

Crystal sizes in intrusions of different dimensions: Constraints on the cooling regime and the crystallization kinetics

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Abstract—Crystallization in magmas depends on the crystallization kinetics as well as on the thermal regime. A dimensional analysis is presented which allows a simple understanding of the characteristics of crystallization. Characteristic scales for the rates of nucleation and crystal growth are used, denoted by I_m and Y_m respectively. The time-scale is given by $\tau_c = (Y_m^3 I_m)^{-1/4}$, and is close to the time required for crystallization to start in supercooled magma. The crystal size scales with $(Y_m/I_m)^{1/4}$, which provides a powerful constraint on the values of the nucleation and growth rates. The influence of the form of the kinetic functions for nucleation and growth is investigated. The form of the growth function relatively unimportant, in contrast to that of the nucleation function. In natural conditions following magma emplacement in cold country rocks, temperatures are continuously changing. Local scaling laws apply, whatever the boundary conditions are, with the characteristic time and crystal size given by $\tau = (Y^3 I)^{-1/4}$ and $R = (Y/I)^{1/4}$, where Y and I are the local rates. τ is the time to achieve crystallization and R the mean crystal size in a given piece of magma, and Y and I are the rates at which crystals were nucleated and grown locally. From petrological observations and laboratory crystallization experiments, the time-scale at high undercoolings is 2×10^5 sec. This gives the characteristic time for crystallization near the margins of intrusions. The time-scale is close to 10^8 sec in equilibrium conditions prevailing in the interior of large magma chambers. These can be compared to the characteristic times for cooling by conduction and convection. Several regimes are defined, depending on the intrusion dimensions. This allows a classification based on the average crystal size which agrees with petrological observations. The detailed study of dikes, sills and igneous complexes of different dimensions will allow constraints on poorly known conditions and parameters, such as the nucleation rate at small undercoolings.

INTRODUCTION

IN NATURE, magmatic crystallization proceeds in continuously evolving conditions, with both the melt temperature and composition changing. In large magma chambers, the end result is extremely complex, and the igneous record is difficult to decipher. Most studies so far have relied on equilibrium phase diagrams (MORSE, 1980) and on various dynamical processes such as thermal and compositional convection (HESS, 1960; JACKSON, 1961; WAGER and BROWN, 1968; MORSE, 1969; IRVINE, 1974; MCBIRNEY and NOYES, 1979; KERR and TAIT, 1986). The influence of the crystallization kinetics generally has been overlooked, despite a growing number of dynamic crystallization experiments in the laboratory (GIBB, 1974; WALKER *et al.*, 1976; DONALDSON, 1979; GROVE and BENCE, 1979; LOFGREN, 1980; KIRKPATRICK *et al.*, 1981; TSUCHIYAMA, 1983; BAKER and GROVE, 1985). These studies emphasize that crystals often nucleate metastably, and that the order of appearance of different crystalline phases follows that of their respective ease of nucleation (KIRKPATRICK, 1983). They further illustrate two critical facts. One is that nucleation is usually suppressed to undercoolings of several tens of degrees. The other is that the cool-

ing rate determines which phase nucleates first. The consequence is that the internal differentiation of magma bodies may not follow equilibrium paths, which presents an obvious problem when interpreting petrological observations. MORSE (1980, p. 229) and others (KIRKPATRICK, 1983; BAKER and GROVE, 1983) have recognized the petrological implications.

To estimate cooling rates in natural conditions, a common practice is to use simple thermal models such as those by JAEGER (1968) and then to evaluate the consequences for dynamic crystallization. What is seldom recognized is that the kinetics themselves influence the cooling rate because the thermal evolution is determined not only by the heat loss mechanism but also by the crystallization rate through the release of latent heat (KIRKPATRICK, 1976; BRANDEIS *et al.*, 1984). The evolution of crystallization in a magma body depends therefore on two factors: the kinetics of crystal nucleation and growth, and the cooling regime. The first depends on local conditions (temperature) and the second on conditions in the whole magma body. Both are poorly known for natural systems, which prevents the direct interpretation of petrological observations. Only slow progress can be expected in the near future from laboratory crystallization

experiments for several reasons. One is that it is difficult to study natural silicate melts with complex chemical compositions and phase relationships. Another is that such natural melts take long times to crystallize. A third reason is that true crystallization conditions are continuously changing, and determined by the coupling between cooling and latent heat release. To proceed towards quantitative understanding of igneous rocks, one can try an integrated approach where field data are used to constrain poorly known parameters and processes. For example, if the thermal regime is known, crystal sizes can be calculated using the crystallization equations (BRANDEIS *et al.*, 1984; BRANDEIS and JAUPART, 1986a). Comparisons with field data can then be used to calibrate the crystallization kinetics. On the other hand, if the kinetics are known, the field data can be converted to temperature and hence yield information on the cooling regime. BRANDEIS and JAUPART (1986a) have used the dimensional analysis of crystallization equations for conduction cooling together with crystal size data from dikes to obtain values for the nucleation and growth rates. The analysis is valid for thin dikes. Conduction is not, however, the main mechanism of heat transport in large magma bodies (SHAW, 1965). In those, convection dominates and changes the cooling history. Because the efficiency of convection is directly related to the size of the body, constraints can be obtained by comparing magma intrusions of different dimensions. For example, dikes of various thicknesses can be studied and compared to large magma chambers.

In this paper, we attempt to define the crystallization behaviour and thermal regime of intrusions of different dimensions, using constraints from the distribution of crystal size. Recent advances on how to model dynamic crystallization and scaling laws for crystal size variations are summarized. We define and give plausible values for the characteristic crystallization time and compare it to the cooling time for magma bodies of different sizes and chemical compositions. The plan is the following. First, a brief review of crystallization kinetics is given. Then, crystallization by conduction at the margin of a dike is addressed. The influence of different expressions for the kinetic functions is investigated. Finally, the various cooling regimes are reviewed and the thickness range for intrusions discussed.

KINETIC CONTROLS ON CRYSTALLIZATION

The effects of crystallization kinetics on natural samples are well documented, most notably in lava flows and mid-ocean ridge pillow basalts where pi-

geonite, the equilibrium liquidus phase, is absent (BRYAN, 1972; BAKER and GROVE, 1985). The first hint at kinetic effects in the petrological literature came perhaps from WAGER (1959) who recognized that nucleation could control the abundance of the different mineral phases that appear in layered igneous complexes. The first specific study on silicate melts was by GIBB (1974) on Columbia River basalt, who found that the temperature at which plagioclase begins to crystallize varies as a function of cooling rate. Since then, many dynamic crystallization experiments have been made (see references above). Following common practice in metallurgy, these are carried out under two different conditions: isothermal (constant temperature) and continuous cooling (constant cooling rate). The evolution of the crystallized product is followed as a function of time, yielding TTT (time-temperature-transformation) and CT (continuous cooling-transformation) diagrams respectively (SHEWMON, 1969). There are, unfortunately, no simple relationships between the two types of experiments on the same starting material (DONALDSON, 1979; TSUCHIYAMA, 1983). Thus, it is difficult to use them for extrapolation to natural conditions which are transient and often out of the range of the laboratory.

Another approach is to study directly the crystallization process by determining the rates of nucleation and growth. Nucleation and growth are distinct phenomena obeying different rules. In silicate melts, new crystal formation results either from the presence of foreign material in the melt, such as impurities or nuclei from a distant source, and from the fortuitous formation of molecular clusters of critical size, *i.e.*, nuclei. These are called heterogeneous and homogeneous nucleation respectively. The rates for both phenomena can be expressed as a function of temperature and undercooling following kinetic theory (JOHNSON and MEHL, 1939; TURNBULL and FISHER, 1949). The general nucleation rate function has a bell-shape, illustrated in Figure 1 and subsequently called "shape 1 function". Crystal growth, on the other hand, requires two steps: solute must be transported to the crystal surface, a process that is usually controlled by chemical diffusion, and then oriented into the crystal lattice (attachment) (KIRKPATRICK, 1975; BARONNET, 1984). The attachment kinetics are relatively well understood and the corresponding growth-rate function also has a bell-shaped curve (BARONNET, 1984), illustrated again in Figure 1. In reality, when crystals grow large and fast, diffusion of solute through the melt becomes limiting. Unfortunately, there is no simple method to treat diffusion in a crowded environment with many crystals

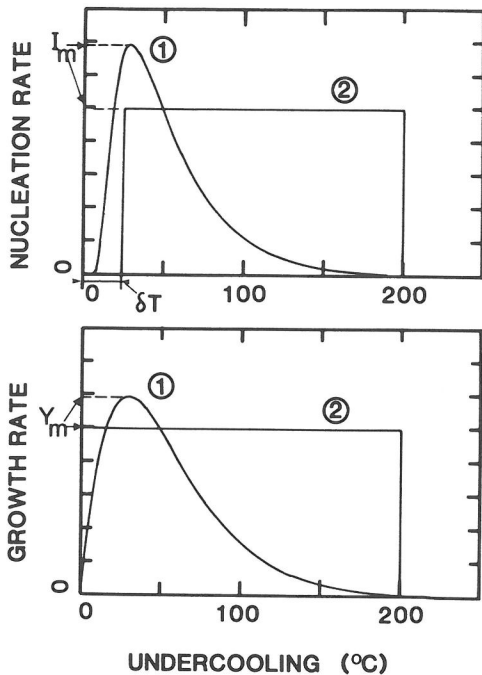


FIG. 1. The functions for the rates of crystal nucleation and growth, expressed as a function of undercooling. The scales are arbitrary. To define these kinetic functions completely, the scales I_m and Y_m must be specified. The bell-shaped curves correspond to standard kinetic theory (BRANDEIS *et al.*, 1984) and are termed "shape 1" functions. "Shape 2" functions are the simplest ones, and are used to investigate the influence of the shapes of the nucleation and growth curves. In each case, the nucleation and growth scales, are used for dimensional analysis. For shape 1 functions, these are the peak rates. In the shape 2 case, the scale is the constant value taken by the function. Note the delay of nucleation δT , which is the undercooling range for which nucleation is negligible.

and there is no general expression for the growth rate function. Under natural conditions, it seems that the attachment kinetics are the controlling phenomenon (KIRKPATRICK, 1977).

This brief summary probably makes it clear that there is no obvious theoretical way to specify the functions for nucleation and growth. The common practice in industrial crystallizers is to use empirical expressions derived from experimental data (RANDOLPH and LARSON, 1971, p. 110). To this end, two different steps are required. One is to specify some characteristic value for the growth and nucleation rates. These will be called the growth and nucleation scales. The other is to specify the form of the function. The bell-shaped curves of Figure 1 have been determined from standard kinetic theory (BRANDEIS *et al.*, 1984). One of the aims of this paper is to investigate the influence of the kinetic

functions on the crystallization behaviour. This will define the precision required from experimental data to allow realistic quantitative models.

Data are available on nucleation and growth rates in silicate melts. Most experiments show bell-shaped nucleation and growth curves, and in the following, we use published values to constrain the peak rates. The most complete set of measurements are those by FENN (1977) and SWANSON (1977). From these, BRANDEIS *et al.* (1984) demonstrated that the peak nucleation rates had to be between 10^{-2} and $10^2 \text{ cm}^{-3} \text{ sec}^{-1}$. In a basaltic lava lake, KIRKPATRICK (1977) estimated values for plagioclase ranging from about 10^{-2} to $1 \text{ cm}^{-3} \text{ sec}^{-1}$. More recently, TSUCHIYAMA (1983) obtained a value of $10^{-2} \text{ cm}^{-3} \text{ sec}^{-1}$ for diopside in the system $\text{CaMgSi}_2\text{O}_6\text{-CaAl}_2\text{Si}_2\text{O}_8$. The total range of possible values is, therefore, quite large, covering four orders of magnitude. Data on growth rates are more numerous and have been compiled by DOWTY (1980). From those, it appears that the growth rate decreases as the chemical system becomes more complex. For natural compositions, peak growth rates should be smaller than $10^{-7} \text{ cm sec}^{-1}$. In his natural crystallization experiment, the cooling lava lake, KIRKPATRICK (1977) gave values between 10^{-10} and $10^{-9} \text{ cm sec}^{-1}$ for plagioclase. These values were obtained at small undercoolings and provide lower bounds for the peak rates. From these considerations, peak growth rates in silicate systems range from 10^{-9} to $10^{-7} \text{ cm sec}^{-1}$.

There is therefore quite a significant body of experimental data, showing significant differences among the various systems. One important question is to assess whether these differences lead to important variations in crystallization conditions. This question will be addressed in this paper. Once the kinetic functions for nucleation and growth are specified, it is possible to write the equation for crystallization involving the coupling between cooling and latent heat release. The first effort in the geological literature was by KIRKPATRICK (1976). However, he made a mathematical error which prevents reliable results. DOWTY (1980) investigated the test-case of a fixed rate of heat loss. He showed that latent heat release is very important and suppresses large undercoolings, bringing temperatures close to the liquidus even for rates of heat loss that are high by geological standards. His calculations give useful insights into the crystallization behaviour of silicate melts, but do not take into account the coupling with heat flow through country rocks and the continuous evolution of heat loss conditions as crystallization proceeds. Following these studies, BRANDEIS *et al.* (1984) tackled the

full problem and presented solutions for conduction cooling in a variety of cases. BRANDEIS and JAUPART (1986a) later made a dimensional analysis of the same equations. In this paper, we summarize their results and investigate the consequences for the cooling regimes of magma bodies of different dimensions. In return, these considerations will allow constraints on the nucleation and growth rates.

DIMENSIONAL ANALYSIS FOR CRYSTALLIZATION

We consider a problem in one dimension only (z), perpendicular to the contact between magma and surrounding rocks. It is assumed that heat transfer is dominated by conduction, which is valid in boundary layers close to rigid boundaries (roof, floor and side-walls). The heat equation is written:

$$\partial T / \partial t = \kappa \partial^2 T / \partial z^2 + L / c_p \partial \Phi / \partial t \quad (1)$$

where c_p is the isobaric heat capacity, κ thermal diffusivity, T temperature, t time and L the latent heat per unit mass. κ is thermal diffusivity. Φ is the crystal content per unit volume and takes values between 0 and 1. $\partial \Phi / \partial t$ depends on the rates of nucleation and growth according to the equation (KIRKPATRICK, 1976):

$$\partial \Phi / \partial t = 4\pi [1 - \Phi(z, t)] Y(t) \int_0^t I(v) \times \left[\int_{t_v}^t Y(u) du \right]^2 dv. \quad (2)$$

I and Y are the rates of nucleation and growth, respectively, and t_v is time of nucleation.

We consider the simplest cooling experiment. At time $t = 0$, magma with initial temperature T_L (the liquidus) is emplaced in country rocks that are colder by an amount ΔT . The initial conditions are that both magma and country rock are initially isothermal:

$$T(z, 0) = T_L \quad \text{for } z > 0 \quad (\text{magma}) \quad (3a)$$

$$T(z, 0) = T_L - \Delta T \quad \text{for } z < 0 \quad (\text{country rocks}). \quad (3b)$$

The boundary conditions are:

$$T(+\infty, t) = T_L \quad (3c)$$

$$T(-\infty, t) = T_L - \Delta T. \quad (3d)$$

Note that these boundary conditions are specified at infinity. In practice, the magma body is of finite dimensions. This is not limiting for short times because conduction propagates slowly and only affects

a boundary layer that advances into uncrystallized magma. The main limitation of these equations is the conduction approximation, which will be discussed later.

The physical properties (κ , c_p) are assumed to be constant. The temperature scale is ΔT , and the scales for the rates of nucleation and growth are denoted by I_m and Y_m . A time-scale appears when Equation (2) is made dimensionless:

$$\tau_c = \{ Y_m^3 I_m \}^{-1/4}. \quad (4a)$$

Because heat transfer is by conduction, the corresponding length-scale is simply given by:

$$d_c = (\kappa \tau_c)^{1/2}. \quad (4b)$$

The non-dimensional variables are denoted by primes:

$$t = t' \tau_c \quad (5a)$$

$$z = z' d_c \quad (5b)$$

$$I = I' I_m \quad (5c)$$

$$Y = Y' Y_m \quad (5d)$$

$$T = T' \Delta T. \quad (5e)$$

With these, Equations (1) and (2) can be made dimensionless, which introduces a non-dimensional number called the Stefan number:

$$\sigma = \frac{L}{c_p \Delta T}. \quad (6)$$

The Stefan number is a measure of the importance of latent heat in the temperature equation. If it is small ($\sigma \ll 1$), temperature is given by the heat equation without latent heat. In geological cases, the Stefan number is of order 1, which implies that latent heat must be taken into account.

Crystallization proceeds within a region of finite thickness called the crystallization interval, which advances into uncrystallized magma. The moving boundary between fully crystallized and crystallizing magma is called the crystallization front and is such that the crystal content Φ is equal to 0.99 (Figure 2). Its coordinate is denoted by $X(t)$. At $z = X(t)$, the temperature is $\theta(t)$. The crystallization interval is defined as the zone where $0.01 < \Phi < 0.99$ (Figure 2) and has thickness $\epsilon(t)$. Both $X(t)$ and $\epsilon(t)$ scale with the crystallization length-scale defined by Equation (4b).

It is also possible to calculate the crystal size. In dimensional variables, a unit volume of crystallized material comprises N crystals, with mean radius R :

$$N(z) = \int_0^\infty [1 - \Phi(z, t)] I(t) dt \quad (7a)$$

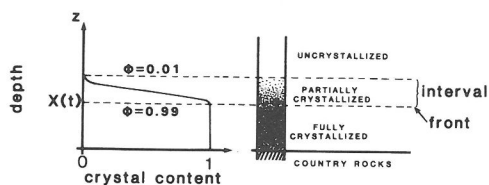


FIG. 2. Definition of the crystallization variables. The crystallization front is the moving boundary between crystallizing and fully crystallized magma. Its coordinate is denoted by $X(t)$. The region where magma is partially crystallized is called the crystallization interval, defined as the zone where $0.01 < \phi < 0.99$. Its thickness is $\epsilon(t)$.

$$R(z) = [4/3\pi N(z)]^{-1/3}. \quad (7b)$$

A size-scale appears when Equation (7) is made dimensionless (BRANDEIS and JAUPART, 1986a):

$$R_c = \{Y_m/I_m\}^{1/4}. \quad (8a)$$

The dimensionless crystal size is obtained by:

$$R' = R/R_c. \quad (8b)$$

To summarize, knowledge of characteristic scales for the rates of nucleation and growth allow the definition of a time-scale, a length-scale and a size-scale, *i.e.* of all the important crystallization parameters. An important result has already been obtained. Both the time-scale and the size-scale depend on the nucleation rate are raised to the power ($1/4$), which shows that they are weakly sensitive to its value. The four orders of magnitude uncertainty on I_m is therefore not critical. The influence of the growth rate is slightly more important, but there are better data.

Because Equations (1) and (2) are coupled, the problem has to be solved numerically. We first derive dimensionless relationships for the different crystallization parameters. As they all depend on the kinetic rates, the influence of their shape on the crystallization behaviour will be investigated. Because both exhibit a steep increase just below the liquidus, it is worthwhile to evaluate how sensitive the results are to their shape. The influence of σ (Stefan number) is weak and only changes the values of the coefficients in the different relationships (BRANDEIS and JAUPART, 1986a).

SCALING LAWS FOR CRYSTALLIZATION

To derive the various scaling laws, "shape 1" nucleation and growth functions are used (Figure 1). These have been determined by using kinetic theory and laboratory data. For these, the scales are given by the peak values. An important parameter is the nucleation delay δT , which is the minimum un-

dercooling for the formation of nuclei. In the following, the Stefan number is taken to be 0.55, which corresponds to high temperature contrasts (BRANDEIS and JAUPART, 1986a).

The evolution of crystallization

The position of the crystallization front $X(t)$ is related to time according to a power-law (Figure 3):

$$X(t) = \lambda t^n \quad (9)$$

where the exponent, n , is close to $1/2$, but slightly different. The $1/2$ power-law corresponds to the ideal case of latent heat release at a fixed melting point (JAEGER, 1968) or of a binary alloy without kinetic effects (WORSTER, 1986). The crystallization kinetics do not alter significantly this simple law, due to the control by heat diffusion.

The evolution of the crystallization interval thickness $\epsilon(t)$ is shown in Figure 4. The general evolution is a slow increase with few oscillations due to the discontinuous character of the nucleation process (BRANDEIS *et al.*, 1984). For this value of σ , the data can be fitted with an error of a few per cent with a law (in dimensionless variables):

$$\epsilon(t) = 0.3t^{0.31}. \quad (10)$$

As cooling proceeds, undercoolings in the crystallization interval decrease. This is illustrated by the evolution of the undercooling at the crystallization front, shown in Figure 5. The general evolution is

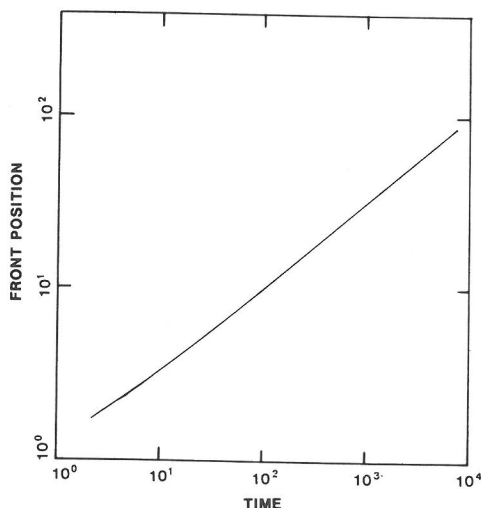


FIG. 3. Position of the crystallization front, $X(t)$, versus time in dimensionless variables for a Stefan number of 0.55. Time $t = 0$ marks the emplacement of magma in country rocks which are colder by an amount of ΔT .

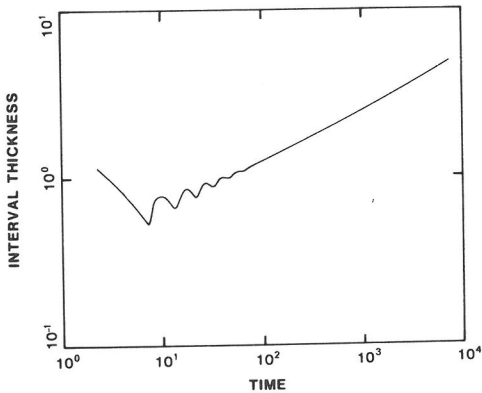


FIG. 4. Evolution of the interval thickness, $\epsilon(t)$, as a function of time in dimensionless variables for a Stefan number of 0.55.

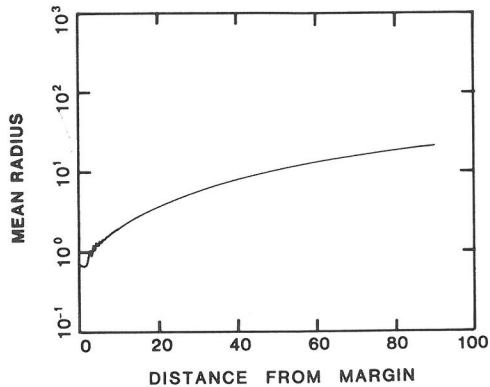


FIG. 6. Mean crystal size as a function of distance to the margin in dimensionless variables for a Stefan number of 0.55.

a decrease towards a small value which is equal to δT (BRANDEIS and JAUPART, 1986a). There are a few oscillations due to nucleation steps. These are damped as there is a tendency to achieve an equilibrium between latent heat release and heat loss. Note that θ has not been calculated for times $t < 1$ (in dimensional variables, for times smaller than the crystallization time-scale), because crystallization has not started and hence no crystallization front can be defined. This shows that τ_c [Equation (4a)] is close to the onset time for crystallization.

The crystal size

The variation of dimensionless crystal size as a function of the distance to the margin is shown in Figure 6. Near the margin, the dimensionless crystal size is close to 1. This shows that the size-scale, R_c , is equal to the crystal size there. Two kinds of factors must be considered for extrapolation to natural

conditions. The first is that, as crystallization proceeds, processes other than thermal conduction may become limiting. In particular, the whole magma chamber is cooling, which implies a change in boundary condition (3c) for the temperature equation. A second factor is that most crystallization occurs in equilibrium conditions at small undercoolings. The measurements by FENN (1977) and SWANSON (1977) only allow constraints on the peak nucleation rate. At small undercoolings, a detailed understanding of how the nucleation rate varies with temperature is lacking (see the discussion on oscillatory crystallization by BRANDEIS *et al.*, 1984). Specifically, the shape of the nucleation function must be known with precision. In the preceding calculations, the nucleation rate tends to zero continuously as θ tends towards the nucleation delay, δT . The unavoidable consequence is that the crystal size would eventually reach infinity, which is not realistic. This stresses the need for reliable data on nucleation rates at small undercoolings. The influence of the shape of the two kinetic functions is now addressed.

THE FORM OF THE KINETIC FUNCTIONS

We have so far relied on reasonable expressions for the relationships between the kinetic rates and temperature. We compare those, referenced as shape 1 functions, to the simplest ones: box-car functions, referenced as shape 2 (Figure 1). These functions have a constant value throughout the crystallization range and are zero elsewhere. They represent the limit-case of a discontinuous behavior. For the nucleation process, this is not unreasonable and approximates the effect of a finite energy barrier for the formation of one nucleus. In-

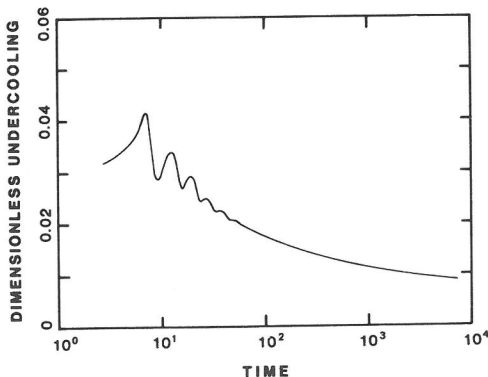


FIG. 5. Dimensionless undercooling at the crystallization front, $\theta(t)$, as a function of dimensionless time for a Stefan number of 0.55.

tuitively, these functions represent the obvious way to achieve a constant crystal size throughout the crystallization sequence. We show calculations for the crystal size for all possible combinations of shape 1 and shape 2 functions (Figure 7). We keep the same procedure for making variables dimensionless. For shape 2 functions, the scale is given by the constant value. In this section, the Stefan number, σ , is taken equal to 0, as this limiting case is simple and yet shows of the most features of the crystallization behaviour (BRANDEIS and JAUPART, 1986a).

How crystal size varies reflects the shape of the nucleation function. For a shape 2 nucleation function, the crystal size is constant after a small transient (Figure 7). The value is close to the size-scale defined by Equation (8a), with a proportionality constant between 0.5 and 0.7 depending on the shape of the growth function.

It is concluded that the shape of the nucleation function critically influences the results, contrary to that of the growth function. Nucleation is, therefore, the process governing crystallization. The main limitation of these calculations is, thus, the lack of constraints on the nucleation function. Crystal growth does play a role in determining the numerical constants in the scaling laws derived above, but does not have any effect on the crystallization behaviour. It is nucleation which is responsible for temperature oscillations as well as the tendency to maintain a constant crystal size.

DISCUSSION

Before applying this analysis to crystallization in magma bodies of different thickness, its validity in

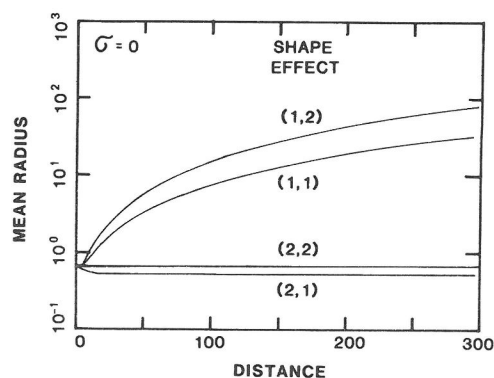


FIG. 7. Evolution of the mean crystal size as a function of the distance to the margin for several kinds of kinetic functions. The numbers in brackets represent the type of function used for nucleation and growth, respectively (shape 1 or 2, see Figure 1). The Stefan number is set equal to 0.

all cases needs assessment, as several assumptions have been made. These principally are two. First, chemical diffusion was neglected in the expressions for the kinetic rates. Second, cooling was achieved by heat conduction.

The crystallization kinetics

Crystal growth is controlled by chemical diffusion and reactions at the crystal-melt interface. The rate controlling process is diffusion at large times for a single crystal, as shown by the time variation of growth rate at fixed undercooling (LOOMIS, 1982; LASAGA, 1982). Diffusion is also important for rapidly cooled magma, for example in lava flows and chilled margins, resulting in spherulitic or dendritic crystal morphologies. In the interior of large intrusions, crystals do not exhibit these morphological instabilities and growth is assumed to be controlled by the interface attachment kinetics (CAHN, 1967; KIRKPATRICK, 1975; BARONNET, 1984). Also, there are thick adcumulate layers (WAGER and BROWN, 1968). Compositional convection can occur in the porous cumulate pile (MORSE, 1969; TAIT *et al.*, 1984; KERR and TAIT, 1986), bringing the required chemical components to achieve adcumulate growth.

Nucleation is the controlling phenomenon for crystallization. At any given depth, nucleation occurs essentially once and is followed by growing the existing nuclei (BRANDEIS *et al.*, 1984). Crystallization occurs in a thick region which moves into uncrystallized magma. The exact growth rate function has no influence on the results (Figure 7). By considering a mean value for the growth rate, which takes into account chemical diffusion and other processes, the scaling laws can be applied successfully. The comparison between petrological observations and calculations yields values for the peak nucleation and growth rate that are close to the experimental data (BRANDEIS and JAUPART, 1986a).

An important result of this study is that the shape of the nucleation function is a critical parameter, especially at small undercoolings. The fact that the crystal size does not vary in the interior of large igneous complexes shows that the nucleation rate does not tend to zero continuously. There is probably a limiting value, which presumably corresponds to a finite energy barrier for the formation of a nucleus of critical size.

We conclude that, in order to explain both the remarkably regular variation of the crystal size at the margins of dikes (Figure 8) as well as the uniformity of crystal sizes in the interior of magma chambers, one needs a combination of shape 1 and

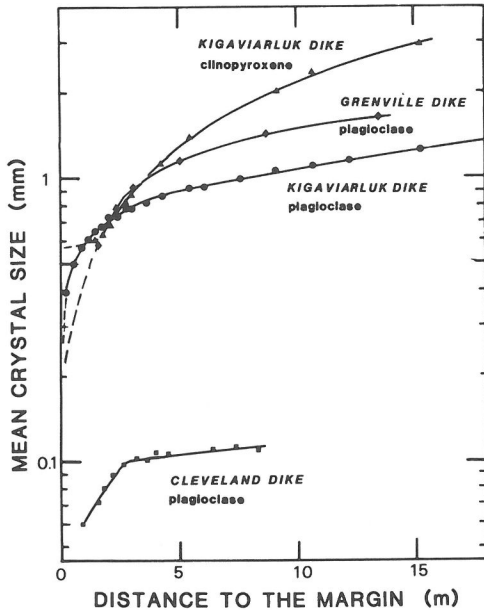


FIG. 8. Variations of crystal sizes in natural dikes (from WINKLER, 1949 for the Cleveland dike; GRAY, 1970 for the Kigaviarluk and Grenville dikes).

shape 2 functions for the nucleation rate. For scaling purposes, one must carry out two separate analyses. One is for highly transient conditions pertaining to the margins, where shape 1 functions must be used. The other is for equilibrium conditions pertaining to the interior where crystallization proceeds at small undercoolings, where a shape 2 nucleation function must be used.

Thermal regime

Two assumptions have been made in regard to the thermal regime. First, cooling is by conduction which is valid for thin dikes and sills, as well as at the bottom of large magma bodies. There, crystallization occurs mainly *in situ* (CAMPBELL, 1978; MCBIRNEY and NOYES, 1979) in stagnant layers isolated from convection (JACKSON, 1961; JAUPART *et al.*, 1984). At the bottom, there is a steep viscosity increase due both to lower temperature and higher crystallinity, and hence, any motions occurring in the chamber interior do not affect the crystallization interval. This indicates that conduction is the dominant means of heat transfer there (see also HUPPERT and SPARKS, 1980). The second assumption is made in boundary condition (3c), where we have, in fact, treated the case of an intrusion of infinite dimensions. In reality, this is not the case, which has implications investigated in the last section.

Local scaling laws

Equations (4a) and (8a) give correct values for highly transient conditions. In fact, the crystal size evolution shown in Figure 6 clearly reflects the evolution of the crystallization temperature (Figure 5). The same is true for the crystallization interval. From Equations (2) and (7), both the crystal size and the crystallization time can be defined locally, by using the instantaneous values of I and Y at the crystallization temperature, θ :

$$R = [Y(\theta)/I(\theta)]^{1/4} \quad (11a)$$

$$\tau = [I(\theta)Y(\theta)^3]^{-1/4} \quad (11b)$$

By using the curve for undercoolings at the crystallization front (Figure 5) and local relationships [Equation (11)], one can reproduce to a good approximation the curves for the crystal size and the crystallization interval. Even though conditions are highly transient, the local scaling procedure gives reasonable results, and should be useful even when cooling is affected by other processes such as convection. If the mean crystal size and the growth rate are known, Equation (11a) gives the local value of the nucleation rate, independently of the thermal regime and the conditions at the other boundaries.

BRANDEIS and JAUPART (1986a) have used the data from dike margins (Figure 9) to constrain the values of the maximum kinetic rates. The nucleation and growth scales, I_m and Y_m , are found to be close to $1 \text{ cm}^{-3} \text{ sec}^{-1}$ and $10^{-7} \text{ cm sec}^{-1}$, respectively. For transient conditions prevailing in early stages at the margins, the characteristic time scale is $2 \times 10^5 \text{ sec}$, and will be termed τ_1 .

In equilibrium cooling conditions that should prevail in the interior of large magma bodies, crystallization occurs at small undercoolings and hence at smaller rates. From petrological observations, we have suggested ranges of 10^{-7} to $10^{-3} \text{ cm}^{-3} \text{ sec}^{-1}$ for the local nucleation rate and a range of 10^{-10} to $10^{-8} \text{ cm sec}^{-1}$ for the local growth rate. These values agree with laboratory measurements (DOWTY, 1980). For those conditions, the time scale has a minimum value of 10^7 sec and is probably close to 10^8 sec . An upper bound is 10^9 sec . This time will be termed τ_3 . It is large, which has important implications detailed below.

THE TIMES FOR CRYSTALLIZATION AND COOLING

A brief summary is worthwhile at this stage. In the simple cooling experiment studied above, crystallization starts after a finite time close to τ_1 at undercoolings of several tens of degrees (Figure 5).

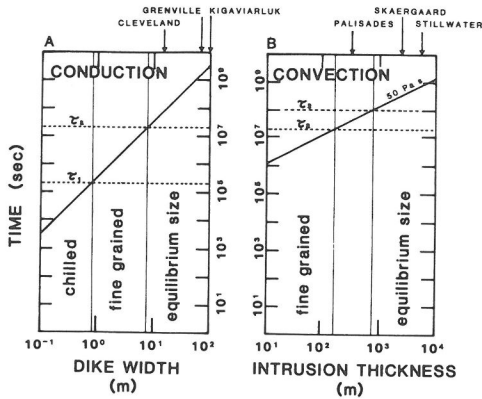


FIG. 9. Comparison between the characteristic times for cooling and crystallization in various cases. (A) Conduction cooling for dikes of different widths. The thick curve is from Equation (12) and gives the characteristic time for conduction cooling. τ_1 is the time for the onset of crystallization. Dikes which cool faster than τ_1 cannot nucleate crystals and hence are chilled. τ_2 is the time needed to achieve equilibrium cooling conditions. Dikes which cool faster than τ_2 crystallize at high undercoolings and hence develop small crystals. (B) Convection cooling in sills and magma chambers. The thick curve is from Equation (13) and gives the characteristic time for convection cooling. Intrusions which cool faster than τ_2 crystallize at high undercoolings and hence develop small crystals. τ_3 is the characteristic time for crystallization in equilibrium conditions at small undercoolings. If the cooling time is close to τ_3 , crystallization has time to record the effects of convective processes operating in the intrusion interior. This implies that complex igneous structures will be found in the solidified rocks.

At later times, undercoolings in the crystallization interval decrease and equilibrium cooling conditions are approached after a time, τ_2 , which is about $10^2 \times \tau_1$ (Figures 5 and 6). There is a marked difference between the cooling rates achieved in the initial transient regime, and the equilibrium rate. Note that the time needed to reach equilibrium conditions is solely determined by the crystallization kinetics. For the preferred values of the nucleation and growth scales, equilibrium conditions are reached once the thickness of crystallized magma exceeds about 5 m. This corresponds to the flattening of the crystal size curve in natural dikes (Figure 8).

In order to fit the dike data, values of $1 \text{ cm}^{-3} \text{ sec}^{-1}$ and $10^{-7} \text{ cm sec}^{-1}$ are required for the nucleation and growth scales. These are compatible with data from laboratory experiments (see above). This shows the use of crystal size data from intrusions. As discussed above, in any volume of igneous rock, the crystal size is given by the instantaneous values of the nucleation and growth rates that prevailed at

the time it crystallized. From both the theory given in this paper and the observations, the crystal size reaches a nearly constant value close to 1 mm in the interior of dikes away from the margins. This size is also that which is observed in almost all igneous complexes (see the compilation in Brandeis *et al.*, 1984). This observation suggests that most crystallization occurs under similar conditions with similar instantaneous values of the nucleation and growth rates.

Further progress in quantitative models necessitates knowledge of the nucleation and growth functions at small undercoolings. To this end, one possibility is to investigate intrusions of different dimensions. The calculations so far have relied on the assumption that the magma body is infinite. In this case, after the initial transients, uniform conditions are realized throughout the crystallization sequence, with, for instance, similar undercoolings and hence similar crystal sizes (Figures 5 and 6). In reality, a magma body is of finite dimensions and cools. The result is that it is no longer possible to assume a fixed temperature away from the crystallization interval, as was done explicitly in boundary condition (3c). This conclusion leads to a variety of cases depending on how temperature varies in the intrusion interior. For example, the limiting case is that of a very thin dike that cools very rapidly, leaving no time for crystals to nucleate, and hence chills. In the following, various possibilities for cooling conditions are discussed. The arguments are rough, and meant to illustrate basic principles and to indicate the direction for future studies. The idea is to compare the characteristic times for various phenomena. These can be defined to within maybe one order of magnitude, which is not critical because the range of natural conditions is much larger.

Conduction cooling in dikes of different widths

Conduction is the dominant mechanism of heat transport in thin dikes. For larger magma bodies, convection is important and modifies the thermal history. Because our understanding of convection in magmas is far from complete (Brandeis and Jaupart, 1986b), it is not easy to define the limit between conduction-dominated and convection-dominated cases. According to Shaw (1965), convection should be important in magma bodies exceeding thicknesses of a few tens of meters. For dikes that are vertical intrusions with very large aspect ratios, we consider that conduction dominates up to 100 m width. This spans the range of most dikes, including the Cleveland, Grenville and Kigavialuk

dikes. Those dikes are 16 m, 60 m and 106 m wide respectively (WINKLER, 1949; GRAY, 1970). If conduction dominates, a rough estimate for the cooling time is:

$$tc \simeq \frac{D^2}{4\kappa} \quad (12)$$

where D is the dike width and κ thermal diffusivity, for which we take a value $7 \times 10^{-7} \text{ m}^2 \text{ sec}^{-1}$. This cooling time ranges from $3.6 \times 10^3 \text{ sec}$ for a 10 cm-wide dike to $3.6 \times 10^9 \text{ sec}$ for a 100 m-wide dike (Figure 9a). τ_1 , the time for the onset of crystallization in supercooled magma, is about $2 \times 10^5 \text{ sec}$ and, therefore, falls within this range. Thus, in thin dikes, cooling occurs before crystals can nucleate, which leads to chilling. According to our estimates, this should be true up to a width of 1 m (Figure 9a), which agrees roughly with common field observations. Another point is that it takes time, τ_2 , or about $2 \times 10^7 \text{ sec}$, to achieve equilibrium crystallization conditions with crystal sizes of about 1 mm. This implies that dikes which cool faster than τ_2 will not experience such conditions, which means higher undercoolings and hence smaller crystal sizes. According to our rough comparison, this should correspond to dikes thinner than about 10 m (Figure 9a). Note that in the 16 m wide Cleveland dike, the crystal sizes are smaller than in the 100 m wide Kigavialuk one (Figure 8), which agrees roughly with the prediction. The point is that studying dikes with widths in the range of 1–10 m means spanning different cooling histories implying different crystal sizes. Detailed investigations could therefore allow constraints on the crystallization kinetics for a range of undercoolings.

Convection cooling in thick sills and large magma chambers

As already discussed, convection is likely to be important in magma bodies which are thicker than a few tens of meters. In the following, we compare magma bodies with thicknesses ranging from 10 m to 10 km. One observation is that the Palisades sill, which is 330 m-thick (WALKER, 1940), has crystal sizes of less than 1 mm in its interior, somewhat smaller than values in larger intrusions (BRANDEIS *et al.*, 1984). Let us focus on crystallization at the bottom which is controlled by conduction (see JAUPART and BRANDEIS, 1986). We have seen that it takes about $2 \times 10^7 \text{ sec}$ to reach equilibrium crystallization conditions. Now, the characteristic time for convective cooling is (JAUPART and BRANDEIS, 1986):

$$t_v = 15D^2\kappa^{-1}\text{Ra}^{-1/3} \quad (13)$$

where Ra is the Rayleigh number defined by:

$$\text{Ra} = \frac{\rho g \alpha \Delta T D^3}{\kappa \mu} \quad (14)$$

D is the whole chamber thickness, μ is dynamic viscosity and α thermal expansion coefficient. This equation shows that the time for convective cooling is proportional to the intrusion thickness. Figure 9b shows its variation as a function of D for a viscosity of 50 Pa sec representative of basalts. Note that it is $6 \times 10^5 \text{ sec}$ for a 10 m thick sill and that it exceeds 10^8 sec for kilometer-sized chambers.

It appears, therefore, that intrusions thinner than about 300 m should cool significantly before equilibrium crystallization conditions can be reached (Figure 9b). This is like the dike case discussed above, implying higher undercoolings in the crystallization interval and hence smaller crystal sizes. One should, therefore, expect the Palisades sill to be finer grained than the large Skaergaard and Stillwater chambers, which is indeed the case. The agreement between the observations and this simple argument is not perfect: the Palisades sill lies slightly away from the boundary separating intrusions with equilibrium crystal sizes (Figure 9b). Again, the point is that making the agreement better will provide constraints on the rates of nucleation and growth. The results obtained so far are encouraging and suggest that such a study will yield meaningful information.

There is another consequence. We gave an estimate of about 10^8 sec for τ_3 , the crystallization time under equilibrium conditions. This is the typical time taken for a volume of magma to become fully crystallized in the deep interior of a large magma chamber. A useful question is whether this volume of magma experiences varying temperatures due to the cooling of the intrusion. In other words, will it contain a record of the evolution of the whole magma body? As seen in Figure 9b, times for convective cooling are similar to τ_3 for most known basic and ultrabasic complexes including the Skaergaard and the Stillwater intrusions. This conclusion suggests that crystallization is slower than convective processes which occur in the interior of large magma chambers, and hence that it is able to record their effects in solidifying rocks. This also explains in a rough way why large intrusions exhibit igneous structures which are much more complex than those from small sills and dikes.

CONCLUSION

We have derived a series of scaling laws which allow a simple analysis of the crystallization behav-

ior. For crystallization, the most important process is nucleation, whose rate can be constrained by laboratory experiments and petrological observations. Knowledge of these kinetic rates allows calculation of the characteristic crystallization time. A simple analysis is made to compare crystallization and cooling times in intrusions of various dimensions. Different regimes are defined, which lead to observable variations in the crystal size. By using numerical values for the nucleation and growth scales, we find that chill features should be frequent in meter sized dikes. The important point is that observations of crystal sizes and morphologies in intrusions of various dimensions can be used to constrain the crystallization time, and hence the crystallization kinetics. Furthermore, the analysis allows for the treatment of different viscosity values. Hence, using data from dikes of varying chemical compositions could also prove useful.

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