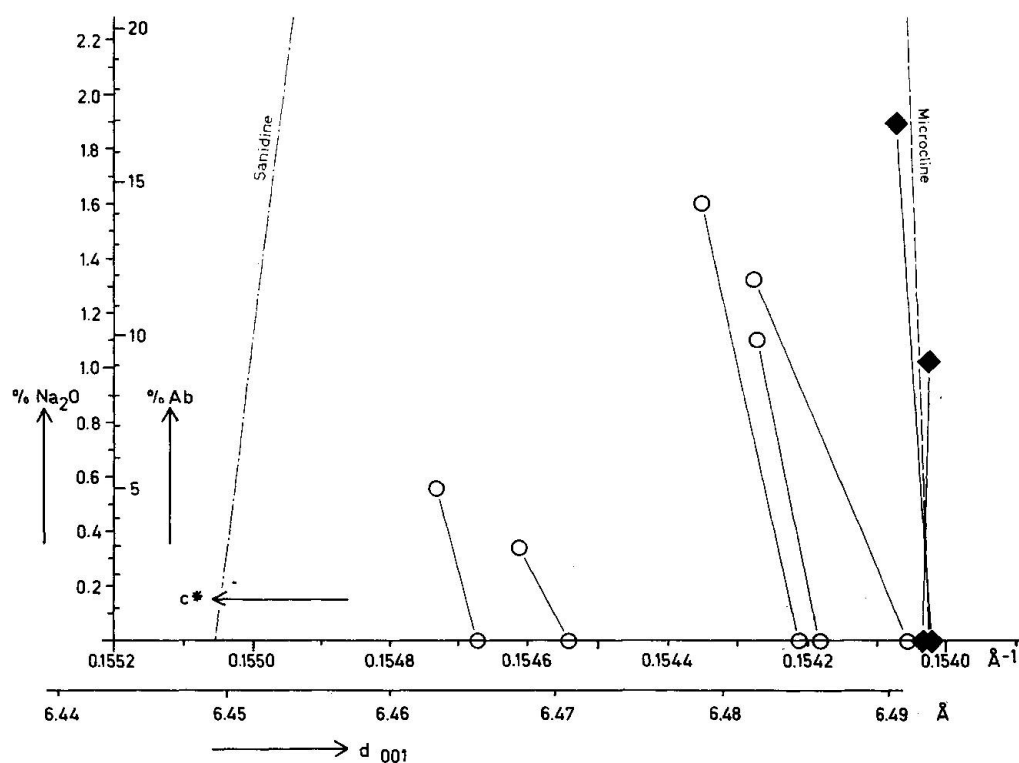


Fig. 4b. b^* against Na-content of selected specimens before and after heating in KCl. (Corresponding values are connected by lines.) See Fig. 2a for the meaning of the symbols and Fig. 4a for the limits of error.

that the adularia values fall within a field limited by two lines of which one is characteristic for maximum microcline and the other for sanidine.

The samples on which Figure 2a is based were also used to determine their b^* and c^* values. This was effected by measuring the 20 of 060 and 002. The correlations of b^*/Na and c^*/Na are plotted in Figures 3a and 4a. Again the adularia values occupy fields that are limited by lines characteristic of maximum microcline and sanidine.

Extrapolation of the limiting lines of Figures 2a, 3a, and 4a to a zero Na-content leads to the a^* , b^* , c^* values given in table 1 which are considered to be characteristic of Na-free maximum microcline and sanidine.

D. It is interesting to note that the a^* values of *Na-poor* adularias approach the values corresponding to sanidine. The b^* and c^* values, on the other hand, appear to have an opposite tendency ("microclitic" behaviour).

E. In order to check the values in table 1 obtained by extrapolation of the values of Figures 2a, 3a, and 4a, as well as to obtain further information concerning the adularia state, Na-free material was prepared as described in section IIb. The results are plotted in Figures 2b, 3b, and 4b. The slope of several lines in these figures is somewhat puzzling but shall not be discussed at this stage. Nevertheless, the data obtained in these experiments again indicate that the adularias investigated have properties intermediate between microcline and sanidine.

IV. DISCUSSION

As our knowledge of the structural states of adularia is still limited (cf. Part I), an investigation of the mineral's b^* and c^* values appeared to be of interest, because these values are rather sensitive to the structural state of alkali feldspars and in particular to the Al/Si-order/disorder relations. (See Table 1; microcline is rather ordered, sanidine rather disordered.) For this reason 105 analysed adularia specimens were selected

Table 1. d values of Na-free *K*-feldspars in their most disordered and their most ordered state

	a^*	b^*	c^*	d_{100}	d_{010}	d_{001}
Sanidine	0.12931	0.076715	0.15506	7.7336	13.0353	6.4491
Microcline	0.12959	0.077208	0.15402	7.7161	12.9520	6.4925

for measurement of their a^* and Δ^1 values and 36 of these were used for the determination of a^* , b^* , c^* by means of the powder method as well of the triclinicity (Δ). The values of a^* , b^* , c^* were plotted against the Na-content (Figs. 1, 2a, 3a, 4a). The main results are as follows:

1. There is a good correlation a^*/Na as was to be expected.
2. There are virtually no correlations b^*/Na or c^*/Na but rather a considerable scattering of the b^* and c^* values with respect to the Na-content. (Figs. 3a and 4a.) However, all values of b^* and c^* lie between such values that are characteristic of maximum sanidine and maximum microcline. On an average the adularia-values resemble those of microcline more than those of sanidine. This average result also becomes apparent when the b^*/c^* relations are plotted as in Fig. 5 which incidentally shows a rather good correlation between the b^* and c^* values.
3. In view of the known limits of error, the data of Fig. 5 do not, however, indicate the existence of an exact one-to-one correspondence between the b^* and c^* values. As the b^*/c^* relation appears to be rather

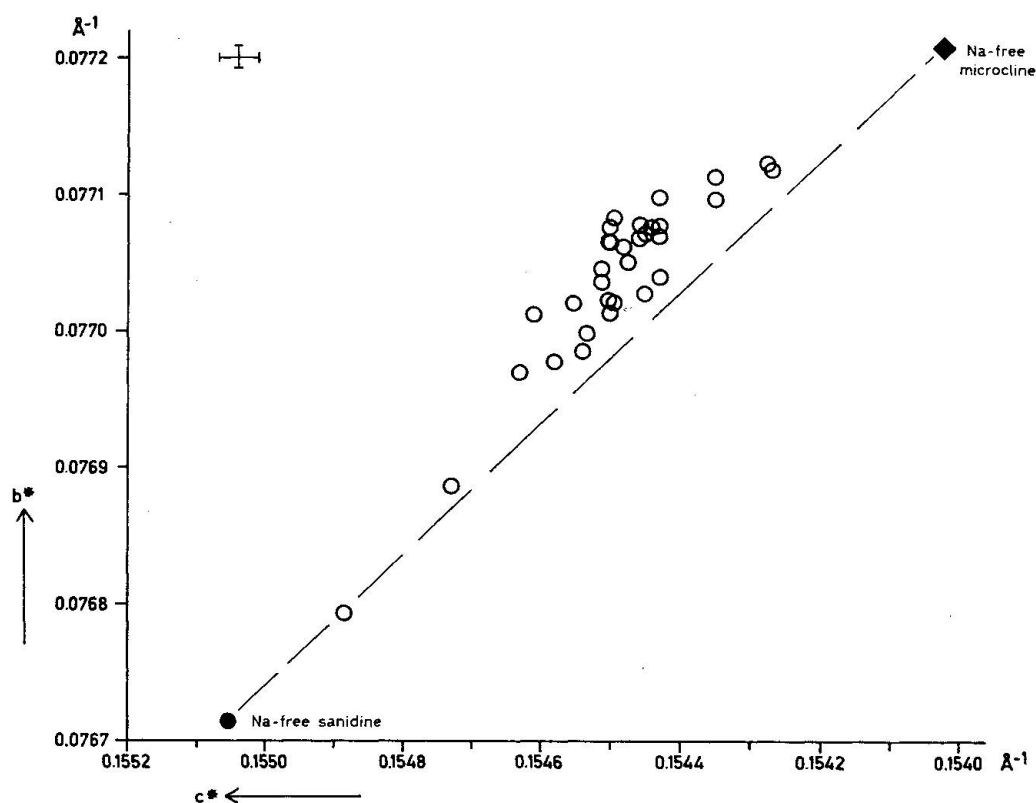


Fig. 5. b^*/c^* -relations of adularias with Δ -values < 0.2 (circles). The limit of error is indicated by the cross. The black dots and squares correspond to the values of Table 1.

sensitive to the degree of Al/Si-order/disorder, it may be concluded that the scattering of the points in Fig. 5 is due to the development of "unstable" states. These could either be unstable in the sense of "unstable" Al/Si-distribution structures or of submicroscopically "twinned" (balanced or unbalanced) domain textures or finally in both respects.

4. As most of the material used for this investigation had Δ -values smaller than 0.2, no correlations between lattice constants and triclinicity can be discussed at present. On the other hand, the b^*/c^* correlations as depicted in Fig. 5 provide further support for the suggestion that apparently monoclinic adularia *may* have a rather high degree of Al/Si order.

5. Some samples were "desodified" by the treatment described above (II, c). The results of this treatment on the lattice constants are shown in Figures 2b, 3b and 4b. It is interesting to note that this treatment changed the lattice constants but did *not* affect Δ within our limits of error. (We thank Prof. P. Ribbe for the suggestion to check on this point.)

6. The optical data on adularia correlated to X-ray data published by BAMBAUER and LAVES (1960) indicate that adularia *may* exist in a rather disordered state (analogous to sanidine). The present investigations provide further support for these conclusions. Adularia may grow in states more disordered than those corresponding to equilibrium conditions. During geological time an approach would be made to the equilibrium state of maximum microcline. The paths along which this approach (and correspondingly the Al/Si distribution) is effected may vary somewhat according to circumstances. Only very few adularias ever reached the equilibrium state of maximum microcline. Most adularias found in nature have only partially reached a state of equilibrium and are actually in states not to be considered as stable in the sense of LAVES (1960). Considering the infinite number of three-fold Al/Si distributions possible in triclinic feldspars and in view of the fact that the texture of "submicroscopically twinned" domains may have an important influence on physical (e.g. optical) properties, no surprise can be felt about the difficulty of formulating a clear-cut definition of "adularia". "Adularia" is a "mineral" but need not necessarily be a "stable" phase in a thermodynamical sense. From a structural and textural point of view, it may be a "sanidine", a "microcline", or an "orthoclase", or anything intermediate between these states that are (in principle) capable of being rather rigourously defined. In this connection reference should be made to an interesting paper by ANSILEWSKI (1958) on "adu-

laria" hydrothermally formed in gneisses. A further discussion of the matter is given in an accompanying paper by LAVES and VISWANATHAN (1967).

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