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The crystal morphology of plagioclase feldspar produced during isothermal supercooling and constant rate cooling experiments

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ABSTRACT. The crystal morphology of plagioclase feldspar in seven mafic rocks has been studied by isothermal supercooling and constant rate cooling experiments at 1 atmosphere using the 'wire-loop' suspension method. There is a systematic variation in crystal morphology with supercooling ($-\Delta T$), but the transitions between the different morphologies are gradual, and consecutive forms sometimes occur together. As $-\Delta T$ increases, the sequence of plagioclase morphology is generally laths → skeletal forms → fan spherulites. During isothermal supercooling the position of the plagioclase morphological fields is shown to be dependent on $-\Delta T$, superheating ($+\Delta T$), and also on time to a greater or lesser degree. Variations in melt composition and viscosity affect the position and slope of the boundaries of the fields. In the constant rate cooling experiments, the morphological type is dependent on the cooling rate and can be used to a limited extent as an indicator of the range of supercooling over which the first crystals nucleated and grew.

THE occurrence of non-equilibrium plagioclase crystal morphologies in volcanic, hypabyssal and plutonic rocks is widespread and has been attributed to supercooling, rapid cooling, and hence rapid growth (e.g. Wyllie and Drever, 1963; Lofgren, 1971*a*; Moore and Lockwood, 1973).

Several workers have produced a variety of plagioclase crystal morphologies (laths, skeletal forms, and fan spherulites) during isothermal supercooling experiments (e.g. Lofgren, 1971*b*; Fenn, 1973; Lofgren, 1974; Gibb, 1974; Lofgren and Donaldson, 1975*a*) and constant rate cooling experiments (Lofgren, 1973 and 1974; Lofgren and Donaldson, 1975*a*).

Throughout this study the crystal morphological terms of Lofgren (1974) will be used in the description of the plagioclase morphology.

Samples and experimental methods. The crystal morphology of plagioclase produced during isothermal supercooling of seven mafic rocks and constant rate cooling experiments of three of these seven has been investigated: a hawaiite (028) from Oldugigar 10 km west of Hekla, South Iceland; a basanite (331) from Cangreja, Tenerife in the Canary Islands; an ankaramite (336) from Montana Dornajo, Tenerife; a trachybasalt (339) from west of the Garachio Teno Peninsula, Tenerife, and two olivine tholeiites (G.1 and G.60) and a tholeiite (G.18) from dykes on the Island of Arran, Scotland. The chemical compositions of these rocks are given in Corrigan (1982).

All experiments were made at 1-atmosphere. In all the melts studied, plagioclase is the equilibrium silicate liquidus phase and the liquidus temperatures are given in Table I. The experimental apparatus and methods are described by Corrigan (1980, 1982).

For the *isothermal supercooling experiments*, after superheating (12-24 hours) the temperature was lowered at about 7 °C per minute to a pre-determined sub-liquidus isotherm, taking care to avoid undershoot, and held within 2 °C of this temperature for the desired run time before quenching into water at room temperature. During each run the gas mixture was adjusted to appropriate values for the new thermal conditions so that the oxygen fugacity remained constant. The time required for plagioclase nucleation over a *range of supercooling* ($-\Delta T$) was determined, and supercooling 'lines' could be drawn for each of the samples (Corrigan, 1982).

In the *constant rate cooling experiments*, during the initial superheating (12-24 hours) the gas mixture was set to give an f_{O_2} of 10^{-8} or 10^{-9} atmospheres. The charges were then cooled rapidly to 10-15 °C above the liquidus temperature, with an adjustment in the gas

TABLE I. *Liquidus temperatures of samples*

Sample no.	Liquidus (°C)	f_{O_2}
028	1166	10^{-8}
331	1168	10^{-8}
336	1210±3	10^{-8}
339	1176	10^{-8}
G.1	1258±5	10^{-9}
G.18	1166	10^{-8}
G.60	1237±6	10^{-9}

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mixture to give the same f_{O_2} at the liquidus temperature. The charges were then cooled at the pre-determined rates (0.3-11.8 °C/hr) through the liquidus temperature to various degrees of $-\Delta T$ and quenched. The $CO_2:H_2$ ratio in the atmosphere was not altered during these runs, so that the f_{O_2} decreased slightly with cooling. This procedure is justifiable since Hamilton and Anderson (1967) have shown that anhydrous basaltic magma cooling under conditions of constant total composition will have a small decrease in f_{O_2}, the amount depending on how much cooling has occurred.

Isothermal supercooling studies

Plagioclase morphologies. The variation of crystal morphology found in samples 028, 331, 336, 339, G.1, G.18 and G.60 with changing $-\Delta T$ conditions was similar to that observed by Lofgren (1974) in melts of the $CaAl_2Si_2O_8$-H_2O system and in remelted ocean ridge basalts, except that dendritic growth forms were not identified. Lofgren (1974) and Donaldson (1976) during their studies of the variation of plagioclase and olivine morphologies with $-\Delta T$ did not indicate what effect time would have on the form of crystals grown from supercooled melts. The present study indicates that the boundaries between plagioclase morphological fields in supercooled melts are dependent on time to a greater or lesser degree. Other factors such as superheating and melt composition could be important in determining the exact shape and position of these fields.

In a multicomponent basaltic melt, crystallization of liquidus plagioclase will enrich the residual melt in low temperature constituents and the $-\Delta T$ value will be decreased as isothermal crystallization progresses. Hence, in a given charge the stable morphology for the initial $-\Delta T$ value will be the most non-equilibrium crystal form (e.g. skeletal crystals and fan spherulites) and later more equilibrium forms (e.g. laths) will represent the stable morphologies for the smaller $-\Delta T$ values as isothermal crystallization progresses (Lofgren, 1974).

At small $-\Delta T$ values plagioclase assumes an equilibrium lath-like habit with planar grain boundaries. Increasing $-\Delta T$ leads to non-equilibrium skeletal crystallization with internal and external crystal boundaries sometimes having lobate forms. Within the skeletal group there is no obvious systematic variation from simple to complex skeletal types as $-\Delta T$ increases. With further increases in $-\Delta T$, fan spherulites become the stable form. This habit is initiated by a crystal beginning to branch with one or more branches. The fans at small $-\Delta T$ values are coarse and open, but with increasing $-\Delta T$ they become finer and more closed. The branching, which is usually asymmetrical, occurs initially at small angles to the principal fibre axis and later from both the original fibre and subsequently developed branches. Rare bow-tie spherulitic forms do occur. At the smaller $-\Delta T$ values the individual plagioclase laths in the fans often assume a skeletal form, but as $-\Delta T$ increases the laths appear to become predominantly non-skeletal. As in the skeletal group, grain boundaries need not be planar but can be lobate and cellular. Some of the main forms are shown in fig. 1.

In sample G.18, fan spherulites were generally absent and instead at $-\Delta T = 64$ °C the skeletal morphology was replaced by much thinner acicular crystals which were rarely skeletal and which were sometimes intergrown with second-phase clinopyroxene fan spherulites. This reversal to a 'more equilibrium' crystal morphology could be explained by the fact that once clinopyroxene nucleates in this sample it crystallizes more rapidly the plagioclase because of its simpler crystal structure (Corrigan, 1982), leading to lower $-\Delta T$ values with the result that further plagioclase nucleation assumes morphologies characteristic of lower $-\Delta T$ values. It is of interest in this respect that a melt supercooled to $-\Delta T = 85$ °C for 2 hours contained acicular plagioclase in the clinopyroxene-rich areas of the bead but the plagioclase had a skeletal and fan spherulitic form in the clinopyroxene-depleted areas.

The effect of $-\Delta T$ *and time on morphology.* In order to determine the effect of $-\Delta T$ and time on the morphologies produced, the growth forms present in each of the charges were plotted on $-\Delta T$ versus time diagrams for each of the seven melts investigated (fig. 2*a*-*g*). The boundaries between the fields of different growth forms were assumed to be linear and were placed at the largest $-\Delta T$ values consistent with the data. In each individual charge the most non-equilibrium growth habit was taken as the stable form at a particular $-\Delta T$ condition. Most of the plots define the skeletal to fan spherulitic boundary, which in fig. 2*a*-*c* is dependent on $-\Delta T$ and time, although its slope varies. The presence of charges with only skeletal forms in the fan spherulitic field of fig. 2*c* might be due to failure to observe fan spherulites through biased thin sectioning. In fig. 2*d* it is not clear whether the boundary is dependent on time, and in fig. 2*e*-*g* the boundary appears to be relatively independent of time. In fig. 2*e*-*f* the morphological boundary between laths and skeletal forms is also independent of time.

The effects of melt composition and viscosity on morphology. To determine the effect of bulk composition on plagioclase morphologies grown from supercooled melts, fig. 2*a*-*c* can be directly compared since the three samples were superheated initially to approximately the same $+\Delta T$ value

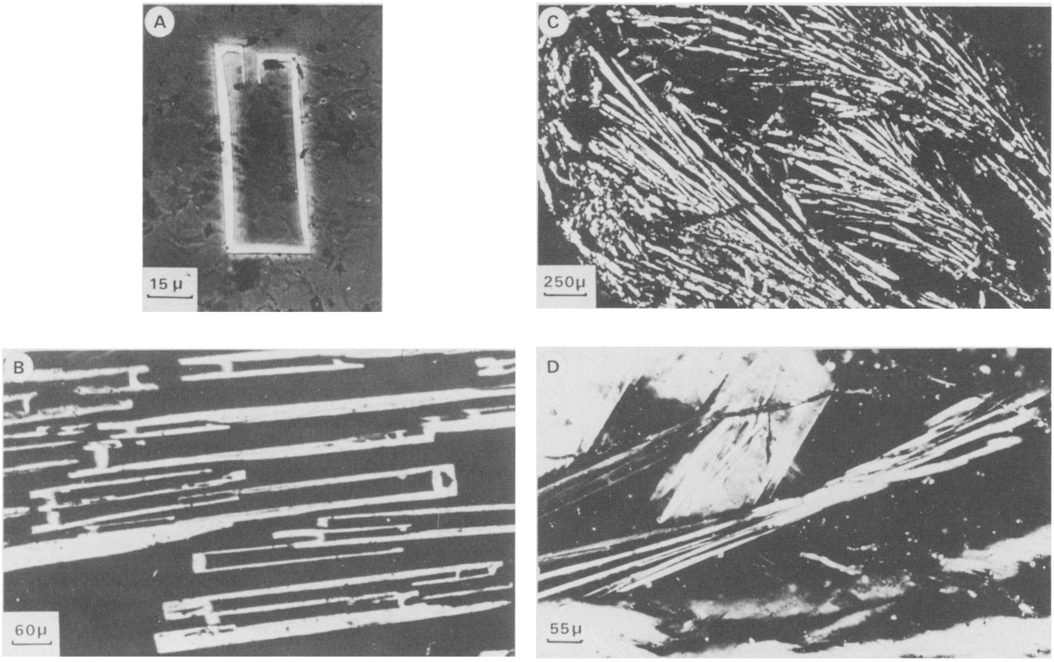


FIG. 1. (*a*) Skeletal plagioclase open at one end with hooked terminations, grown during isothermal supercooling of sample G.60. $+\Delta T = 42\,°C$, $-\Delta T = 36\,°C$, $T = 1201\,°C$, time = 2.75 hours. Partially crossed polars. (*b*) Skeletal chain plagioclase, grown during isothermal supercooling of sample G.1. $+\Delta T = 22\,°C$, $-\Delta T = 96.5\,°C$, $T = 1161\,°C$, time = 8 hours. Crossed polars. (*c*) Open and coarse branching plagioclase fan spherulites, grown during isothermal supercooling of sample 339. $+\Delta T = 24\,°C$, $-\Delta T = 101\,°C$, $T = 1075\,°C$, time = 20 hours. Crossed polars. (*D*) Plagioclase bow-tie fan spherulite, grown during isothermal supercooling of sample G.60. $+\Delta T = 43\,°C$, $-\Delta T = 84.5\,°C$, $T = 1152.5\,°C$, time = 1 hour. Crossed polars.

(sample 339 is ignored as the skeletal/fan spherulitic boundary is uncertain). The $-\Delta T$ value of the skeletal/fan spherulitic boundary at fixed time is plotted against melt viscosities (calculated at the appropriate isotherms) in fig. 3. The boundary moves to smaller $-\Delta T$ values at lower melt viscosities. This could be attributed to the decreased viscosity leading to greater diffusion in the melt and thus aiding nucleation and increasing the likelihood that new nuclei on the skeletal crystals will reach the critical size and begin to branch.

The effect of $+\Delta$T *on morphology.* Lofgren (1974) suggested that $+\Delta T$ could be a potential morphological control, but ignored it in his studies. It is likely that $+\Delta T$ would affect the shape, size, and position of the morphological fields since increasing $+\Delta T$ has been shown to displace the supercooling 'line' markedly towards greater $-\Delta T$ values (Corrigan, 1982).

The effect of differences in $+\Delta T$ on the skeletal/fan spherulitic boundary and the size of the skeletal field for sample 028 is shown in fig. 2*h*. The same sample (028) at $+\Delta T = 36\,°C$ (fig. 2*i*) shows the fan spherulitic forms close to the supercooling 'line' and the disappearance of the skeletal field. Increasing $+\Delta T$ moves the supercooling 'line' to progressively larger $-\Delta T$ values and raises the morphological field boundaries to smaller $-\Delta T$ values, leading, as in fig. 2*i*, to no skeletal field and to fan spherulites being the first form to crystallize. The results indicate that the morphology of crystals in natural rocks is not just dependent on $-\Delta T$, but is also a complex function of $+\Delta T$.

Further evidence that $+\Delta T$ controls the position of the boundary is shown in fig. 4, where the 5, 10, and 20 hour lines from fig. 3 are reproduced and the corresponding $-\Delta T$ values from the apparently time-independent boundaries for samples G.1, G.18, and G.60 are plotted. It is evident that the results for samples 028, 331, and 336, which were superheated by similar amounts and those for samples G.1, G.18, and G.60 which were superheated by different amounts are inconsistent, suggesting that the position of the morphological boundaries is controlled to some extent by the degree of super-

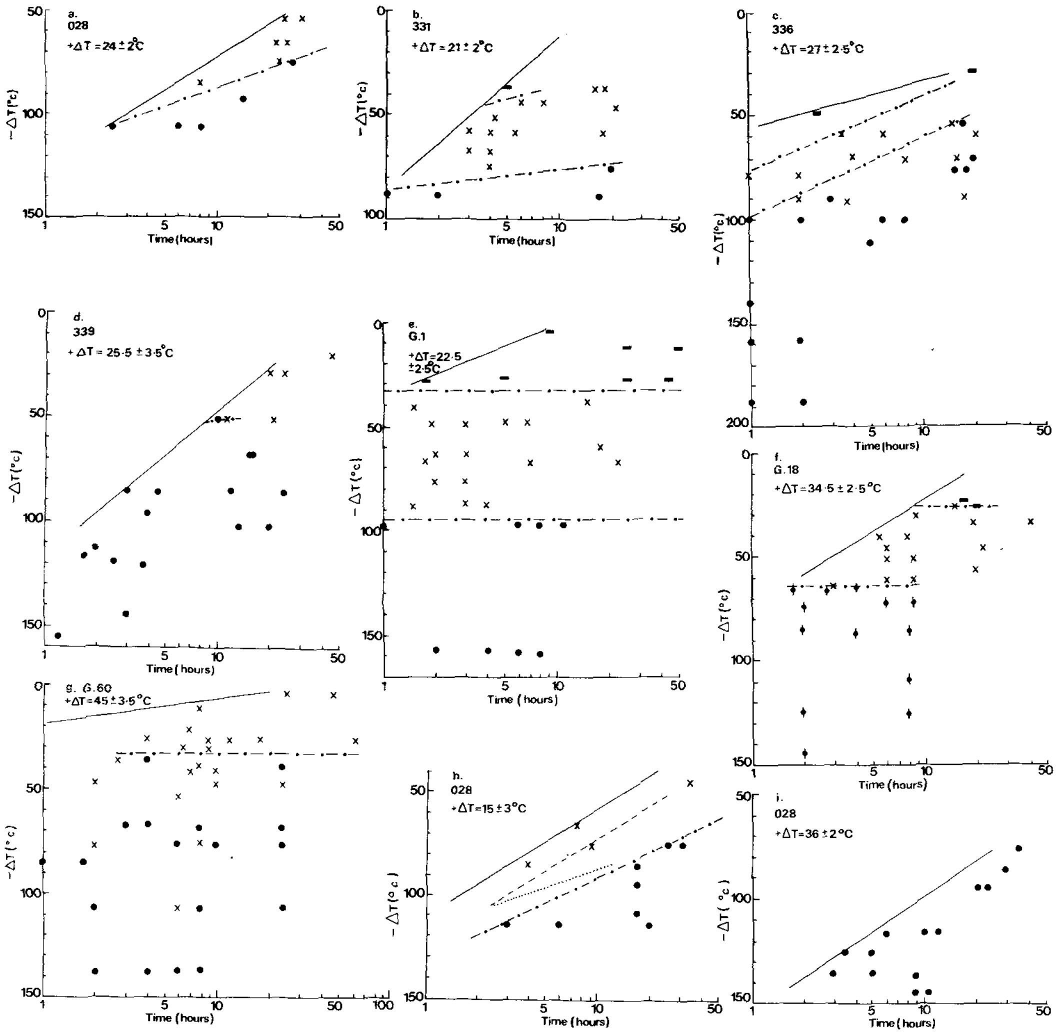


FIG. 2 (*a–i*). Plagioclase morphologies for samples 028, 331, 336, 339, G.1, G.18, and G.60 as a function of $-\Delta T$ and time. ▬ = Laths, × = Skeletal forms, ● = Fan spherulites, ⧫ = Acicular forms. The solid lines are plagioclase supercooling 'lines' (Corrigan, 1982). The dotted/dashed lines are boundaries between crystal morphological fields. For (*h*) the dotted line is the boundary between the skeletal and fan/spherulitic forms and the dashed line is the isothermal supercooling 'line' from (*a*).

heating. The data are, therefore, consistent with the view that $+\Delta T$ may have a significant affect on the growth morphology observed at any isotherm.

Constant rate cooling studies

For the composition G.18, the same plagioclase morphologies were observed as in the isothermal supercooling experiments. Lofgren (1974) observed similar sequences of plagioclase crystal morphologies with increasing cooling rate as with increasing $-\Delta T$ but by contrast Donaldson (1976) observed for olivine that the sequence of morphologies as a function of increasing $-\Delta T$ was only in partial accordance with the sequence of morphologies grown in cooling rate experiments.

In the present experiments two distinct habits were observed: polysynthetically twinned skeletal plagioclase with less skeletal plagioclase in the same charge; and smaller, thin, acicular skeletal and non-skeletal plagioclases, abundant in the

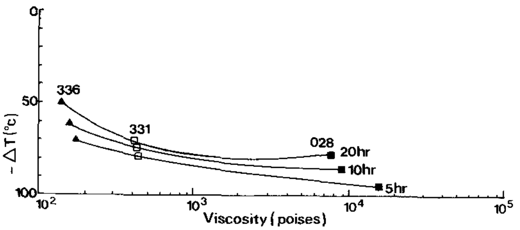


FIG. 3. Positions of the skeletal/fan spherulitic boundary for different crystallization times in terms of $-\Delta T$ and viscosity for samples 028, 331, and 336.

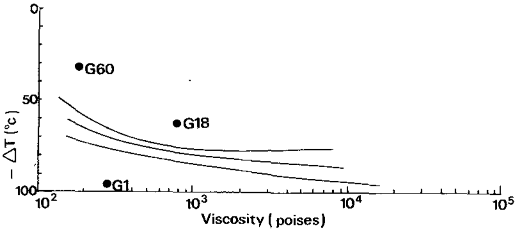
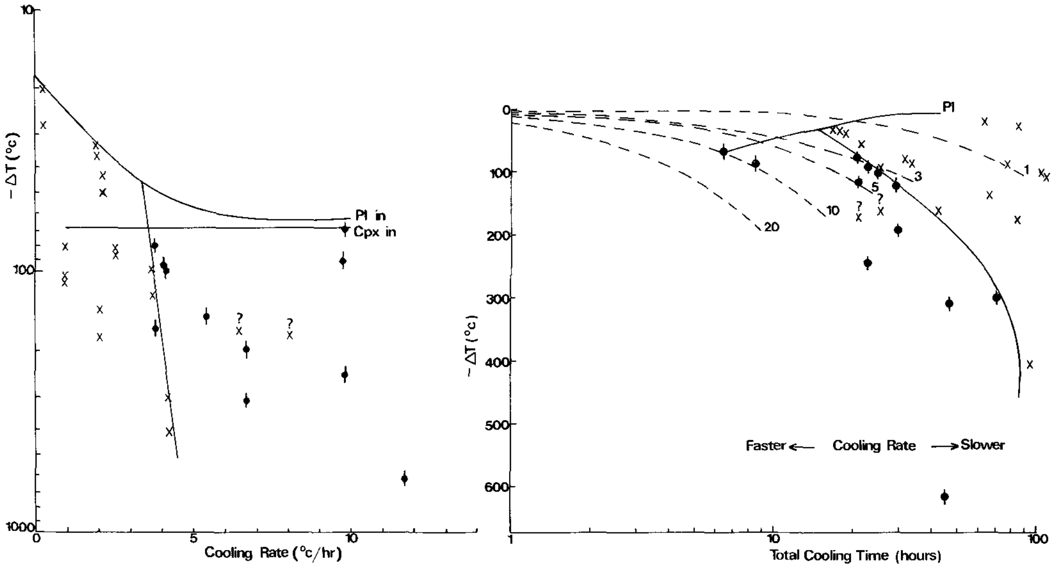


FIG. 4. Positions of the skeletal/fan spherulitic boundary for samples G.1, G.18, and G.60 compared with those in fig. 3. For these samples the boundaries are essentially independent of time (see figs. 2*e*–*g*).

charges containing dense branching clinopyroxene spherulitic forms. In a single charge, large skeletal plagioclase crystals sometimes coexist with acicular (skeletal and non-skeletal) plagioclase crystals, the actual proportions seeming to be related to the abundance of clinopyroxene fan spherulites. As in the isothermal supercooling experiments, the acicular morphology could be attributed to the fact that clinopyroxene growth releases latent heat and thus reduces $-\Delta T$. The distribution of these often patchily zoned morphological types as a function of $-\Delta T$ and cooling rate is given in fig. 5 and as a function of $-\Delta T$ and of total cooling time in fig. 6, where the most non-equilibrium form has been used to typify each charge. In both figures it is apparent that, except for two seemingly anomalous points, the morphological type is dependent on the cooling rate, with slower cooling rates favouring the skeletal morphology and faster cooling rates favouring the acicular morphology. Lofgren and Donaldson (1975*b*) showed in a series of monotonic cooling rate experiments with H_2O-undersaturated (approximately 5 wt. % H_2O) plagioclase melts, that 'elongate crystals' grew at slower cooling rates than 'acicular crystals'. Fig. 6 indicates that the boundary between the two morphologies is time dependent. From fig. 5 it is evident that the type of morphology present can be used to a certain



FIGS. 5 and 6. FIG. 5 (*left*). Plagioclase morphology as a function of $-\Delta T$ and cooling rate for sample G.18. × = skeletal laths and ♦ = acicular laths. FIG. 6 (*right*). Plagioclase morphologies produced during constant cooling rate runs using sample G.18 as a function of $-\Delta T$ and total cooling time. Symbols as for fig. 5. The solid curve (labelled Pl) denotes the 'plagioclase in' line from fig. 5. The numbered curves (dashed) refer to cooling rate curves.

extent as an indicator of the range of supercooling over which the first crystals nucleated and grew. For the acicular forms the $-\Delta T$ values would be in the range of 45–63 °C. The presence of skeletal forms, however, cannot be used as an indicator of the range of $-\Delta T$ over which they first crystallized since the slope of the morphological boundary in fig. 5 indicates that at cooling rates of 3 to 4.5 °C/hour acicular crystals will grow at small $-\Delta T$ values but with further supercooling they will transform to, or become overgrown to form, skeletal crystals. Similarly, from fig. 6, a melt cooling at 1 °C/hour will nucleate only skeletal crystals whereas a melt cooling at 3 °C/hour will first nucleate acicular crystals but at about $-\Delta T =$ 80 °C crystallization will move over into the skeletal field. A melt cooling at 5 °C/hour will nucleate acicular crystals and it seems likely that if this cooling curve does eventually intersect the morphological boundary it will do so at such a large $-\Delta T$ value (approximately 450 °C) that the melt kinetics will prevent a morphological change from taking place. A melt cooling at rates faster than 10 °C/hour will nucleate only acicular forms.

In the second sample studied (G.1) a characteristic feature was that plagioclase often occurred with a bimodal size distribution in a single charge, large, twinned, skeletal laths and sometimes open fan spherulitic forms coexisting with interstitial groundmass twinned plagioclase which was not normally skeletal. In many cases the larger crystals stemmed from the wire support and are thought to have nucleated early and heterogeneously (hence their large size). In one particular case a run cooled at 2 °C/hour and quenched at $-\Delta T = 166$ °C contained only large plagioclase crystals and no granular plagioclase. This fact could be explained if the first plagioclase to nucleate did so heterogeneously, and its subsequent growth depleted the melt in components necessary for further plagioclase nucleation. It is of interest in this respect that the only mafic phase to nucleate in this charge was olivine. Presumably the calcium necessary for the nucleation and growth of clinopyroxene had had its activity in the melt lowered. At faster cooling rates greater than 10 °C/hour there was a tendency for the plagioclase to be acicular in habit and to be intergrown with clinopyroxene fan spherulites. At a cooling rate of 208 °C/hour plagioclase grew as fine fan spherulites.

In the third sample (G.60) plagioclase sometimes occurred in two crystal sizes (e.g. at a cooling rate of 2.2 °C/hour and $-\Delta T = 132.5$ °C large boxwork plagioclase occurred with minor smaller, skeletal, and non-skeletal laths) or sometimes as just dominant large skeletal grains (e.g. at a cooling rate of 5.4 °C/hour and $-\Delta T = 154$ °C). At a cooling rate of 5.4 °C/hour, but with quenching at $-\Delta T = 242$ °C plagioclase crystallized as open branching fan spherulites. At a cooling rate of 10.7 °C/hour and quenching at $-\Delta T = 228$ °C the large plagioclase crystals had either a fan spherulitic form or else were skeletal with diffuse, ragged crystal boundaries, which may indicate that they had reacted with the melt.

Conclusions

In the two types of experiments similar plagioclase morphologies grew in the samples investigated, but the conditions at which they grew varied in each type of experiment, e.g. for sample G.60 plagioclase fan spherulites first grew at $-\Delta T =$ 56 °C during the isothermal supercooling experiments but were not observed in the constant cooling rate experiments until $-\Delta T = 242$ °C at a cooling rate of 5.4 °C/hour.

As a result of the isothermal supercooling experiments, the position of the plagioclase morphological fields is shown to be dependent on $-\Delta T$ and also on time to a greater or less degree. Variation in melt composition affects the position and slope of the boundary between the skeletal and fan/spherulitic fields, and for charges superheated to the same extent it is apparent that after a certain time the boundary between the two fields exists at smaller $-\Delta T$ values for lower melt viscosities. Superheating affects the position of the skeletal/fan spherulitic boundary and the size of the skeletal field, such that the size of the latter decreases with increasing $+\Delta T$. If certain mineral morphological changes are a function of $+\Delta T$ and time as well as $-\Delta T$, then this exerts severe limitations on the possible use of morphology as a petrological tool and indicator of the $-\Delta T$ at which crystals nucleated and grew in igneous rocks.

In the constant rate cooling experiments plagioclase morphology appears to be dependent on the cooling rate, but it has limited uses as an indicator of the range of supercooling over which the first crystals nucleated and grew.

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REFERENCES

Corrigan, G. (1980) Unpub. Ph.D. thesis, Univ. of Sheffield.

——(1982) *Mineral. Mag.* **46,** 31–42.

Donaldson, C. H. (1976) *Contrib. Mineral. Petrol.* **57,** 187–213.

Fenn, P. M. (1973) Unpub. Ph.D. thesis, Univ. of Stanford, USA.
Gibb, F. G. F. (1974) *Mineral. Mag.* **39**, 641-53.
Hamilton, D. L., and Anderson, G. M. (1967) In *Basalts*, New York (Interscience), 445-82.
Lofgren, G. (1971*a*) *J. Geophys. Res.* **76**, 5635-48.
——(1971*b*) *Geol. Soc. Am. Bull.* **82**, 111-24.
——(1973) *Trans. Am. Geophys. Union.* **54**, 482.
——(1974) *Am. J. Sci.* **274**, 243-73.
——and Donaldson, C. H. (1975*a*) *Trans. Am. Geophys. Union*, **56**, 468.
————(1975*b*) *Contrib. Mineral. Petrol.* **49**, 309-19.
Moore, J. G., and Lockwood, J. P. (1973) *Geol. Soc. Am. Bull.* **84**, 1-20.
Wyllie, P. J., and Drever, H. I. (1963) *Trans. Roy. Soc. Edin.* **65**, 155-77.

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