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Alkali Feldspar Crystallization under non Equilibrium Conditions from Pneumatolytic, Hydrothermal and Diagenetic Environments

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Abstract

Primary unstable or metastable phases will usually crystallize when feldspars grow under pneumatolytic, hydrothermal or diagenetic conditions, that is at rather low temperatures and pressures, in presence of water without fusion of the rock. By Al/Si diffusion a secondary phase is reached as the system tends towards the stable state. Ion mobility is of main importance for the advance of this process. In *plutonic* and *volcanic* rocks disordered phases will crystallize stably. They can be chilled or cool rapidly tending towards equilibrium along non-equilibrium paths. When cooling is very slow equilibrium paths may be followed (GOLDSMITH and LAVES 1954b). The relation of this process to time and pressure was studied by KARAMATA (1961). From *fluid phases* primary disordered feldspars crystallize metastably, as crystallization is rapid. Ordered phases are reached rather soon, due to the great ion mobility even though pressure is not too considerable in cracks and hollow druses (RAMBERG 1949). Intermediate phases may also subsist. From *hydrothermal* environment any degree of order can be expected. Crystallization speed is variable depending mainly on the concentration gradient and ion transport in the solution. Usually rapid crystallization produces disordered phases. Ordering after crystallization is sluggish. Intermediate phases are frequent. Under *diagenetic conditions* (authigenic feldspars) crystallization speed is very low and ion mobility is poor. Very slow crystallization can produce primary triclinic phases around a monoclinic nucleus. Intermediate phases are frequent. Ordering hardly progresses at all after crystallization.

Under any of these latter conditions none of the feldspar thermometers hitherto proposed can be applied with sufficient certainty.

Zusammenfassung

Unter pneumatolitischen, hydrothermalen und diagenetischen Verhältnissen kristallisieren meist instabile oder metastabile Feldspatphasen, bei verhältnis-

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mässig tiefer Temperatur, die unter dem Schmelzpunkt des Gesteins liegt, in Gegenwart von Wasser und — im Fall von Kluft- oder Drusenfeldspäten —, selbst im fluiden Zustand ohne Einwirkung eines starken Druckes. Diese Feldspatphasen erreichen durch einen diffusen Al/Si-Ordnungsprozess verschiedene intermediäre Zustände, die dem Gleichgewichtszustand zustreben. Die Ionenbeweglichkeit ist für den Fortschritt dieses Vorganges äusserst wichtig. Bei *hochthermalen* und *mittelthermalen Gesteinen* können ungeordnete Phasen im Gleichgewicht kristallisieren; bei langsamer Abkühlung können sie auch auf dem Wege zum Gleichgewicht in den stabilen Zustand übergehen. Bei schneller Abkühlung hingegen streben sie dem Gleichgewichtszustande zu, in dem sie mannigfaltige intermediäre, instabile Zustände durchlaufen (GOLDSMITH und LAVES 1954b). KARAMATA (1961) untersuchte den Zusammenhang dieses Ordnungsvorgangs mit dem tektonischen Druck und der Zeit bzw. Alter des Gesteins. Bei niedrigem Druck entstehen in Klüften und Drusen durch schnelle Kristallisation aus fluidem Zustand meist metastabile Phasen. Die geordneten Zustände werden in der Regel bald erreicht, aber es können auch intermediäre Phasen erhalten bleiben. Aus *hydrothermalen Umgebung* kann jeder Ordnungsgrad erwartet werden. Die Kristallisationsgeschwindigkeit ist sehr verschieden und hängt hauptsächlich von der Konzentrationsänderung und Ionenzufuhr der Lösung ab. Oft kristallisieren instabile oder metastabile Phasen. Der diffuse Al/Si-Ordnungsprozess schreitet nur langsam voran. Intermediäre Phasen sind häufig. Unter *diagenetischen Verhältnissen* (authigene Feldspäte) kristallisieren die Feldspäte äusserst langsam; die Ionenbeweglichkeit ist beschränkt. Bei sehr niedriger Kristallisationsgeschwindigkeit können primäre geordnete Phasen entstehen, die wahrscheinlich um einen monoklinen Keim wachsen. Intermediäre Phasen sind häufig. Der Ordnungsprozess geht im festen Zustand sehr langsam vor sich.

Die bis jetzt vorgeschlagenen Feldspatthermometer können unter den hier untersuchten Bedingungen nicht angewandt werden.

INTRODUCTION

The properties, structural state and composition of feldspars have always been related to their crystallization or stability temperatures in an attempt to find a geological feldspar-thermometer of general applicability. This type of thermometer however seems to apply only in a very restricted number of cases, and usually not under the low temperature conditions here discussed. Feldspars will crystallize or even reprecipitate metastably under these conditions (rather low temperature and pressure in presence of water). High-temperature modifications may be chilled and later transform sluggishly into the stable form, often following non-equilibrium paths. To avoid erroneous conclusions it must therefore be born in mind that crystallization and stability temperatures may be widely different, higher or lower. Also that the final state can depend to a greater extent on the speed with which the system tends to

equilibrium, than on either of these temperatures. Usually not even the lower limit of the temperature of crystallization can be determined from the highest persisting form, as suggested by MACKENZIE and SMITH (1961) and other authors. Recently geological feldspar-thermometers have been proposed and discussed from different viewpoints by several authors: BARTH (1959, 1962), ORVILLE (1962b), CHRISTIE (1962), HEIER (1961), MACKENZIE and SMITH (1961) etc. A severe criticism, mainly of BARTH's feldspar-thermometers was published by DIETRICH (1961) and also by WINKLER (1961).

We believe that under pneumatolytic, hydrothermal and diagenetic conditions none of the feldspar-thermometers hitherto proposed can be applied with sufficient certainty, as the main condition for their correct application — that the feldspar phases shall be in equilibrium — is usually not fulfilled.

Referring to feldspar thermometry based on the structural state of the alkali feldspars the paper of MACKENZIE and SMITH (1961) is mainly discussed, as in this publication previous work is also considered. These authors establish a stability range of the different feldspar phases arranged according to their Al/Si order (for definitions see the original text). (See also BARTH, 1965.)

In potassium feldspars the degree of order was determined by them by admitting a direct relation between their optic axial angle and the degree of Al/Si order. Difficulties in relating this degree of order with the $2V$ values have already been discussed (M. DE SÁENZ 1966). Other aspects of MACKENZIE and SMITH's (1961) paper were discussed by LAVES (1961) at the end of that publication.

MACKENZIE and SMITH (1961) further discuss the relation between the crystallization temperature of alkali feldspars with their present structural state. To some extent they also take into account the crystallization speed. Thus they consider that among authigenic feldspars adularia might have grown quicker than maximum microcline which may be primarily triclinic. However, they maintain the alternative possibility — that intermediate authigenic feldspars crystallized at higher temperatures than maximum microcline. Proceeding from this second consideration they establish the stability limit of maximum microcline at 200°C. Similarly a limit of 300°C was given by BARTH (1959). The importance of the different speeds with which stable states are approached has already been pointed out by LAVES (1961). (Comment to MACKENZIE and SMITH 1961 § E.) This fact has certainly been neglected by many authors. Under pneumatolytic, hydrothermal and diagenetic

conditions it is certainly a main factor in determining the present state of the feldspar phase. The evidence put forward to establish the stability range of maximum microcline does not seem convincing. To determine such a limit it would be necessary to prove that under diagenetic conditions feldspars crystallize stably or transform into the stable state, or nearly so. If this is denied the only admissible alternative is that intermediate authigenic phases had not sufficient time to reach equilibrium and would otherwise probably have transformed into maximum microcline. If equilibrium crystallization of authigenic feldspars is admitted as a general rule, the occurrence of authigenic microclitic sanidine (M. DE SÁENZ 1963) and authigenic analbite (BASKIN 1956) cannot be explained, as both are near to high temperature modifications. Also other authigenic monoclinic or intermediate feldspars than those described by BASKIN (1956) and GOLDSMITH and LAVES (1954b) should not occur. In fact, it seems to happen very frequently that metastable and unstable phases crystallize at low temperatures, equilibrium crystallization being very rare. Under diagenetic conditions different factors, mainly ion — supply towards the growing feldspar crystal can determine whether crystal growth near to equilibrium conditions is possible. Authigenic feldspars seem to maintain their original degree of Al/Si order nearly unchanged and complete ordering apparently never occurs as to our knowledge crosshatched authigenic maximum microclines have not been described. Under hydrothermal and pneumatolytic conditions secondary ordering is the fundamental process determining the actual degree of Al/Si order of the feldspar phase. This process depends on temperature and pressure as well as on the presence of volatiles, especially water, and mainly on time. As conditions vary considerably with time after the beginning of feldspar crystallization extremely complex sequences can sometimes be found.

CHARACTERISTICS OF CRYSTAL GROWTH UNDER PNEUMATOLYTIC, HYDROTHERMAL AND DIAGENETIC CONDITIONS

General considerations on mineral crystallization under these conditions are given by NIGGLI (1948) and RAMBERG (1949, 1952).

Pneumatolytic conditions in the case of crack or druse-feldspars imply the presence of water above the critical point, temperatures below the melting point of the rock and rather low pressures. Crystallization from such a fluid phase, which is in fact a solid-gas solution, is fast. Ion mobility is considerable so that Al/Si diffusion as well as Na/K exchange can occur

rapidly. (Compare WYART and SABATIER 1961, ORVILLE 1962a etc.) The low viscosity of the solution permits the growth of large, well developed, idiomorphic crystals. Substitution processes penetrate deep into the rock or large crystals, also recrystallization seems to take place easily. Usually disordered crystals will grow as the crystallization speed will not allow an initial ordered distribution of the Al and Si ions, even though ion mobility is great. Potash feldspars and probably also albite nucleate in the monoclinic forms (BROWN 1962). Primarily potash feldspars usually grow as monoclinic individua as can be proved by their crosshatched twinning (LAVES 1950) and monoclinic morphology. If pneumatolytic conditions are maintained during some time ordering can advance quickly. (However it should be remembered that under the same conditions ordered feldspars may also undergo disordering by re-precipitation from a non-saturated solution [GOLDSMITH and LAVES 1954a].) Al/Si ordering always progresses more easily in $\text{NaAlSi}_3\text{O}_8$ than in KAlSi_3O_8 ; albite is nearly always found in its low temperature form in low temperature rocks, but KAlSi_3O_8 occurs frequently in intermediate states. This will occur specially when crystals form at the lower temperature limit of the pneumatolytic stage and the system rapidly cools down below the critical temperature. Adularia or common orthoclase (for definitions see LAVES 1960) will then be found. Recrystallization is also favoured by pneumatolytic conditions. Crosshatched twinning which is first submicroscopic will tend to become coarser. Sometimes rather large single crystal areas will be found. The regular crosshatching can even disappear and microclines twinned only after the albite or pericline law with irregular contact faces will be found. Such twinning can, therefore, not be taken as a proof of primary triclinic crystallization. Compare for example the samples described by BARTH (1959). The microcline described by this author may primarily have been monoclinic and have suffered an intense recrystallization later. Although KARAMATA (1961) proved that recrystallization speed increases considerably with pressure the influence of this factor will only be of minor importance as in our case high pressure conditions are not considered.

Certainly all the secondary processes discussed above can be interrupted by very rapid cooling either at their very beginning or after some time. High temperature phases or intermediate ones, from pneumatolytic environments can, therefore, also be expected.

The factors that determine the relation between crystallization temperature and the degree of order of the alkali feldspar phases are so complex and diffusive processes are so intense in the fluid phase that a

geological thermometer based on the composition or the structural state of the feldspar phases from pneumatolytic environments can not be expected to give any reliable results.

Under *hydrothermal conditions*, that is in presence of hot water below the critical temperature Na/K diffusion is rather active in the feldspar solid phase as well as between the feldspar phases and the hydrous solution. It is however limited to the contact zones and the influence of the solution mainly affects cracks and fractures or the outer rims of the rock, penetrating into the solid phase only to a minor extent. Zoning due to different Na/K relation is frequent. Recrystallization also occurs mainly near the crystal surface; crosshatched microclines mantled with single microcline crystals are found. The progress of crystallization under these conditions mainly depends on the concentration gradient in the solution, which in turn varies with the temperature gradient, with changes in pressure affecting evaporation, and with variations in composition due to ion contributions from outside the system. (On transfer of matter through rocks compare RAMBERG 1949 and 1952, p. 174—211.) The progress of crystallization also depends on the bulk composition of the solution, its viscosity and its temperature, all of which factors regulate the ion mobility in the solution. They decide the compromise between crystallization speed and ion mobility, thus regulating the possibility of approaching equilibrium. Usually primary disordered crystals seem to deposit from hydrothermal environment. If, however, crystallization is very slow, primary triclinic feldspars might be produced. EULER and HELLNER (1961) succeeded in crystallizing a highly disordered triclinic potassium feldspar in the laboratory under hydrothermal conditions. As in nature crystallization can occasionally occur at much lower speeds, somewhat more ordered feldspars may also be expected. This however does not seem to occur frequently. Ion transport towards the crystals is often rather irregular and "staircase faces" develop, e. g. (hk0) faces with different h and k values one of which is repeated several times (see M. DE SÁENZ 1967, fig. 5). Under these conditions ordering seems to follow non-equilibrium paths and adularia with a rather high degree of order and triclinic symmetry is often found. (CHAISSON 1950, BAMBAUER and LAVES 1960 etc.)

From *diagenetic environment* feldspar crystallization is very slow, compared with crystallization from the fluid phase or under hydrothermal conditions. Although ion mobility is very poor, the chance of primary triclinic alkali feldspars growing, seems to be better, although nucleation according to GOLDSMITH and LAVES (1954) and BROWN (1962) probably always occurs in the monoclinic state. BASKIN (1956) describes authi-

genic maximum microclines with triclinic morphology and a characteristic twinning probably due to the growth of primary triclinic feldspar on a monoclinic nucleus. Any degree of order can be expected to occur under these conditions the result depending mainly on the concentration gradient in the liquid solution. High-temperature phases have been found e.g. analbite by FÜCHTBAUER (1950), identified by BASKIN (1956) and microclitic sanidine by M. DE SÁENZ (1963).

Under diagenetic conditions ordering hardly seems to advance at all; the primary phases are preserved almost unchanged, as Al/Si ion diffusion is extremely slow in the solid phase at low temperature and pressure (compare KARAMATA 1961). This seems to explain why in BASKIN'S samples analbite could be preserved in spite of the ease with which ordering progresses in $\text{NaAlSi}_3\text{O}_8$. This is also the reason why to our knowledge crosshatched authigenic maximum microclines have not been described. Besides it does not seem to be reasonable to expect ordering to progress so far as to yield single crystals of maximum microcline when under hydrothermal conditions this does not occur either. Equilibrium crystallization is possible even for the classical examples of Ostwald's step rule, when crystallization speed is sufficiently slowed down.

COMPOSITION OF THE FELDSPAR PHASES

WINKLER (1961) in a discussion of the feldspar-thermometers has already stated that "... the composition of coexisting feldspars depends in a complicated manner on temperature, bulk composition and pressure and their composition can at present not give any indication of their temperature of crystallization". — We only wish to stress that the hydrous, low-temperature, and low-pressure conditions, referred to in the present paper, are probably the least favourable ones for the possible application of such geological thermometers.

Fluid phases and hydrous solutions are usually not in equilibrium with all existing solid phases, although small laboratory samples reach equilibrium rapidly (ORVILLE 1962a). The initial composition of these solid — liquid or solid — gas solutions is influenced by the composition of surrounding minerals. This is an account of leaching and ion exchange processes. It is also affected by the crystallization sequence of the paragenetic minerals. The composition of these solutions changes continuously with time, during and after crystallization. Migration of the solutions due to differences in specific gravity, temperature or pressure (ORVILLE

1962 a) and solubility of all pre-existing and paragenetic minerals, mainly that of the different feldspars, depending on their structural state and retrograde solubility (MOSEBACH 1955), and selective solubility also can produce considerable changes in the Al/Si lattice and especially substitution of one alkali feldspar by another, through alkali ion exchange. In pneumatolytic environments these substitutional processes proceed deeply into the rock; under hydrothermal conditions surface action of the solution is more frequent. The temperature and pressure mainly affect the intensity of the substitutional process, especially near the critical temperature (transition from the pneumatolytic to the hydrothermal stage) but the composition of the final phases, which is usually not uniform, depends rather on the relative Na/K ion concentration in the solution and in the solid phase. At similar temperatures albitization as well as microclinization can occur. These processes can be completed or intermediate compositions can be preserved due to partial substitution. Unsubstituted remnants are also frequent. We have hitherto studied one example of microclinization (M. DE SÁENZ 1965) and another of albitization (M. DE SÁENZ 1967), confirming these conclusions.

FINAL CONSIDERATIONS

At low temperatures, below the melting point of the rock and at low pressures, crystallization can occur from pneumatolytic, hydrothermal or diagenetic environments. In the *fluid phase* (pneumatolytic conditions) crystallization, ordering, ion exchange and recrystallization proceed rapidly. Primary disordered potassium feldspars crystallize metastably tending rapidly to equilibrium. Recrystallization can produce almost single crystals of microcline. Also sodium feldspars are often primarily monoclinic. If pneumatolytic conditions pass rapidly into lower temperature ones, any intermediate state of ordering or recrystallization can be expected. Under *hydrothermal* conditions, crystallization can be rapid depending on the concentration gradients. It has also been shown that under these conditions low temperature phases can be transformed into high temperature ones by dissolution and reprecipitation of the metastable phase (GOLDSMITH and LAVES 1954a) so that the degree of saturation of the hydrothermal solution has also to be considered. Usually primary disordered phases crystallize. Ordering progresses only sluggishly so that often intermediate phases are found. Intermediate primary phases may also form. The path along which ordering proceeds is usually

not the equilibrium one, so that "triclinic" or "monoclinic" adularias are frequent. Under *diagenetic* conditions crystallization is so slow that primary triclinic phases can also be expected. Anyway intermediate phases of all stages even high temperature ones will occur. Ordering hardly progresses at all after crystallization.

From all these observations it may be concluded that under the low temperature conditions described here, no geological thermometers based on the degree of Al/Si order of the alkali feldspar can be applied. Not even the lowest possible crystallization temperature can be estimated. Feldspar thermometers such as these proposed by BARTH (1959) etc. will probably also provide erroneous or very uncertain information about the temperature of crystallization. Many factors other than temperature combine in determining the final composition of the phases. Interpretation of twinning must also be very carefully considered as recrystallization will produce important changes in the distribution of the crystal individuals, and even single crystals may be produced from originally crosshatched microcline.

We believe that no general scheme relating the present phase characteristics of the feldspars discussed here with the crystallization conditions can as yet be provided.

Note: This paper contains the general conclusions of the thesis submitted for the degree of Dr. en Química of the University of Uruguay, Montevideo. The experimental results were discussed in previous papers (M. DE SÁENZ 1963, 1965, and 1967). Results published by other authors were also considered.

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