NUCLEATION, CRYSTAL GROWTH AND THE THERMAL REGIME OF COOLING MAGMAS

Geneviève Brandeis, Claude Jaupart, and Claude J. Allègre

Laboratoire de Géochimie et Cosmochimie, Institut de Physique du Globe et Département des Sciences de la Terre, Université Paris

Abstract. Crystallization at the margin of a quiet cooling magma has been studied numerically, taking into account the kinetics of crystallization. The variables are the latent heat value, the growth and nucleation functions, the initial magma temperature, and the thermal contrast between magma and country rock. We have investigated a wide range of values for these parameters corresponding to natural conditions. We show that after a highly transient stage, crystallization tends toward an equilibrium between heat production (latent heat release) and heat loss. Given the small diffusivity of country rocks, latent heat release is the main factor controlling the temperature evolution. In order to minimize the latent heat release, crystallization occurs at a temperature where nucleation is small. This can be close to either the liquidus or the solidus, depending on the initial conditions. The main process controlling crystallization is nucleation and not crystal growth. Nucleation occurs as a series of sharp pulses followed by longer periods of crystal growth. The nucleation pulses give birth to thermal oscillations. These oscillations can be sustained if the interior magma temperature is above the liquidus independently of the heat loss mechanism. We show that the phenomenon occurs on the scale of a few centimeters which corresponds to the inch-scale layering of many ultrabasic complexes. The model allows us to calculate crystal sizes which are in good agreement with geological observations. The crucial parameters which determine crystal size variations near the margins of igneous bodies are the initial thermal conditions as well as the nucleation and growth functions. In the main cooling regime close to the liquidus, significant size variations can be created by small thermal disturbances.

1. Introduction

The dynamic crystallization of magma bodies involves both thermal and chemical transport phenomena, because the rate of crystallization depends on temperature as well as composition. Studies of phase equilibria in silicate systems have been numerous since the pioneering work of Bowen [1928], but have usually been independent of the cooling history. Recently, cooling models for magma chambers have been proposed [Jaeger, 1968; Usselman and Hodge, 1978; Spera, 1980]. These assume that crystallization occurs at a fixed melting point and probably give reasonable thermal predictions for large times, but are helpless for understanding the fine structures which are observed in igneous rocks. These struc-

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Paper number 4B1067 0148-0227/84/004B-1067\$05.00. tures must depend in one way or another on local crystallization conditions.

The first attempts to study dynamic crystallization have been tackled from an experimental point of view. Since the first experiments of Fouqué and Michel-Levy [1882], systematic investigations have been made on nucleation and crystal growth rates [Winkler, 1947; Klein and Uhlman, 1974; Kirkpatrick et al., 1976; Swanson, 1977; Fenn, 1977] and on rock textures [Jahns and Burnham, 1958; Lofgren, 1971, 1974, 1980; Donaldson, 1976, 1977]. However, these experiments are usually carried out at constant cooling rate, or at constant temperature, which makes it difficult to extrapolate them to natural conditions which are essentially transient and often out of the range of the laboratory.

Thus it is difficult at present to integrate structural and petrological observations in a well-founded physical framework. For example, the spectacular rhythmic layered structures which are observed on a variety of scales in fossil magma chambers [Wager and Brown, 1968] have been interpreted in several different ways Maaløe, 1978; McBirney and Noyes, 1979; Irvine, 1980, 1982; Huppert and Sparks, 1980; Chen and Turner, 1980]. These interpretations often rely on an assumed thermal regime. There is clearly a need for a sound understanding of the thermal evolution of magmas in the intermediate range spanning the time needed for the growth of one crystal to that needed for the crystallization of a whole layer. It is the purpose of this paper to fill this gap. Kirkpatrick [1976] investigated various models of crystallization, but his calculations do not exhibit the importance of latent heat release as he made a small but significant error (Appendix C). Dowty [1980] showed that latent heat release is the determining factor but did not take into account the coupling with heat flow through country rocks. We pursue these studies in more realistic conditions.

The crystallization regime of a magma is determined by latent heat release, heat transport in the magma, and heat removal through country rocks. The corresponding boundary conditions are complex and probably variable spatially Spera et al., 1982]. In a magma chamber, crystallization occurs on all sides, along vertical or sloping walls, and at the top and bottom. In a large aspect ratio chamber, most crystallization takes place in the bottom boundary layer [Huppert and Sparks, 1980] where conditions are much simpler. There is probably no hydrothermal convection in the country rocks below since they are heated from above. Also, there is probably no convection in the magma close to the crystallizing layer. Recent fluid dynamical experiments have indicated the presence of a stagnant layer isolated from convective stirring at the bottom of magma chambers [Jaupart et al., 1984]. Thus it is likely that crystallization in the bottom boundary layer

is controlled solely by conduction. We present numerical calculations based on this assumption. Our aim is to show how nucleation and crystal growth adjust to heat loss conditions. Our calculations represent a first step for other cases with more complex transport phenomena. We try to generalize our results using simpler equations and the insight gained from our numerical experiments.

We present a set of numerical experiments in which the role of the main parameters, the nucleation and the growth rates, are investigated systematically. We follow the temperature evolution of magma and calculate crystal sizes. We compare our results with petrological observations.

2. The Kinetics of Crystallization of Magmas

2.1. Nucleation Rate (I)

Nucleation is classified as either homogeneous, if it arises through random thermal fluctuations in the liquid, or heterogeneous if the presence of another phase facilitates the process. For magmas, most nucleation occurs heterogeneously on preexisting grains Kirkpatrick, 1977; Dowty, 1980]. The factors which affect the nucleation rate are the same as for homogeneous nucleation, for which the theory is well known. The major difference is the critical interval δT [Dowty, 1980] over which the nucleation rate is negligible. The &T is larger in the case of homogeneous nucleation. Using general expressions of homogeneous nucleation theory and parameters adequate for heterogeneous nucleation, it is possible to obtain suitable expressions for the nucleation rate (I) in the general form [Turnbull and Fischer, 1949; Turnbull, 1950, 1952]

I = K exp(
$$-\Delta G_a/RT$$
) exp($-\Delta H_{tr}/RT$) (1)

where ΔH_{tr} is the transport enthalpy, ΔG_a the critical nucleus free energy, T the absolute temperature. The pre-exponential term depends on the geometry of the system and on temperature. A good approximation for it is [Turnbull and Fischer, 1949]



Fig. 1. Two curves of nucleation rate (I) versus undercooling Θ , depending on parameters K_1 , K_2 , and K_3 (see text). The critical nucleation delay δT is small, corresponding to heterogeneous nucleation.



Fig. 2. Three curves of growth rate (Y) versus undercooling Θ , depending on parameters K_4 , K_5 , and K_6 (see text).

$$K = K_{\perp} T$$
 (2)

where K₁ is a constant. Dowty [1980] uses a different expression, but the overall shape of the nucleation function is not changed significantly. Calculations done with both types of functions yield essentially the same results. ΔG_a is the energy barrier for the formation of nuclei of critical size, and can be written as [Turnbull and Fischer, 1949]

$$\Delta G_a = K_2 R / (T_L - T)^2$$
(3)

where T_L is the liquidus temperature. K_2 is considered as constant in a first approximation. For a given magma, (I) is thus a function of the single variable undercooling $\odot = T_L - T$. Typical examples are given in Figure 1.

Parameters K₁, K₂, and K₃= Δ H_{tr}/R determine, respectively, the maximum rate of nucleation I_{max}, the critical δ T of nucleation, and the interval of temperature $\epsilon_{\rm I}$ over which nucleation occurs. Estimates of I_{max} are obtained in Appendix A. K₂ depends on various physical parameters which have been discussed in detail by Dowty [1980] and takes values from 10⁵ to 10¹³ K³. These determine δ T which is always found to be greater than 5°C. We consider the lowest values (between 5° and 10°C), which correspond to heterogeneous nucleation. Donaldson [1979] has shown experimentally that δ T is smaller than 13°C for olivine crystals in basaltic melts. Finally, K₃ is such that $\epsilon_{\rm I}$ is of the order of 100°C (typical interval of crystallization). This represents an enthalpy Δ H_{tr} varying from 20 to 200 kcal/mol.

2.2. Growth Rate (Y)

Crystal growth is controlled by two factors. At the beginning of crystallization, the limiting processes are the interface reactions [Allègre et al., 1981], with a growth rate depending only on undercooling [Kirkpatrick, 1975]. Chemical diffusion becomes the limiting process at larger times [Loomis, 1982; Lasaga, 1982], but it is clear that the effect of temperature dominates in highly transient cooling conditions. Kirkpatrick [1977] has shown that growth in lava lakes is

	L x10 ⁵	ΔT ₀ ,	T ₀ -T _L	, ,	I V-	¥ -		Y	 V -	I _{max} ,	ε _Ι ,	Ymax,	εy, °C	t _{fin} *,
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TABLE 1: List of Numerical Experiments

*tfin is duration of the experiment. Here, $c_p = 1.09 \times 10^3 \text{ Jkg}^{-10} \text{ C}^{-1}$, $\kappa = 7 \times 10^{-7} \text{ m}^2/\text{s}$, and $\rho = 2.8 \times 10^3 \text{ kg/cm}^3$.

controlled by interface attachment kinetics. Our aim in this paper is to investigate the role of nucleation far from equilibrium, and we consider for simplicity, growth rates which are only temperature dependent. The growth rate (Y) is written

$$Y = K_{+} \exp(-\Delta G'/RT)$$

$$x \left[1 - \exp(-\Delta G'/RT) \right]$$
(4)

 $\Delta G'$ is the activation free energy and depends on temperature. We take it to be equal to ΔH_{tr} in a first approximation. $\Delta G''$ is the bulk chemical free energy difference between melt and crystal, and is roughly proportional to the undercooling Θ [Kirkpatrick, 1975]. Y is thus a function of the single variable Θ (Figure 2). Parameters K₄, K₅= $\Delta G''$ /R, and K₆= $\Delta G'$ /R determine, respectively, the maximum growth rate Y_{max}, the temperature at which Y is maximum, and the width of the crystallization interval ϵ_Y . We consider values of Y_{max} between 10⁻⁴ and 10⁻⁷ cm/s (see the compilation by Dowty [1980]). Parameter K₅ has little effect on the growth rate and is fixed at a value of 20. Parameter K₆ is taken equal to K₃.

3. Basic Equations and Boundary Conditions

At time t=0, magma is emplaced in cold country rocks. We consider the one-dimensional problem. Both magma and country rock are initially isothermal, with a temperature difference ΔT_0 . We assume that crystallization takes place in the magma only. The heat equation is written in the outer medium (country rock) and in the inner medium (magma) as follows:

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$$\partial T/\partial t = \kappa \partial^2 T/\partial z^2$$
 (5)

$$\partial T/\partial t = \kappa \partial^2 T/\partial z^2 + 1/\rho c_n \partial Q/\partial t$$
 (6)

where c_p is the isobaric heat capacity, ρ is density, and κ is thermal diffusivity. $\partial Q/\partial t$ is the heat production rate per unit volume due to crystallization and is equal to

$$\partial Q/\partial t = L_v \partial V/\partial t$$
 (7)

where L_v is the latent heat per unit volume and $\partial V/\partial t$ the volumetric rate of increase of crystals. We assume that crystallization occurs in the form of a single phase. Models for processes involving simultaneous nucleation and growth have been developed for metals by Johnson and Mehl [1939] and Avrami [1939, 1940, 1941]. These can be applied to igneous systems, although nucleation is not of the same type [Kirkpatrick, 1976]. As rates of nucleation and growth depend on temperature, the relationship is [Kirkpatrick, 1976]

$$U/\partial t = -4\pi U Y \int_{0}^{t} I(\tau)$$

$$[\int_{\tau}^{t} Y(u) du]^{2} d\tau \qquad (8)$$

where U represents the volume fraction of melt remaining uncrystallized at time t. U is simply equal to 1-V. A unit volume of crystallized material comprises N crystals, with a mean radius R*:

$$N = \int_{0}^{t} U I dt \qquad (9)$$

$$R^{\star} = (4/3\pi N)^{-1/3}$$
(10)

The boundary conditions are

z > 0 t = 0 $T = T_0$ (11)

z < 0 t = 0 $T = T_0 - \Delta T_0$ (12)

$$z = +\infty$$
 $t > 0$ $T = T_0$ (13)

 $z = -\infty \qquad t > 0 \qquad T = T_0 - \Delta T_0 \quad (14)$

where T_0 is the initial magma temperature. It is more convenient to think in terms of dimensionless undercooling defined by

$$\Theta^* = (T_{L} - T) / \Delta T_0$$
 (15)

This system of equations has been written in dimensionless form and solved numerically. Details about the method are given in Appendix B. Numerical experiments have been performed for a wide range of parameters (Table 1). Parameters κ , ρ , and c_p have been kept constant because they vary little for magmas (their values can be found in Table 1). A general discussion with dimensionless variables is made in section 7.

4. Thermal Evolution During Crystallization

All experiments show similar effects, and we describe here one typical experiment. We follow the evolution of the temperature and of the fraction crystallized at different depths in the magma.

4.1. Experiment 6

In experiment 6, magma is intruded at the liquidus in country rocks which are colder by



Fig. 3. Evolution of the dimensionless undercooling as a function of time at various distances from the margin (full line). Note the change of scale at $t=2.5 \times 10^4$ s. The solution without latent heat release is represented by dotted lines. Large dots mark the beginning of crystallization.



Fig. 4. Plot of the number of crystals versus time. Depths are the same as in Figure 3. Large dots and squares indicate the beginning and end of crystallization. Note that nucleation occurs as a short pulse. Crystallization then proceeds by growing the existing crystals, and the number of crystals remains constant. At depth z=12.4 cm, nucleation does not start until a later time (τ_2) .

100°C. Magma cools (Figure 3), but crystallization does not start immediately because a finite length of time is necessary for the formation of nuclei and their growth to a significant size. Time τ_1 (Figure 4) marks the onset of significant crystallization. Before τ_1 , undercooling is high and nucleation rates are high. The number of nuclei produced is therefore large and the final crystal size is small. At time τ_1 , latent heat release is important, temperature increases markedly, and the production of nuclei is stopped (Figure 4). Crystallization proceeds by growing the existing crystals. This is an important feature of disequilibrium cooling experiments.

The first thermal shock does not affect the fluid over a large distance (less than 12 cm, Figure 4). As crystals of the first generation grow to their final size, latent heat release becomes less important and temperature decreases again. This allows crystallization to proceed. A second batch of nuclei is generated at time τ_2 (Figure 4), with fewer crystals because temperature is closer to the liquidus. The final crystal size will therefore be larger than near the contact. Temperature rises again, which again stops the nucleation, and the scheme is repeated again.

4.2. Thermal Conditions During Crystallization

The progress of crystallization can be followed by the movement of the crystallization front, which is at depth z where V is equal to 99%. Except during the initial period which follows magma emplacement, crystallization occurs at decreasing undercoolings close to the liquidus. This general decrease (Figure 7) is such that nucleation slows down. There is a tendency to achieve an equilibrium between crystal production and growth on the one hand, and heat loss through country rocks on the other hand. The temperature profile tends to a roughly constant gradient





Fig. 5. Dimensionless undercooling versus depth at various times for experiment 2. Numbers along the curves are times in units of 10^4 s. The margin is at z=0 (vertical line). Note that the country rock (z<0) is reheated. Note also that the curves tend to a more or less constant gradient which is quite large (a few ^OC/cm).

which corresponds to a constant rate of heat loss by conduction (Figure 5). Note that, except for slight effects at the beginning of crystallization (Figure 5a), the temperature gradient is always negative, i.e., corresponding to heat being transported away from the magma. This shows that latent heat release does not act as a heat source into the uncrystallized magma.

Another important parameter is the crystallization interval which we define as the region where V is between 1% and 99%. We find that its thickness is very small, of a few centimeters. Latent heat release increases with the thickness of the crystallization interval, which must be limited.

Thermal instabilities occur during the whole crystallization sequence. We have verified that they do not have a numerical origin (Appendix B). The first instability has been explained above in detail (at time τ_1). In the cooling conditions envisioned here, crystallization thus proceeds by the episodic nucleation of crystals at a temperature close to the liquidus. This is due to the inability of country rocks to lose heat at a rate equal to that of heat production by crystallization. Note that temperature gradients are large (of the order of 10 C/cm, Figure 5) corresponding to high heat flows of about 10^5mW/m^2 out of the magma. These high heat flows even allow the contact zone to stay at the solidus, as explained below. Conduction through the crystallized pile thus occurs with rates of heat loss comparable to hydrothermal conditions [see Spera et al., 1982 .

5. Parametric Study

5.1. The Influence of "Internal" Parameters: Y, I, L

In all these experiments, the initial temperature is equal to the liquidus.

The influence of the growth rate is mainly due to parameter Y_{max} , (experiments 1-4; for more details, see Brandeis [1983]). At low Y_{max} , τ_1 is large, and a large magma thickness is undercooled by a significant amount. Crystallization starts at temperatures far from the liquidus, where nucleation rates are high. This leads to a fine-grained matrix. On the contrary, if Ymax is large, τ_{\downarrow} is small and temperatures in the whole magma are quickly brought back to the liquidus. The first nucleation episode does not occur over a large distance. Because nucleation occurs closer to the liquidus, the mean crystal size increases more quickly with distance. This also shows that crystal sizes will be greater than at small growth rates.

In a second set of experiments (6-8, Table 1), we changed the value of the nucleation rate I_{max} . R[#] is smaller for large I_{max} near the contact but increases very quickly afterward and becomes even slightly greater than for small I_{max} (Figure 6). This shows again that the magma cannot be in equilibrium in a state where nucleation is high.

In a third set of experiments (6,9,10; Table 1), the only varying parameter is the nucleation delay δT . Above all, this parameter determines



Fig. 6. Mean radius as a function of distance for different values of I_{max} (experiments 6-8).

the equilibrium conditions of crystallization, which are such that the nucleation rate is small, i.e., at a temperature very close to $T_L - \delta T$. The latent heat was varied between 2×10^5 and

The latent heat was varied between $2x10^{\circ}$ and $8x10^{5}$ J/kg [Bottinga and Allègre, 1978]. This represents less than 1 order of magnitude variation, and the effect is not very important.

5.2. Cooling Conditions

All the experiments have been performed with the same growth and nucleation functions.

In a first series of experiments (6,11,12, 13,22), the only variable parameter was ΔT_0 . One of our aims was to compare the crystallization history of magma bodies intruded at different depths. T_0 is equal to the liquidus. At large ΔT_0 , temperature oscillations are quickly damped, because undercoolings are generally high and a new nucleation step does not require a large temperature decrease. Note that the undercooling at the crystallization front depends weakly on ΔT_0 (varying by only 10°C when ΔT_0 goes from 50°C to 800°C, Figure 7).

Variations of ΔT_0 imply differences in the crystal size evolution away from the margin: the variation is less rapid if ΔT_0 is large and the crystal size decreases with increasing ΔTO (Figure 8). For large ΔT_0 (experiments 13 and 22), temperatures near the contact are lower than the solidus, and crystallization starts farther away. Because a significant temperature gradient is maintained (Figure 5), there is a thin zone near the contact where temperatures may remain close to the solidus (Figures 8 and 9). When temperatures are brought down to solidus values, they pass through the nucleation and growth maxima. The ensuing release of latent heat depends on how much time is spent around the maxima. If temperature decreases fast enough,

there is not enough nucleation, the crystallization front moves away at a fast rate, and the zone stays cold. Thus for large enough values of ΔT_0 , there will be a subsolidus zone which remains uncrystallized and an adjacent one which crystallizes around the solidus. This corresponds well to the centimeter-sized aphanitic layers observed near the boundaries of dikes [Gray, 1970]. The thickness of this zone increases with ΔT_0 (Figure 8).

In a second set of experiments (18,19), T₀ is greater than the liquidus by 100°C. Temperatures are quickly brought back to the liquidus and oscillate very quickly with only slight damping (Figure 10). The interior temperature is above the liquidus; hence there is a significant thermal gradient into the crystallizing layer. This prevents widespread simultaneous nucleation. The thickness of the crystallizing layer is smaller than in the preceding cases (0.5-1.5 cm instead of 3-4 cm). Each nucleation step thus occurs over a small distance and in a small amount of time. This results in rapid and highamplitude temperature oscillations because the smoothing effect of heat conduction is ineffective.

6. Discussion

6.1. Limitations of the Model

We have investigated many different nucleation functions with varying degrees of steepness around the liquidus, and the same behavior has always been observed. In all cases, I is taken to



Fig. 7. Evolution of the undercooling at the crystallization front for different values of ΔT_0 . T_0 is at the liquidus.



Fig. 8. Mean radius as a function of distance to the contact for different values ΔT_0 . The arrows indicate the thickness of the zone which crystallizes at subsolidus temperatures. Note that this thickness increases with ΔT_0 . Note also that the mean crystal size close to the contact does not depend on ΔT_0 .

be zero at the liquidus. This may not be true at very small undercoolings where the initial number of sites (heterogeneities) plays an important role which is unfortunately impossible to evaluate. Experimental and theoretical work at low undercoolings is thus required to improve our understanding of dynamic crystallization. These limitations do not affect our main results. We have shown that temperature tends toward an equilibrium between heat loss and heat production, at a small and roughly constant undercooling. This will hold for any model of heterogeneous nucleation. Thus the nucleation rate is approximately constant, which leads to homogeneous grain sizes in the interior of magma bodies, in good agreement with observations (Table 2).

We have neglected chemical diffusion, but we have shown that nucleation is the controlling factor of crystallization. At a given depth, nucleation essentially occurs only once, and crystallization proceeds by growing the existing nuclei. The exact mechanism of crystal growth is



Fig. 9. Plot of the fraction crystallized versus time at various depths for experiment 13. Note the much smaller rate of crystallization at depth z=4.9 cm. There, crystallization proceeds close to the solidus.

thus less important and does not limit the validity of our results.

Finally, we have treated a limited class of thermal models. As stressed before, we have always observed the same qualitative behavior and we feel that our catalog of solutions is extensive enough to study most cases. Only the quantitative estimates are likely to differ. Consider, for example, that a heat flux is imposed at the crystallization front, which would correspond to the roof of a magma chamber where there is convection. This can be understood using our results for initial magma temperatures above the liquidus. The resulting thermal structure is similar, with a temperature gradient into the



Fig. 10. Evolution of the undercooling at the crystallization front, when the initial magma temperature is above the liquidus. Numbers along the curves are values of ΔT_0 .

Dike	Thickness, m	Main Minerals	Size, mm	Distance to Contact, m	Reference	
Cleveland	16	plagioclase	0.055 - 0.11	0 - 8	Winkler [1949]	
Grenville	60	plagioclase clinopyroxene	1.2 - 3.5 1 - 3	1 - 13 1 - 13	Gray [1970]	
Kigaviarluk	106	plagioclase clinopyroxene	0.8 - 2.8 1.2 - 6	0.3 - 22 0.3 - 22	Gray [1970]	
Palisade	300	plagioclase olivine clinopyroxene	0.5 - 1.5 0.15 - 0.25 0.01 - 60	0 - 300 0 - 25 ?	Walker [1940]	

TABLE	2a.	Crystal	Sizes	in	Igneous	Bodies:	Dikes	and	Sills
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crystallizing layer. The consequence is a slower cooling rate (compare Figures 7 and 10) because country rocks must lose both the heat produced by crystallization and the imposed heat flux. There is also the possibility that hydrothermal convection is operating, at least at the top of a magma chamber. We show in section 7 that this type of heat loss results in the same general behavior.

6.2. Comparison With Previous Models

Dowty [1980] already attempted to calculate crystal sizes. Assuming a constant rate of volumetric heat loss, he showed that once a certain number of nuclei has appeared, temperature is brought back to the liquidus. Our results complement his, specifying in particular the thickness of the crystallization interval and the heat flow values reached naturally by conduction cooling.

Our model predicts that crystallization tends to occur at an equilibrium temperature below the liquidus. In a sense, this justifies the assumption of Jaeger [1968] that magma behaves as an ideal body, i.e., that latent heat is released at a fixed melting point. According to his model, the crystallization front position is given by a power law λ/t , where λ is the solution of an implicit equation. Consider, for example, experiment 13. Using the same latent heat value, we have solved Jaeger's equation for that same value of the initial temperature contrast and for a phase change occurring at an undercooling of 10° C, which corresponds to the equilibrium value at the crystallization front (Figure 7). The agreement with our numerical results is good, but the slopes are slightly different (Figure 11). The numerical model predicts somewhat smaller crystallization rates at large times because the

Intrusion	Height, m	Main Minerals	Mean size, mm	Distance From Base of Intrusion to Measure- ment Site, m	Reference
Skaergaard	3000	plagioclase pyroxene oxides	1 - 2 0.6 - 1 1 - 2	1600 (UZa) 1600 (UZa) 1600 (UZa)	McBirney and Noyes [1979]
		olivine	0.05 - 3	-100 (hidden zone)	Maaløe [1978]
Stillwater	5000	olivine bronzite chromite bronzite chilled marginal gabbro	1 - 4 1 - 4 0.1 - 0.4 0.7 - 1 0.3	ultramafic zone 300 - 1100 0 - 300 contact	Jackson [1961]
Rhum	1200	olivine plagioclase	1 1	?	Brown [1956]
Eatern Bushweld	7500	bronzite olivine	1 1 - 3	1200 - 2300 1200 - 2300	Cameron [1975]

TABLE 2b. Crystal Sizes in Igneous Bodies: Major basic intrusions

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Intrusion	Main Minerals	Mean Size, mm	Observations	Referenc	ce
Duke Island	olivine pyroxene	0.2 - 2 4 - 10	size-graded layer 5-25 cm thick	Irvine	[1963]
Great Dyke	olivine - chromite	1 - 3	chromitite layer	Wilson	[1982]
Klokken Complex	felspars pyroxene	1.5 5	reverse modally graded layer	Parsons	[1979]
Imilik	plagioclase pyroxene olivine	0.25 - 2 0.25 - 2 0.25 - 4	size-graded layer 1 m thick	Brown and Farmer	[1971]
Nunnarsuit	felspars olivine pyroxene	2.5 1 1	normal modally graded layer	Parsons and Butterfield	[1981]
Ilimaussaq	felspars nepheline pyroxene	5 2 - 5 5	little size grading	Sörensen	[1968]
Chebucto Head	quartz plagioclase biotite K-felspar all of the same size all of the same size	2 - 4 2 - 5 2 - 3 1 - 3 1 0.5 - 4	reverse size- graded layer 30 cm thick microgranite layer leucogranite layer	Smith	[1975]
Southern Greenland	plagioclase biotite pyroxene	3 - 6 1 - 2 3		Emeleus	[1963]
Jimberlana	bronzite olivine chromite	$ \begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	(contact)	Campbell	[1977]
Ploumanac'h	biotite quartz oligoclase	2 - 3 1 - 8 3 - 7	reversed size- graded layer	Barțière	[1981]

TABLE 2c. Crystal Sizes in Igneous Bodies: Other Intrusions

crystallization temperatures and thicknesses are still changing. We stress that the equivalent phase-change temperature is not equal to the liquidus and cannot be determined a priori. We have shown that it depends strongly on the initial conditions. In the case of low ΔT_0 , the evolution of the crystallization front does not follow a power law (Figure 11) because the crystallization interval is of large thickness, which does not correspond to Jaeger's assumptions.

7. Simplified Formulation and Qualitative Behavior of the Solutions

Thermal oscillations result from the interaction of two mechanisms, heat loss and latent heat release, which are coupled through temperature. To show the essential physical aspects and the critical parameters of the problem, we now propose a simpler system of equations which has the same behavior as our numerical solutions. We take a lumped approach for the mean temperature in the crystallizing layer:

$$\frac{dT}{dt} = -\beta (T-T_m) + \frac{L}{c_p} G \qquad (16)$$

Heat loss is modeled as the exchange between magma at temperature T and country rocks at temperature T_m , β characterizes its intensity, and G is the growth rate of the crystallized fraction (dV/dt). Because the relation-ship between temperature and nucleation and growth rates is non linear, there is no simple form for G as a function of T. Because of its finite thickness, the



Fig. 11. Position of the crystallization front for various values of ΔT_0 . Dashed curves represent the solution of Jaeger [1968] for a fixed phase-change temperature θ_0 .

layer responds to a temperature change with some delay τ . This has already been suggested by Samoylovitch [1979]. G is thus the solution of the following differential equation:

$$\tau \, dG/dt + G = f(T) \tag{17}$$

Here f(T) has the form of a bell-shaped curve (Figure 12). For our present purposes, it is sufficient to consider a simple Gaussian law:

$$f(T) = A \exp \left[- (T - T_e)/b \right]^2$$
 (18)

The system is best discussed in the (T,G)phase-plane [Arrowsmith and Place, 1982], where all solutions are conveniently represented as trajectories. Sustained oscillations can be predicted under specific conditions (Figure 12). There are four types of solutions depending on the values of the parameters and on the respective positions of isoclines (dT/dt = 0 and dG/dt = 0). When the isoclines do not intersect (case 1), all trajectories tend to fixed point B (T_m, 0), which corresponds to a uniform temperature equal to that of country rocks. For solutions to be oscillatory, the isoclines must intersect. This occurs when the dimensionless variable β^* ,

$$\beta \star = \beta \frac{(T_e - T_m) c_p}{L A}$$
(19)

is smaller than 1. If condition (19) is met, there is another stable fixed point, labeled A (Figure 12). The behavior of solutions in its vicinity is described by parameter δ [Arrowsmith and Place, 1982]:

$$\delta = -\frac{df}{dT} |_{T=T_A}$$
(20)

and dimensionless variable τ^* defined by

$$\tau^* = \frac{L \,\delta \,\tau}{c_p} \tag{21}$$

Trajectories converge toward stable A, either directly (case 2), oscillating with strong damping (case 3), or oscillating with little damping (case 4, Figure 12). The four types of solutions are realized for different values of β^* and τ^* (Figure 13). Sustained oscillations (case 4) require that β^* be smaller than a limit-value β_1 and also that τ^* be neither too small nor too large.

Parameter τ measures the response of the crystallizing layer to temperature changes and is a function of its thickness: a thick layer is slow to adjust. We have seen that the thickness remains quite small, even for high rates of heat loss. The δ has a large range of variations because of the shapes of the nucleation and growth curves and is therefore a critical parameter. From (20) it is a measure of the sensitivity of nucleation and growth to temperature changes. Damping decreases with increasing δ , in agreement with our numerical experiments. Large values of δ correspond to the solutions of Figure 10 (high undercoolings).

This schematic discussion shows that three variables enter the problem: the intensity of heat loss, the thickness of the crystallization interval, and the sensitivity of crystallization rate to temperature changes. They are not independent, the first one determining the other two. Our experiments show that the thickness of the crystallization interval varies little and that higher rates of heat loss essentially lead to higher undercoolings and therefore to greater values of δ .

We now discuss the system quantitatively. We have always found values of β smaller than β_l , even for efficient hydrothermal convection. We also estimate that τ^* remains between values of 10 and 10^5 . We find therefore that crystalli-





Fig. 12. Trajectories in the (T,G) phase-plane. The units are arbitrary. The straight line is isocline dT/dt=0 where the tangents to the trajectories are vertical. The bell-shaped curve is isocline dG/dt=0 where the tangents to the orbits are horizontal. When the two isoclines intersect each other, all trajectories converge toward either fixed point B $(T_m,0)$ or fixed point A. Sustained oscillations with low damping are possible around A.



Fig. 13. Graphical representation of the four possible solutions in the (τ^*, β^*) space. In each region, the corresponding behavior is illustrated schematically (circled numbers refer to cases detailed in the text). The white inner region is where sustained oscillations occur. β_1 is the maximum value of β^* above which there is no oscillatory solution. Arrows indicate the range of parameters τ^* and β^* in geological conditions.

zation always occurs with oscillations, although the degree of damping varies (Figure 13).

8. Implications and Comparison With Petrological Observations

8.1. <u>Undercoolings and Cooling Rates in</u> Crystallizing Magmas

Magma crystallizes in two different regimes. The initial regime is highly transient with temperatures significantly below the liquidus and high cooling rates. The second regime is one of quasi-equilibrium at a low undercooling determined by the density of heterogeneous nuclei and the initial thermal conditions. The cooling rate is low. In thick dikes and magma chambers, most crystallization should occur in this regime. There is good evidence that this is indeed the case [Morse, 1979].

8.2. Inch-Scale Layering

We have shown that temperature oscillations occur on a spatial scale of a few centimeters (Figures 6,8), which is similar to that of inchscale layering [Wager and Brown, 1968]. The origin of inch-scale layering has been usually ascribed to "oscillatory nucleation" [Zy1, 1959; Wager, 1959; Vannier, 1976; Maaloe, 1978; McBirney and Noyes, 1979; Morse, 1979]. Crystallization in most basaltic chambers tends to occur at a cotectic composition, as was emphasized by Morse [1979], for example. Consider two cotectic crystal phases A and B. The crystallization of A, say, changes the liquid composition. When nucleation stops, the liquid composition is in the stability field of crystal B. Temperature decreases and, at sufficient undercooling, the nucleation of B starts. This eventually brings back the liquid composition in the stability field of A, and the cycle repeats itself. We have shown in

this paper that the phenomenon occurs for thermal reasons on the scale of a few centimeters, which is observed in the field. We have also emphasized that the most favorable conditions for sustained oscillations correspond to interior magma temperatures above the liquidus (Figure 10).

8.3. <u>Crystal Sizes in Dikes and</u> Larger Intrusions

For the whole set of models considered here, we find a total range of crystal sizes of 0.01 to 10 mm. This corresponds very well to the values observed in all types of igneous rocks (Table 2). We predict that, in general, crystal size increases away from the margin (Figures 6,8) and should eventually reach a more or less constant value when quasi-steady thermal conditions are achieved. This agrees with measurements made in dikes (Figure 14). Our calculated sizes are close to the observed ones. During dike emplacement, heat advection associated with the flow of magma is important and may change the boundary conditions. However, flow velocities are high (~1 m/s) [Einarson and Brandsdottir, 1980] and a 10 km long dike is emplaced in about 10^4 s, which is short compared to cooling times (Figure 4). Our calculations emphasize that size variations depend on the initial thermal conditions. Note that in three cases, the Kigaviarluk and Grenville Dikes and the Palisade Sill (Figure 14), the plagioclase data are in striking agreement, which indicates similar cooling conditions. Pushing the reasoning to the limit, these data suggest that the initial temperature contrast was greater than about 600°C (Figure 8), which is compatible with likely crustal conditions. We also suggest on the same grounds that the initial temperature contrast was slightly greater for the Kigaviarluk Dike than for the Grenville Dike. The Cleveland Dike is an exception both because the variation occurs over a smaller distance and



Fig. 14. Variations of crystal sizes in dikes. Sources for the data are given in Table 2. Data for the Kigaviarluk and Grenville dikes are nucleation density measurements and have been transformed into crystal sizes using relation (10). The squares represent results obtained with experiment 13.

because the interior sizes are smaller. This points to a significantly higher thermal contrast. Together with the small thickness of the dike, this implies higher undercoolings and therefore smaller crystals. This discussion illustrates the interest of crystal size measurements.

Consider now the case of large magma chambers. Although there is little doubt that some form of convection operates in such large objects [Bartlett, 1969], Jaupart et al. [1984] have shown that the bottom crystallization occurs in a stagnant boundary layer isolated from convective stirring. Replenishment of the chamber by a new influx of magma will change the thermal conditions. The available evidence suggests that replenishment is episodic [Huppert and Sparks, 1980; Condomines et al., 1982] and hence that crystallization occurs mainly in a conduction regime with only brief periods where heat advection plays a role. Thus our calculations are also valid in this context. Magma chambers exhibit complex layered structures where the crystal size is usually remarkably constant. Size homogeneity is a feature of quasi-steady crystallization conditions. In a few cases, the crystal size is greater at the bottom of a layer, which is then termed size-graded (Imilik, Duke Island). In other cases (Ploumanach, Chebucto Head), a layer can be reverse size-graded, which corresponds to "normal" cooling conditions, analogous to the dike examples of Figure 14. We have noted that relatively modest temperature disturbances around the liquidus can lead to large size variations (Figure 8). This suggests that reverse sizegraded layers imply mechanisms of sudden emplacement on a colder substratum.

Sedimentation is often put forward to explain size-graded layers. However, those are not a frequent feature of magma chambers. Our calculations show that the thickness of the crystallization interval is small (a few centimeters). This indicates that sedimentation can only occur on a small scale. Of course, this says nothing about the possibility of settling or flotation from distant crystallizing interfaces.

8.4. Pillow Lavas

Inside a pillow lava, three textural zones can be distinguished Bryan, 1972; Schiffman and Lofgren, 1982]. There is first a vitrophyric rim, usually less than 2 cm thick that may contain some phenocrysts. There is then a spherulitic zone about 3-4 cm thick with skeletal crystal growth in a glassy matrix. The core of the pillow is made of a holocrystalline zone showing intergranular textures. These three zones are usually described in terms of different cooling rates. Recent experimental work [Lofgren, 1983] indicates that differences in textures comparable to those due to varying cooling rates can be produced by varying the kind and density of nuclei. Although the cooling conditions for pillow lavas must involve heat loss in water, our calculations for very high initial temperature contrasts may be used for comparison. As shown in section 5.2., our model predicts the existence of three zones: first, an outermost one where crystallization occurs below the solidus with few crystals growing; second, an intermediate zone close to the solidus; and third, the interior where quasi-equilibrium cooling conditions are achieved. This explains that crystal size is independent of the pillow radius. Our calculated thicknesses for the two outer zones agree with the observations (Figure 9). As shown above, high cooling rates are "frozen" into the two outer zones during the initial cooling period which follows emplacement. The two zones then evolve close to the solidus with low cooling rates. Thus nucleation with high cooling rate characteristics may coexist with growth of low cooling rate characteristics.

9. Conclusion

As discussed in the text, the static nature of our assumptions (no convection or magma flow) applies more specifically to the bottom boundary layer of magma chambers or sills. However, conduction always dominates in boundary layers, and our study provides a general framework for the study of magmatic processes recorded in solidified igneous rocks. Our results are in good agreement with observations of crystal sizes and textural zones in dikes and pillow lavas. The comparison with magma chambers is qualitative because of the more complex thermal phenomena associated with the emplacement and evolution of such large reservoirs. Our model specifies the "normal" conditions of crystallization, i.e., without taking into account possible physical effects (density surges or replenishment) or chemical effects (double-diffusion). Arguments about the puzzling structures of fossil magma chambers can be made by comparison with these "normal" conditions.

Generally speaking, our experiments show that

nucleation and growth are competing against each other. Fast growth is associated with fast nucleation and thus does not lead to large crystal sizes. We have shown that size variations tell us a lot about the thermal conditions prevailing before the onset of crystallization. Further, in the main cooling regime at small undercoolings, if mechanical disturbances such as density surges are associated with slight thermal transients, they will result in measurable crystal size variations. This raises the hope that size measurements may be used in a systematic fashion to investigate the thermal aspects of magma cooling and differentiation.

Following Dowty [1980], we emphasize that latent heat is one of the main controlling factors for the thermal evolution of cooling magmas. Crystallization is achieved through a series of nucleation steps followed by periods of crystal growth. Each step occurs over a few centimeters and is associated with a temperature jump of several degrees. This oscillatory behavior must be compared to the phenomenon of inchscale layering. In all cases, the crystallization interval is of small thickness, which implies that crystal settling occurs on a small scale.

Appendix A: Estimations of the Nucleation Rate

Some measurements of the nucleation density N have been performed over a large range of temperatures at constant undercooling [Fenn, 1977; Swanson, 1977]. We propose here a method for calculating the nucleation rate (I). In the experiments, nucleation and growth rates are constant and equation (8) can be integrated easily:

$$U(t) = \exp(-\pi/3 IY^3 t^4)$$
 (A1)

The density N is then :

$$N = \int_{0}^{t^{*}} I U(t) dt \qquad (A2)$$

where t^* is the duration of the experiment. If it is large enough, N is equal to

$$N^{\infty} = I \int_{0}^{\infty} \exp(-\frac{\pi}{3} IY^{3}t^{4}) dt$$
$$= \frac{\Gamma(5/4) I}{\frac{4}{2}\pi} \simeq \frac{I}{\frac{4}{2}\pi}$$
(A3)

where α is equal to $(\pi/3)$ IY³. This equation holds under the following condition:

$$\alpha t^{*4} > 1 \qquad (A4)$$

Equation (A3) is rewritten as

$$I = N^{4/3} Y x (\pi/3)^{1/3}$$
(A5)

Knowing I from (A5), we calculate α to verify condition (A4). High crystal growth rates are requested as α varies with Y³, and the method has been applied to Fenn's [1977] measurements. Fenn gives curves of nucleation density versus undercooling, which exhibit a maximum. We calculate the maximum nucleation rate at this value. In some experiments the curves have no maximum, and we have taken the data at the largest undercooling. Condition (A4) was always verified. We find that I_{max} is between 10⁻¹ and 10⁻² cm⁻³s⁻¹ for alkaline feldspar crystals (Table Ala).

When condition (A5) is not met, it is always possible to find a minimum value for the nucleation rate by expanding integral (A2). A minimum value for I is thus simply

$$I = N / t^*$$
 (A6)

This second method was applied to Swanson's [1977] experiments on the orthoclase-albiteanorthite-quartz system, as growth rates are small ($\simeq 10^{-9}$ cm/s). Considering (A6), I_{max} is found to be between 10^1 and 10^3 cm⁻³s⁻¹. These values are higher than those deduced from Fenn, because the initial melt composition is drastically different.

The two sets of experimental data give a reasonable range of values. There are a few direct estimations of nucleation rates in less well-

 $I = N^{4} / {}^{3}Y$ N, ст-³ Melt Y. ΔT, °C cm/s (A5) Composition 5x10⁻⁷ 5.4×10^{-2} Ab₉₀Or₁₀+2.7%H₂O 6x10³ 400* 2x10³ 5x10⁻⁶ 1.3×10^{-1} Ab₉₀Or₁₀+5.4%H₂O 200 2.5X10-6 6.3×10^{-2} 2x10³ Ab₉₀Or₁₀+9.0%H₂O 130 10⁻⁶ 3.4×10^{-2} 2.5x10³ Ab_{70} Or $_{30}$ + 1.7% H $_{20}$ 300* 4x10³ 2.5x10⁻⁶ 1.6×10^{-1} Ab / 00r 30+4.3%H 20 210 10³ 2x10⁻⁶ 2×10^{-2} Ab₇₀Or₃₀+9.5%H₂O 110 2.5×10^{-1} 4x10³ 4x10⁻⁶ $Ab_{50}Or_{50}+2.8\%H_{2}O$ 350* 4x10⁻⁶ 8x10² 3x10⁻² Ab₅₀Or₅₀+6.2%H₂O 170

TABLE Ala. Maximum Nucleation Rate Calculations of Alkaline Feldspar Crystals From Density Meaturements [Fenn, 1977]

*No maximum nucleation density in the experiments. We took the data at maximum undercooling.

(Tratal	Walt	۸ . ۳	N	v	I=N/t* (A6)		
Crystar	Composition	°C	cm ⁻³	cm/s	24h	144h	
Quartz	Gr + 3.5% H ₂ O	200	10 ⁸	2x10 ⁻⁹	10 ³	2x10 ²	
	Gr + 6.5% H ₂ O	450*	10 ⁸	2x10 ⁻⁹	10 ³	2x10 ²	
	Gr + 12% H ₂ O	400	10 ⁸	10 ⁻⁸	10 ³	2x10 ²	
Alkali Fs	$Gr + 3.5\% H_2O$	200	10 ⁷	10^{-7}	10 ²	2x10	
	$Gr + 6.5\% H_2O$	450*	10 ⁸	10^{-8}	10 ³	2x10 ²	
	$Gr + 12\% H_2O$	280	10 ⁸	$3x10^{-9}$	10 ³	2x10 ²	
Plagioclase	Gr + 3.5% H ₂ O	350	10 ⁸	10 ⁻⁹	10 ²	2x10	
	Gr + 6.5% H ₂ O	300*	5x10 ⁷	5x10 ⁻⁸	10 ³	2x10 ²	
	Gr + 12% H ₂ O	300	10 ⁸	3x10 ⁻⁹	10 ³	2x10 ²	

TABLE Alb. Maximum Nucleation Rate Calculations From Density Measurements [Swanson, 1977]

*No maximum nucleation density in the experiments. We took the data at maximum undercooling.

controlled conditions. Kirkpatrick [1977] made some measurement on plagioclase crystals in basalts from Hawaiian lava lakes. He found values ranging from 7×10^{-3} to $2 \text{ cm}^{-3} \text{s}^{-1}$ for small under-coolings. Winkler [1947] obtained a value of $10^{-3} \text{cm}^{-3} \text{s}^{-1}$ for nepheline crystals.

Appendix B: Numerical Method and Convergence

The system of equations (5-8) and (11-14), with a nonlinear parabolic partial differential equation, was investigated by Crank and Nicolson [1947] using an implicit method. They treated the case of heat generation by a chemical reaction whose rate depends on temperature. We used a second-order implicit method to solve the heat equation with a truncation error of $O[(\Delta z)^2 + (\kappa \Delta t)^2]$. We transform equation (8):

$$\partial LnU/\partial t = f(\Theta)$$
 (B1)

This ensures that U remains between 0 and 1. Crank and Nicolson [1947] approximated (B2) with

. .

$$\frac{\operatorname{Ln} U_{1}^{n+1} - \operatorname{Ln} U_{1}^{n}}{\Delta t} = \frac{1}{2} \left[f(\Theta_{1}^{n+1}) + f(\Theta_{1}^{n}) \right] \quad (B2)$$

This has a truncation error of order $O[(\kappa \Delta t)^2]$. The calculation of temperatures at time t+ Δt requires several iterations with a relaxation method [Shaw, 1953]. We chose instead to write

$$\frac{\operatorname{Ln} \mathbf{U}_{\mathbf{i}}^{n+1} - \operatorname{Ln} \mathbf{U}_{\mathbf{i}}^{n}}{\Delta t} = \mathbf{f} (\Theta_{\mathbf{i}}^{n})$$
(B3)

which has a truncation error of order $O(\kappa \Delta t)$.

The reason for doing so is that the truncation error of the heat equation is $O[(\kappa \Delta t)^2 + (\Delta z)^2]$. Implicit finite difference schemes are stable for all values of ratio $\lambda = \Delta t / (\Delta z)^2$ and, in particular, for $\lambda = 0.5$, when $(\Delta z)^2$ is of the same order as $\kappa \Delta t$. Therefore the simpler form (B3) is accurate enough and leads to a much simplified system [Ames, 1965].

The convergence in Δz is the most difficult to obtain because of the high value of the latent heat term. Space steps vary from 0.25 to 0.0625 cm. Once convergence in space is achieved, convergence in time is always assured. We used mesh ratios such that $\lambda < 4$, which imply time steps smaller than 10 s. The convergence has been checked for each run, particularly in the case of rapid oscillations (experiments 18 and 19). Each oscillation occurs over more than 100 time steps and is not of numerical origin.

Appendix C: Comparison With the Work of Kirkpatrick [1976]

The initial condition in Kirkpatrick $\begin{bmatrix} 1976 \end{bmatrix}$ is different from ours, as the country rock is assumed to be initially preheated with a temperature gradient equal to $\theta = -8/3z$ (z in centime-



Fig. 15. Comparison between Kirkpatrick's calculations (thin lines) and ours (heavy lines). Dashed lines represent the solution without latent heat release, which has been calculated both analytically and numerically. Note that it is almost identical to Kirkpatrick's solution.

ters). We adapted our model in order to treat the same condition, and the same growth and nucleation rates. We verified that our numerical solution of the equation without latent heat release is identical to the analytical solution [Carslaw and Jaeger, 1959]:

$$\Theta(z) = -0,5zA \operatorname{erfc}(z/2/\kappa t)$$

-A $\sqrt{(\kappa t/\pi)} \exp(-z^2/4\kappa t)$ (A=-8/3) (C1)

Kirkpatrick's solution is hardly different from (Cl), whereas ours branches off at the onset of the crystallization (Figure 15). Moreover, the crystallization front does not move at the same rate: at t= 1.5×10^{5} s, the front only reaches z=60 cm, whereas Kirkpatrick finds it reaches z=60 cm.

This discrepancy is due to an error in Kirkpatrick's equations. Equation (7) had been simplified by setting:

$$dQ/dt = \partial Q/\partial T \times \partial T/\partial t$$
 (C2)

This is not correct because Q is function of both T and t. The proper equation is

$$dQ/dt = \partial Q/\partial T \times \partial T/\partial t + \partial Q/\partial t|_T$$
 (C3)

This second term is the largest. Kirkpatrick's solution amounts, in fact, to ignoring latent heat in the temperature equation (see Figure 15).

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C.J. Allègre, G. Brandeis, and C. Jaupart, Laboratoire de Géochimie et Cosmochimie, Institut de Physique du Globe et Département des Sciences de la Terre, Université Paris 6 et 7, 4, Place Jussieu 75230 Paris Cedex 05 France.

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