The heat treatment of some Norwegian aventurinized feldspars

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ABSTRACT. The morphology and structures of intergrowth precipitates in some Norwegian aventurinized plagioclases have been described in previous publications; data collected during an investigation of a wide variety of heat treatments upon the same specimens are now reported. In detail the behaviour of different feldspar crystals is highly individualistic, though there is a common pattern in sub-solidus reaction(s) with the feldspar; even if the identification of the products is uncertain, it is this process which causes the apparent 'homogenization' of the feldspar.

Some comparisons can be made between the present studies and the more limited experiments of earlier workers, and suggestions for reconciling experimental data are discussed. On the paragenesis of aventurinized feldspars, these heating experiments confirm that mechanism(s) are complex, but that exsolution from an originally iron-enriched feldspar by cooling in the solid state is unlikely to be of more than peripheral importance.

In previous papers (Copley and Gay, 1978, 1979) experimental data on the morphology and structures of intergrowth precipitates in some Norwegian aventurinized plagioclases were described and discussed. This contribution reports the effects of a variety of heat treatments on the same specimens, and considers their relevance to investigation of the aventurinization process in feldspars.

Summary of previous work. In his extensive study of aventurinized feldspars Andersen (1915) describes optical changes in the main hematite-like inclusions within a plagioclase (An₂₂) from Aamland, Söndeled, Norway as crystals were heated at temperatures from $1050\,^{\circ}\text{C}$ to $1260\,^{\circ}\text{C}$ for periods ranging from $\frac{1}{2}$ hr to 22 days. In detail the results are complex but his main conclusions can be stated briefly.

Without exception, outlines of host cleavage fragments were unchanged and crystals remained birefringent after heating; occasionally the feldspar became milky in appearance due to the formation of a fine black dust. The behaviour of lamellar

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inclusions depended on their original opacity; more opaque particles lightened rapidly in colour at about 1050 °C whilst other more transparent inclusions were unaffected at this stage. Reasonably uniform lighter-coloured lamellae then persisted until temperatures of 1230-60 °C were reached when they were no longer visible. Andersen attributed this disappearance to a simultaneous melting of the iron oxide inclusions and adjacent feldspar to form undetectable thin films of glassy material; he also suggested that the colour changes at lower temperatures were due to a phase change from a darker to a lighter form of the oxide. Annealing experiments at 1050 °C on previously 'homogenized' specimens caused reappearance of opaque lamellae in their original positions within the crystals after one or two days; however, he noted a change in their optical character so that they 'were evidently made up of a fine aggregate of an opaque or very dark brown substance'. Transparent particles did not re-appear, though their positions were sometimes occupied by a dense crowding of black dust (possibly a reduced oxide such as magnetite). From such experiments (and other observations) Andersen suggested that 'aventurine feldspars have been formed by unmixing of an originally homogeneous solid solution of the feldspar and hematite (or a ferric compound) in such a manner that thin hematite lamellae have separated along structural planes of the feldspar'.

Subsequent workers have criticized this hypothesis (see Smith, 1974), though only Kraeft and Saalfeld (1967) made any further observations on the effects of heat treatment on aventurinized oligoclases; their limited data were generally consistent with Andersen's work, though indicating that changes in the character of precipitates commenced at much lower temperatures.

Present experimental observations. Most of our heat-treatments were carried out on crystals from the aventurinized plagioclase (An₂₃) described as specimen 3 in Copley and Gay (1979); a few experiments with crystals from other specimens

described in that paper gave results in general accord with those from specimen 3 (which we judge to be comparable to that used by Andersen). Our original intention was both to extend and clarify Andersen's optical studies and to supplement them by X-ray and electron-optical investigation. To this end over 50 crystals with well-defined lamellar inclusions were subjected to a wide range of treatments in air at various temperatures for different periods. Detailed optical observations were made on all crystals, and many were also examined by X-ray diffraction methods; unfortunately it has so far proved impossible to prepare satisfactory specimens of heat-treated materials for electronoptical examination. Reference was made in our 1979 paper to difficulties encountered in the preparation of ion-beam thinned sections of aventurinized plagioclase; reactions in these specimens induced by heat treatment exacerbate these technical problems and electron microscope studies of reaction interfaces must await their solution.

Our optical observations confirm that the precise behaviour of individual crystals is unpredictable in that different temperatures and periods of heating are necessary to promote particular changes. Nevertheless a general pattern in the sequence of events has emerged, starting at temperatures in the range 800-850 °C. Initially the thinner yellowish more irregular platelets begin to 'disappear'; in general they are no longer visible at temperatures slightly above 900 °C. Most of the larger and more euhedral platelets are unaffected at this stage, but at higher temperatures these lighten in colour and eventually disappear; this 'homogenization' process is apparently promoted by extension of the heating period at a given temperature, and becomes much more rapid with increasing temperature. For

most crystals such 'homogenization' can be completed before the feldspar melts in the range 1050-1100 °C; a typical series of observations is summarized in Table I. Occasionally a feldspar crystal did not melt, and maintained its original shape up to about 1200 °C; although original lamellar inclusions were not visible, the crystal was less translucent with infrequent evidence of a dust of minute dark particles. Annealing experiments at temperatures in the range 800-975 °C for periods up to 100 days generated re-crystallized fine-grain polycrystalline aggregates from melted feldspar crystals; it was impossible to detect any re-precipitated nonfeldspathic material. Similar annealing treatments on unmelted feldspars showed that with one exception, changes caused by an original heating were irreversible; in particular, 'homogenized' specimens gave no sign of a re-appearance of iron oxide (or any other) precipitates. The exceptional experiment involved one of the relatively few unmelted specimens 'homogenized' at 1200 °C and subsequently annealed in stages at temperatures around 800 °C for a total period of 100 days. Additional twinning of the feldspar developed together with a few small colourless hexagonal plates on the crystal surface as well as traces of a reddish dendritic growth; it was impossible to identify these reaction products or to recognize any singular characteristic of this specimen which would account for its exceptional behaviour.

X-ray powder diffractometry shows only the host feldspar pattern at all stages, but standard range heavy exposure single crystal oscillation methods can provide some further evidence of the effects of heat-treatment. The onset of optical changes in the range 800-850 °C is associated with significant additions to the diffraction patterns. Fig. 1a shows a

	TABLE I. A	4 typica	l series o	f optical	observations	durina	heat treatment
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Temperature (°C)	Duration (h)	Total time (h)	Summary of observations
841	167	167	About 40% of the thinner anhedral platelets show signs of 'homogenization'; larger more euhedral darker platelets unaffected.
841	166	333	Continuation of process above; about 60% of anhedral platelets affected or apparently disappeared.
841	456	789	As above with about 80% of anhedral platelets 'homogenized'; euhedral platelets still apparently unchanged.
902	260	1049	Only euhedral platelets remain; these have begun to lighten in colour.
984	164	1215	Continuing 'homogenization' of euhedral platelets, most of which are now barely coloured.
1001	122	1335	Almost complete 'homogenization'; only a few residual fragments remaining.
1050	168	1503	Crystal melted.
977	580	2083	Re-crystallization of feldspar aggregate; no visible inclusions.

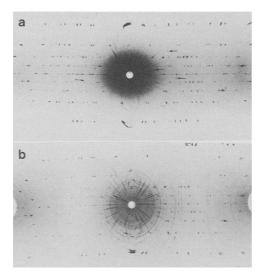


Fig. 1. Standard range oscillation photographs. Feldspar a-axis $Cu-K\alpha$ radiation. (a) Unheated aventurinized plagioclase; arrows indicate some reflexions due to inclusions. (b) The same crystal after heating for one week at $840^{\circ}C$.

typical pattern from an unheated crystal; apart from the main maxima due to the twinned feldspar host, a few additional maxima associated with the inclusions can be seen. Fig. 1b shows the pattern from the same crystal after heating for a week at 840 °C; if any original diffraction spots due to inclusions remain, they are swamped by a complex pattern of powder arcs superposed on the main feldspar maxima. This superposed powder pattern becomes more dominant with increasing optical 'homogenization' until the feldspar melts. From crystal to crystal, the d-spacings of powder arcs are constant (within limits of experimental accuracy); there are, however, variations in relative intensities and grain size which may be related to the temperature and duration of heating. For re-crystallized melted specimens, the complex feldspar powder pattern obscures any additional superposed powder arcs; annealing treatments of unmelted specimens showed no detectable change in the superposed patterns. There is no evidence of a preferred orientation texture at any stage.

We conclude that optical 'homogenization' involves an irreversible localized reaction at feldsparinclusion interfaces. In this reaction, the multicomponent products are similar in all specimens varying only in their relative amounts and grain sizes. At present the only data on these reaction products are the d-spacings listed in Table II; their accuracy is not very high even though values quoted are averaged from many measurements on

different oscillation photographs. After consideration of possible products from a reaction between plagioclase (An₂₃) and Fe₂O₃, it is our tentative conclusion that those most likely to form are an Fe-bearing feldspar accompanied by Fe-Al oxide(s). Whilst this does not conflict markedly with the data of Table II, it must be regarded as an unproven hypothesis until more reliable evidence can be obtained.

TABLE II. d-spacings of powder lines due to reaction products (Å)

4.9	3.25	2.55	2.10	1.74	1.43	
4.3	3.00	2.50	1.95	1.60	1.39	
4.15	2.85	2.40	1.87	1.57	1.32	
3.85	2.65	2.20	1.83	1.48	1.28	

The values quoted are averaged from measurements of the powder lines common to most heat-treated specimens. The accuracy is not high due to experimental circumstances, and values of d-spacings are unlikely to be more reliable than about $\pm 5\%$. Relative intensities are variable depending on the development of the reaction products.

Discussion. The present experiments demonstrate that the detailed behaviour of particular aventurinized feldspar crystals subjected to heat treatment is so erratic that strict correlation is not possible even from fragment to fragment of the same specimen, much less from specimen to specimen, locality to locality, etc. We are conscious that the results which have been described can only be regarded as a preliminary excursion into a complex problem which requires much further study. It is not our intention to pursue this problem; nevertheless this record of our data may be helpful to future workers in this field.

It is worthwhile to note that in their more general trends many of our observations are comparable to those of Andersen, although temperatures of about 200 °C lower produce similar optical changes in the hematite-like precipitates (note that Kraeft and Saalfeld mention similar temperatures for the few heat treated specimens that they examined). Whilst we can be certain of our furnace calibration but can have no reliable assessment of Andersen's temperatures, it seems most unlikely that so large a discrepancy arises from such a simple instrumental cause; its origin is more probably to be found in variability in sample chemistry and environment, differences in specimen size and heating duration and, or, some more complex factors not presently apparent. By their very nature aventurinized feldspars, even from the same bulk sample

from a particular locality, are chemically inhomogeneous; the general spread of our observations on specimen 3 (together with a similar pattern for the few crystals from other specimens that we have investigated) is compatible with such inhomogeneity. Rather more puzzling is the widespread melting of our specimens in the 1050-1100 °C range since Andersen reports heating experiments at temperatures about 150-200 °C higher (as do Kraeft and Saalfeld for one specimen). With such chemical variability and sample inhomogeneity, one cannot predict from published phase diagrams when the complex phase combinations that exist in aventurinized feldspars will melt; our experiments demonstrate the occurrence of reactions which may well lower melting points below those expected for plagioclase containing some K. Realizing that this raises the possibility of major experimental studies (particularly for systems containing iron oxides), we tried to reconcile 'anomalous' melting behaviour with the effects of crystal size and heating duration. In some simple experiments a number of fragments from 0.5-5.0 mm diameter were heated in air in the same crucible. After 160 h at 950 °C no crystal had melted, but further heating for 130 hrs at 995 °C caused smaller crystals to melt; further periods of heating at temperatures up to 1045 °C resulted in the melting of all but the largest fragments. Even such crude experiments indicate that larger crystals tend to melt at higher temperatures and the preservation of feldspar crystallinity in Andersen's experiments could be due to the use of large cleavage fragments (probably greater than 5 mm diameter) and shorter heat treatments (often only up to a few hours).

Whatever the origins of behavioural inconsistencies, our experiments indicate that all changes stem from reaction at the feldspar-inclusion interface. The d-spacings of Table II allow considerable ambiguity in product identification when using comparison with the known line spacings for likely components (particularly those of the complex Fe-Al oxide system). More positive identification and study of this reaction requires electron microscopy and diffraction at the interface; in practice we have found that the interfacial reaction weakens an already fragile attachment of the inclusions to the host feldspar so that they are removed by even the most modest ion-beam bombardment. Nevertheless we can be certain that the reaction products are at least partly crystalline and not completely glassy materials. Moreover, although laboratory annealing experiments cannot entirely preclude the development by exsolution of the original lamellae, the balance of evidence suggests that sufficient local concentrations of ferric ions would be unlikely to be available for the development of large hematite-like lamellae in aventurinized feldspar. Both our experiments (and those of Andersen) indicate that any exsolution of non-feldspathic material yields dust-like precipitates in the form of finely dispersed aggregates. Concerning colour changes in precipitate lamellae prior to melting, these must, in part, be due to a diminution in lamellar thickness, though as the reaction proceeds it is possible that the proposed redistribution of cations in inclusions and surrounding plagioclase (Copley and Gay, 1979) may also be responsible; a transition 'from a darker to a lighter modification of the hematite' proposed by Andersen is too simplistic a suggestion and is yet another area that needs further investigation.

The paragenesis of aventurinized (and clouded) feldspars has been reviewed by Smith (1974) in terms of two main hypotheses (a) exolution from an original Fe-rich feldspar in the solid state and (b) the introduction of Fe-rich inclusions into 'normal' feldspar from external sources. He finds the data both complex and conflicting; no firm conclusion can be reached and he recognizes the possibility that more than one mechanism can be involved. All that is clear from the present work is that heating experiments are unlikely to resolve these difficulties for they present new complications which, even if they were to be more fully investigated, are unlikely to be relevant to the conditions under which these Norwegian aventurinized feldspars were formed. From our examination of untreated and treated specimens we believe that whatever causes feldspar aventurinization in a particular locality, any iron enrichment of primary crystals plays only a minor part in the characteristic textures of the eventual intergrowth. In this context, the nature of the orientational spread of plate-like inclusions (see Fig. 2, Copley and Gay, 1979) could be regarded as consistent with the introduction of foreign material along planes of weakness (such as cleavage and composition planes) in normal plagioclase.

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