

SOME INVESTIGATIONS IN THE CRYSTALCHEMISTRY OF SILICATES

II. The Orientation of Perthite Lamellae in Feldspars.

BY

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Abstract. The perthite problem has been dealt with. The treatment is based on the diffusibility in feldspar minerals. The values have previously been published by the author. It is concluded that the most stable orientation of perthite lamellae results from layer formation normal to the longest axis of the diffusion ellipsoid of the mineral, provided the diffusion body is a positive revolution ellipsoid. In the feldspars at temperatures from about 450° K to about 700° K this seems to be approximately the case. The accurate position of the longest axis is not stated with certainty, but it seems to be situated in the vicinity of the normal to $(\bar{8}01)$. This position is in accordance with the perthite plane frequently found, viz. $(\bar{8}01)$ to (100) .

The perthites of the feldspar group early attracted the interest of mineralogists. In Norway especially Olaf Andersen has worked in this field. In his paper from 1929 he gives an extensive review of the perthite problem. He states that it is possible to discern between three modes of formation of perthites:

1. Exsolution
2. Rhythmic crystallisation
3. Replacement.

Without regard to the genesis he distinguishes between the following types of perthites:

- a) *String perthites.* The lamellae rich in sodium form very small, rather regular strings in the microcline. The strings lie parallel to (010) and at an angle of about $— 73^\circ$ with (001) . The cross-section of the strings is often oblong with the major axis perpendicular to (010) .
- b) *Film perthites.* The lamellae rich in sodium form thin films in the microcline, running perpendicular to (010) and at an angle of around $— 73^\circ$ with (001) . More seldom other directions of the films are found.

- c) *Vein perthites*. This is the most abundant type. The lamellae rich in sodium are variable with regard to size, outline and orientation. Most commonly they are running in directions approaching the (100) face, at angles around $— 65^\circ$ with (001).
- d) *Patch perthite*. This type is still more irregular than the vein type. The parts rich in sodium show a tendency to orient themselves in the same direction as the vein perthites.

The other perthite classes described by Andersen are of minor importance in this case, and they are not dealt with here.

Andersen finds it reasonable to assume that especially in the case of string and film perthites, exsolution processes in the solid state have played an important part. In the other cases it seems as if replacement has been of the greatest importance. In the case of exsolution processes Andersen seems to be of the opinion that a total migration of albite molecules take place. Edmonson Spencer, who has discussed the perthite problem (1937), states that it is sufficient that some of the sodium ions should diffuse into certain parts of the potash sodium feldspar. In these parts the sodium is replacing the potassium ions. This replacement is followed by a contraction of the SiO_4 tetrahedra and a slight turning of their axes, whereby a tiny albite lamella is formed.

This means that even the exsolution perthites may be regarded as a kind of replacement perthite. In the case of the ordinary replacement perthites it is obvious that the actual replacement process is a diffusion process, although the transport of sodium and potassium ions to and from the place of replacement may be caused by circulating solutions. Olaf Andersen (loc. cit.) is discussing the orientation of the perthite lamellae and he is concluding that contraction cracks play an important part in the orientation process. In a broad chapter he is discussing the orientation of the tension during contraction and cooling. Even though contraction cracks may orient themselves in such a way that they can give an explanation of the orientation of the lamellae in some perthites, they fail in the explanation of why perthite lamellae are very seldom found in other directions than approximately $\mp (\bar{8}01)$. It must be remembered that this direction is just one of the possible directions of contraction cracks.

During my work on the diffusibility of cations in crystalline silicates, I have found some features which might give a possible

answer to some of the questions in the problem of orientation and shape of the perthite lamellae. The paper referred to (Rosenqvist 1949) gives the details of the diffusion experiments. Here I will therefore just briefly repeat the results of interest in the case of perthite formation.

In the study of diffusion anisotropy in feldspars it was found that cations as Ra^{++} showed a marked difference in diffusibility in different crystallographic directions of the feldspar minerals. The diffusion in the following three directions were measured: $\perp \{010\}$, $\perp \{001\}$, and \mp the a-axis. The diffusion co-efficients proved both in microcline and in albite to be much higher in the latter direction than in the two other directions. At 1000° K the ratio between the three diffusion co-efficients is found to be 1 : 1.5 : 5.9 in the case of diffusion of Ra^{++} in microcline. In albite the corresponding ratio is 1 : 1.75 : 9.5. The actual values in the 3 directions are at 1000° K in albite $\cong 4.8 \cdot 10^{\pm 10}$ cm²/sec \perp (001); $\cong 8.5 \cdot 10^{\pm 10}$ cm²/sec \perp (010) and $\cong 4.5 \cdot 10^{\pm 9}$ cm²/sec \mp [Ok].

In microperthite the corresponding values are $3.2 \cdot 10^{\pm 9}$ cm²/sec., $2.2 \cdot 10^{\pm 9}$ cm²/sec, $1.3 \cdot 10^{\pm 8}$ cm²/sec. The accuracy may be about ± 25 % except for the value \perp (001) in albite where the accuracy is about ± 50 %.

The diffusion co-efficients depend on the temperature in such a way that the ratio mentioned is decreasing by decreasing temperature. At about 650° — 700° K it seems as if the ratio between the diffusion \perp (010) and \perp (001) approaches unity, and at temperatures of about 450° K the three diffusion coefficients seem to be nearly equal.

The diffusion anisotropy in crystals of lower symmetry is a fact that has not always been recognized. It seems to me, however, that it is of major importance in the case of perthite formation and orientation.

From classical wave theories we have to assume that the locus of diffusing ions, starting from a point in the middle of a crystal after a certain time t represents an ellipsoid. In tetragonal, trigonal and hexagonal crystals this diffusion ellipsoid is degenerating into a revolution ellipsoid, and in cubic crystals it is degenerating into a sphere.

It is not possible, however, without further assumptions mathematically to derive neither the axial ratio nor the orientation of the diffusion ellipsoid in a triclinic crystal only from the three data measured, viz.; the diffusion co-efficients $\perp (001)$ $\perp (010)$ and $\neq a$ -axis.

If we regard the values measured in the feldspars, it is obvious that the anisotropy in three directions practically perpendicular to each other is very pronounced.

We may therefore conclude that the unknown diffusion ellipsoid must possess a pronounced excentricity in one direction. In the two other directions, however, the excentricity probably is much less. We do find that the diffusibility $\perp (001)$ and $\perp (010)$ is nearly the same.

The more the ellipsoid approaches an ellipsoid of revolution the greater is the probability that we should find an axis system of the above mentioned type, that is with two axes perpendicular to each other and nearly the same size, and a third axis normal to the plane of the two others and considerably longer than these.

As an approximation we therefore may assume that the frontier of cations diffusing in feldspar minerals is shaped nearly like a revolution ellipsoid. This diffusion ellipsoid has a high positive excentricity.

From this approximation we can conclude that the axis of quasi rotation lies in or near the face (010) and forms only a small angle with the normal to $(\bar{8}01)$.

To make a check on the position of the diffusion ellipsoid I measured the diffusion co-efficient in the direction $\cong \div 18^\circ$ from the normal to $(\bar{8}01)$. This direction is conjugated to the a axis. Within the probable error I got the same value as $\neq [0kl]$.

Several additional measurements of the diffusion co-efficients in different directions of the same feldspar crystal are necessary to ascertain the position and axial ratio of the diffusion ellipsoid.

The data measured, however, tell us enough to draw several conclusions of interest in the study of perthites. Provided the activity is uniform in all crystallographic directions, the majority of the diffusing cations is diffusing to and from in one direction close to the normal to $(\bar{8}01)$. In all other directions the number of diffusing cations per cm^2 and sec. is less. It seems probable to me that sodium and potassium behave in much the same manner in sodium-potassium-feldspars as Ra and Pb behave in microcline and albite. I have, how-

ever, never had the opportunity to study the diffusion of alkali metals. Because of the low half life value of the radioactive isotopes it will be a difficult task to undertake the necessary experiments.

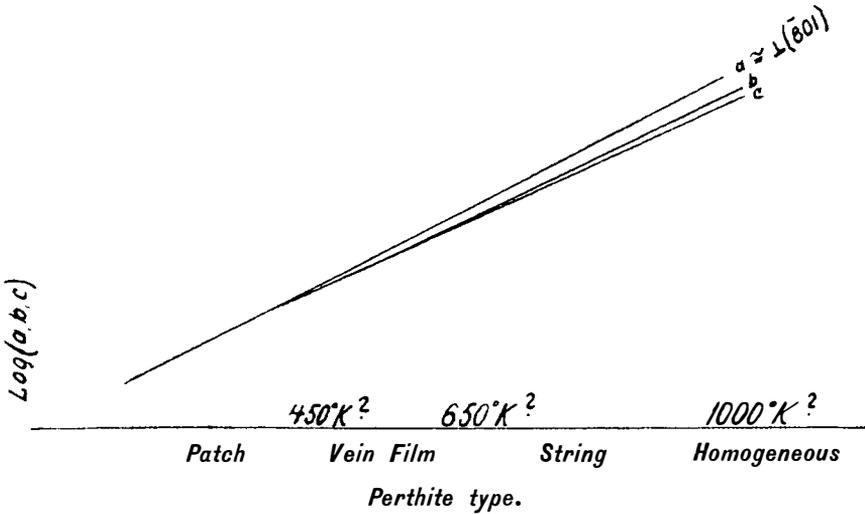
Returning to the perthite problem, we know that at temperatures below the boundary line of the immiscibility gap of the alkali feldspars, the activity of sodium in homogenous mixed crystals is higher than the activity of sodium in albite. Exsolution therefore takes place.

We may assume that the first albite lamellae are distributed haphazardly. These albite lamellae will be in diffusion equilibrium with the mother crystal at constant temperature. In a certain time the same number of sodium ions are precipitating on the lamellae and dissolving from the same lamellae. When the temperature is decreasing, the sodium activity will, however, decrease more in the albite lamellae than in the mother crystal, and a continuous precipitation takes place. From the diffusion experiments, however, we know that the majority of the diffusing ions are diffusing in one single direction. This means that the lamellae normal to this direction will grow more rapidly than all other lamellae. If the cooling process is sufficiently slow, the lamellae in all directions except the one normal to the highest diffusibility will be resorbed and precipitated on the lamellae normal to the highest diffusibility. This is caused by the higher solubility of the smaller crystals. Provided the diffusion ellipsoid is a positive revolution ellipsoid, the stable exsolution perthite lamellae form layers normal to the axis of revolution. If the diffusion ellipsoid is an ordinary triaxial ellipsoid, and the three axes differ much from each other, the stable exsolution perthite lamellae will not form layers of equal thickness, but lenses or strings parallel to the shortest axis. If the diffusion ellipsoid degenerates into a sphere, however, no orientation or shape will be more favoured than any other. In this case it seems likely that the albite lamellae would be irregular and un-oriented.

The theories outlined above, concern the formation of exsolution perthites. Without important alterations the same theories may, however, be employed on replacement perthites.

It seems from the diffusion experiment as if the symmetry of the diffusion ellipsoid is decreasing by increasing temperature. According to these observations it must be concluded that perthites formed at high temperatures must be of the string type, at intermediate tem-

Axial ratio of diffusion ellipsoid



peratures the perthites must be of the film or vein type and at low temperatures the perthites must be irregular of the patch type.

The figure shows this idea in a simplified diagram. The diagram is based on the assumption that sodium is diffusing in microcline the same way as radium.

The diffusion experiments proved that diffusion in the solid state is a very slow process in silicates. It seems therefore of some interest to examine if it is possible to form exsolution perthites by diffusion in a reasonable time.

We may, for instance, consider a temperature of 600° K. At this temperature the diffusion co-efficient of Ra \perp (801) in microcline is $1.10 \div 10^{12}$ cm²/sec. As an approximation we may assume the same diffusion co-efficient in the case of sodium. We may further assume a homogeneous soda potash feldspar containing 25 % albite molecule corresponding to about 2 % Na. This feldspar is assumed kept at a temperature of 600° K. From the exsolution curve we may assume that about $\frac{1}{4}$ of the soda content of the soda potash feldspar is soluble in the microcline phase at this temperature, the rest ≈ 1.5 % must separate as albite. We may as an instance assume that the albite is forming lamellae of 0.2 mm thickness with an average distance of 0.8 mm. The average Na gradient during the exsolution process may thus be calculated to approximately 0.35 g/cm⁴. As the exsolution takes

place from two directions there must diffuse about 0.005 g Na through a crosssection of 1 cm² in order to form a perthite lamella of 0.2 mm thickness. With a diffusion co-efficient = $1.10 \cdot 10^{-12}$ cm²/sec. this means that about $1.4 \cdot 10^{10}$ sec. is necessary, i. e. 500 years

It seems thus not improbable that the diffusion in the solid state may form perthite lamellae of a thickness of 0.2 mm in a reasonable time even though no transport of importance over long distances takes place.

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L I T E R A T U R E

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