LABORATORY EXPERIMENTS WITH AQUEOUS SOLUTIONS MODELLING MAGMA CHAMBER PROCESSES. I. DISCUSSION OF THEIR VALIDITY AND GEOLOGICAL APPLICATION

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ABSTRACT. Experiments with aqueous solutions have been widely used to model magma chamber processes and the origins of layering in igneous rocks. Such experiments have the advantage that they are simple to carry out in a laboratory and they allow a fundamental understanding to be developed on the principles that control solidification and convection in crystallizing fluid systems. They provide data that can be used to assess general physical theories. These theories can then be applied to geological situations. Dynamic similarity between laboratory experiments and natural systems is an important consideration. However, it is not necessary for an experiment to match nature exactly in complexity or even values of dimensionless parameters such as Reynolds Number or Rayleigh Number in order to be informative. If the experiment and natural system are in the same fluid dynamical regime, where a particular physical law applies, then that is sufficient. Although aqueous systems readily generate fluid layering and compositional stratification, the relationship of this layering to rock layering still needs further development.

1. INTRODUCTION

Ideas on the fluid dynamical processes in magma chambers and on their relationship to the characteristics of igneous rocks have been recently influenced by laboratory experiments using aqueous solutions. These experiments have investigated the convective effects that can occur in multicomponent fluid systems where two or more properties with different diffusivities control the density of the fluid. The double-diffusive effects observed in such systems attracted the interest of geologists because of the resemblance of the fluid layering that could form to layered igneous rocks (Turner and Gustafson, 1978; McBirney and Noyes, 1979; Chen and Turner, 1980). Experiments involving crystallization from saturated aqueous solutions have proved to be of particular interest and a wide range of novel fluid dynamical phenomena have been recognised. Some of these phenomena are now thought to be relevant to the interpretation of layering in intrusions.

Since 1978 numerous laboratory investigations have been published

and the topics of double-diffusive convection and compositional convect-
ion reviewed in several recent papers (Huppert and Turner, 1981a;
Sparks et al., 1984; Huppert and Sparks, 1984; Turner, 1985). In view
of these reviews a further synopsis would not serve any useful purpose.
We concentrate on two topics which are particularly relevant to the
interpretation of layered intrusions. First, we consider to what extent
it is reasonable to extrapolate small-scale laboratory experiments with
aqueous solutions of low viscosity to large, slowly cooled, magma
chambers containing high viscosity silicate melts. Second we discuss
how the various kinds of fluid layering observed in laboratory experi-
ments might relate to the various kinds of layering observed in solid
rocks.

2.   JUSTIFICATIONS AND LIMITATIONS

The principal justification for using aqueous solutions is that they
constitute simple chemical systems that can be easily handled and
studied using inexpensive laboratory equipment at room temperature.
Aqueous solutions when combined with other water-soluble organic
compounds such as glycerine, glycols and natrosol can provide systems
with great ranges of physical properties such as density, chemical
diffusivity and viscosity. These features mean that experiments can be
undertaken over a wide range of conditions so that some of the
fundamental principles of crystal growth and associated fluid motions
can be identified and investigated. The chemical composition of many
aqueous solutions can also be determined by simple and rapid techniques
such as the measurement of refractive index and density, and flame
photometric analysis. The topology of the phase diagrams are also
widely varied, allowing investigation of many different situations of
geological interest.
     The relationships between crystallization and fluid dynamic
processes is a new scientific field. There are few references before
1978 to the role of fluid motions in crystal growth in a literature
scattered across many fields, including metallurgy, chemical engineering
and electronics. Models of crystal growth have largely concerned
molecular processes occurring at crystal faces and the surrounding fluid
has usually been considered static. Aqueous solutions are the most
practicable systems in which to discover the fundamental physical
principles involved and would be the logical way to proceed even if the
subject held no significance to geologists.
     We suggest that a close analogy exists with the early days of
experimental petrology. Progress was then made by understanding simple
binary and ternary silicate systems and discovering the fundamental
rules that govern phase equilibria. It proved to be much better to
investigate a system such as Mg_2SiO_4-SiO_2 properly than to immediately
attempt to understand a ten component natural basalt. Eutectic points,
peritectic points, continuous and discontinuous reactions are all
features of fundamental significance to all chemical systems (metals,
aqueous solutions and silicates) whether they are simple or complex. We
consider that exactly the same arguments apply to the fluid dynamics of

crystallization.   There are some aspects of the experiments with aqueous
solutions that reveal fundamental principles independent of the actual
fluid used.   One example of such a principle is that, except for
eutectic compositions, crystallization changes the composition and
density of the fluid locally and, in consequence, generally leads to
fluid motions due to the presence of density gradients.   These motions
can take place in any kind of fluid provided that appropriate driving
forces acting on the fluid are of sufficient magnitude.

A common argument against the use of aqueous solutions is that the
experiments do not scale and there is no dynamical similitude (for
example Rice, 1985).   We have frequently been faced with arguments along
the lines that "magma has a much higher viscosity than water" or "the
Lewis numbers (the ratio of thermal to chemical diffusivity) are much
larger in magmas" or "the crystal sizes in the experiments scale to a
kilometre in magma chambers".   Although there are genuine limitations to
the aqueous solution experiments, such comments do not fully take
account of the combined theoretical and experimental method used in
fluid dynamics and the significance of dimensionless numbers.

A common misconception is that experimental studies have to scale
precisely to the natural situation (i.e. that there has to be total
dynamic similarity).   Dynamical similarity is desirable in cases in
which the theoretical background is unknown or incomplete.   However, if
a fundamental physical law can be verified under one set of experimental
conditions then the law can apply under a completely different set of
conditions whether dynamical similarity holds or not.   The only necess-
ity is that the flow be in the same physical regime.   Some of the
commonly used dimensionless numbers (such as Rayleigh number and
Reynolds number) reflect ratios of forces.   The purpose of the dimension-
less numbers is to identify situations where particular forces or
effects dominate behaviour.

These concepts can be illustrated with a simple example of the fall
of a solid spherical object through a liquid.   The solid object falls
under some circumstances at a terminal velocity according to Stokes Law:

$$V = \frac{\Delta\rho\ g\ d^2}{18\mu} \qquad\qquad (1)$$

where $\Delta\rho$ is the density difference between solid and liquid, g is
gravity, d is the diameter of the sphere and μ is the dynamic viscosity
of the fluid.   The statement that Stokes Law is appropriate whenever the
viscosity of the fluid is large is, however, too superficial and may be
specious.   The correct statement is that Stokes Law is valid whenever
inertial forces are small compared to viscous resistant forces, and this
ratio is termed the Reynolds number:

$$Re = \frac{\Delta V d}{\mu} \qquad\qquad (2)$$

Stokes Law holds as long as Re is approximately less than 1.   Thus

the law is valid for a dust particle settling in air, an extremely low
viscosity fluid, but does not hold for 10 metre sized mantle xenoliths
falling through basalt magma, a fluid with high viscosity. In the
latter case the fall velocity would be determined by a physical law
appropriate for high Reynolds numbers where inertial forces dominate,
and the fall velocity would be almost independent of the liquid
viscosity. If we wished to confirm Stokes Law by experiment this could
be done by conducting experiments at Reynolds numbers of 0.1, but the
law, which is based on a theoretical balance of forces between the
gravitational force exerted by the solid on the liquid and the viscous
drag force exerted by the fluid on the solid, would hold equally well at
any small Reynolds number. Stokes Law can be, and has been, used to
predict how a solid diapir of material might move through the mantle. A
kilometre diameter object with a density of 3200 kg/m^3 would be
predicted to ascend through a mantle, with a viscosity of 10^{18} Pa.s. and
density of 3300 kg/m^3, at a velocity of around 3 x 10^{-8} m/s. The
Reynolds number would be about 10^{-18} and yet no experiments have ever
been conducted at such low values. We would assume that most geologists
would accept such calculations as providing approximate estimates of
velocities of mantle diapirs, yet experiments used to confirm Stokes Law
typically have Re numbers 10^{15} times greater than this natural
situation. The reason that such calculations are valid is that Stokes
Law is an example of a fundamental physical law reflecting the balance
of forces, which has been confirmed by numerous experiments. These
experiments do not have to show dynamic similarity, but merely have to
have Re < 1.

     Another hidden aspect of Stokes Law in its simplest form is that no
other forces or processes influence settling. Presumably a falling
object could be effected by all sorts of other effects. The Coriolis
force must be present, yet in geological situations it is so weak that it
can be neglected. If the falling object is a miscible liquid drop in
another liquid then diffusion could complicate the situation by changing
the dimensions and properties of the droplet with time. If we are
concerned with a liquid drop falling some known distance, then Stokes
Law could still apply if the time-scale for diffusion over a distance
comparable to the drop diameter, d, is large compared to the fall-time.
A useful dimensionless parameter in this case might be (d^2/Dt) where D
is the diffusivity and t is the fall-time. Stokes Law would be valid if
$d^2/Dt \ll 1$. There are many possible complications in nature that could
cause departures from Stokes Law and each situation has to be judged on
its own merits. Surface tension forces can be important in bubble
motion (Sparks, 1978), thermal diffusion in movement of hot diapirs
through the mantle (Marsh, 1982) and chemical diffusion can be important
when a magma droplet moves through a magma of another composition.

     This digression on one simple example of fluid dynamics illustrates
three points that effectively answer the kinds of criticisms cited
above:

     (i) It is the magnitudes and balances of forces and physical
effects which determine the regimes of fluid flow not the actual values
of particular physical properties.

(ii) Experiments designed to confirm theoretical physical laws need not show complete dynamic similarity to the natural situations of interest.

(iii)Extrapolation of physical laws or fluid dynamic models can be made to natural processes that are impossible to study experimentally. The extrapolation will be valid if all the important physical and chemical effects have been included.

These remarks should not be construed as attempting to disguise the limitations of the experiments with aqueous solutions. For example both the Lewis numbers and Prandtl numbers of the experiments are not appropriate for magmas and theoretical developments are inadequate to make confident predictions of how these parameters will influence behaviour in magmas. Indeed experimental systems need to be found which have the same range of Lewis and Prandtl numbers as magmas. One of the central problems and areas of active research concerns the development of physical theories analogous to Stokes Law. In double-diffusive and compositional convection the recognition of new phenomena in tank experiments has so far outpaced the development of physical theories. The prediction of the occurrence and scale of double-diffusive layering in magma bodies remains uncertain until a more comprehensive theory is developed on how the layers grow and on the influence of high Lewis numbers and Prandtl numbers in magmatic liquids (Huppert and Sparks, 1984). In the case of marginal crystallization some progress has been made on theory (Huppert and Worster, 1985; Nilson et al., 1985; Brandeis and Jaupert, 1986) and some measure of agreement has been found between theory and experiment. We cannot be sure that a particular phenomena has geological significance without a sound theoretical framework backed up by good experimental data. In the next section some of the phenomena observed in tank experiments are discussed that might explain layering in igneous intrusions. We briefly comment on the extent to which theoretical developments and geological observations support the occurrence of these processes in magma chambers.

3.   LAYERING

Tank experiments with aqueous solutions have shown that layering can occur both in the liquid and in the solid product by a variety of different mechanisms, and on a range of length scales. Since it is the solid product which is preserved in layered intrusions, the processes that cause layering in the liquid should be carefully distinguished from those that form layering in the solid.

3.1 Liquid Layering

Experiments with aqueous solutions have documented a substantial number of different mechanisms that can cause double-diffusive layers to form and compositional gradients to develop. Geologically the most important situation in which liquid layers form is when there is a stable compositional gradient and an unstable thermal gradient. Layers can be formed in an initial compositional gradient by sidewall heating or

cooling, heating from below or cooling from above. Some of the characteristics and mechanisms have been reviewed elsewhere (Huppert and Sparks, 1984; Turner, 1985). A particularly important mechanism involves sidewall crystallization in which compositional and thermal gradients are generated by crystallization of a homogeneous solution (McBirney, 1980; McBirney and Baker, 1985) and layers can develop in these gradients. Melting processes can also cause layering (Huppert and Turner, 1980). Layering can result from replenishment; by creating new layers of different composition, by reheating existing gradients and by causing new compositional gradients to form by mixing processes (Huppert et al., 1982; Huppert et al., 1986a; Turner and Campbell, 1986). Table 1 summarizes the different layering and stratification mechanisms that have been identified. Layering can form easily in aqueous systems in many different situations.

3.2 Theory

Layering can form by several processes and there is no single comprehensive physical formulation. Each layer-forming situation needs to be considered as a separate case and many of the situations listed in Table 1 do not have a complete and well-established physical theory. Heating a compositional gradient from below has been studied for aqueous systems, and there is good agreement between theory and experimental data (Huppert and Linden, 1979). This physical theory, however, cannot be extrapolated with confidence to magma chambers, because there is still not a complete understanding of how the Lewis number and Prandtl number effect the scale and development of the layers. The detailed structure and instabilities of the double-diffusive interfaces are not completely understood, even in aqueous systems.

Some recent publications have cast doubt on the occurrence of double-diffusive layering in magmatic systems (McBirney, 1985; Spera et al., 1986). McBirney (1985) used linear stability theory as a criterion to establish whether double-diffusive convection can occur and he concludes that many geological situations are not in the correct regime for layers to form. However, non-linear theory shows that motion is definitely possible in cases where linear theory would predict no motion (Huppert and Moore, 1976). The calculations by Spera et al. (1986) are for the special case of steady Rayleigh-Bernard convection and for the parameters they use the flow does not develop a series of layers. These calculations are not, however, appropriate for many double-diffusive systems. For example, they do not predict layers in the case of a salinity gradient heated from below even though layers are observed to form (Huppert and Linden, 1979). Moreover, both are one-dimensional and do not allow for effects at side walls, for example, to influence the flow. We therefore consider that these views are premature because there is not yet a sufficiently well-developed framework of physical theory to make such pronouncements with confidence. On the other hand, we cannot be sure under what circumstances double-diffusive layers can form in magma chambers and some of the mechanisms observed in the tank experiments could well prove to be unimportant in magmas.

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TABLE 1. <u>Dynamical formation of layers</u>

Full accounts of the layering mechanisms can be found in the list of references

Physical situation	Simplest possible initial conditions	Layer dimensions	Main references
1. Heating a compositional gradient from below, or cooling a compositional gradient from above	stable compositional gradient	$h = 51(\varkappa_T/N)^{\frac{1}{2}}$ for aqueous solutions only $\varkappa_T$: thermal diffusivity N: buoyancy frequency	Turner (1968) and Huppert & Linden (1979)
2. Cooling or heating a compositional gradient at a vertical side wall	stable compositional gradient	$h = 0.65\Delta\rho/\frac{d\rho}{dz}$ $\Delta\rho$: density difference between wall and far field at mean far-field composition $\frac{d\rho}{dz}$: far-field density gradient	Huppert & Turner (1980) and Huppert et al. (1984)
3. Crystallization at a vertical wall of a container of finite volume	uniform composition and temperature	not known for any system	McBirney (1980); Turner & Gustafson (1981) and Thompson & Szekeley (1987)
4. Insulated sloping boundary in a two-component system	vertical gradients in both compositions, with one stably stratified and the other unstably stratified	not known for any system	Linden & Weber (1977)
5. Crystallization at a sloping floor in a container of finite volume	uniform composition and temperature	not known for any system	Huppert et al. (1986b)
6. Replenishment of resident fluid by heavy input	anything	given by thickness of replenished layer	Huppert & Sparks (1980)
7. Replenishment of residual fluid by light input	anything	a large entrained layer whose thickness increases with time as long as the input persists	Huppert et al. (1986a)
8. Melting the roof of a granite chamber by a basaltic melt	uniform basaltic layer		

Many of the features of greatest geological interest in the experiments with aqueous solutions involve compositional convection from growing crystals. Quantitative physical theories for compositional convection, compositional boundary layers and development of compositional stratification are not yet fully established, but there is enough known to suggest that these processes do occur in magmas. For simple models of the stability of boundary layers next to growing crystal faces (Huppert and Sparks, 1984) the compositional Rayleigh numbers are similar for aqueous solution experiments and for plausible conditions in magma chambers. The analysis of Nilson et al. (1985) indicates that compositional boundary layers can form by sidewall crystallization in magma chambers and can cause differentiation at geologically significant rates. Compositional effects on density generally overwhelm thermal effects in both aqueous and magmatic systems (Sparks and Huppert, 1984), and we believe that realistic models cannot avoid taking compositional convection into account as a major influence in developing zoning and layering.

3.3 Layering in the Solid

Layers in the solid products of aqueous solution experiments can be formed by a number of mechanisms. Chen and Turner (1980) showed that double-diffusive layers in the liquid would influence the growth of crystals, so that their morphology could change across an interface. They documented subtle changes of habit as crystalline solid layers formed from stratified and layered solutions. Kerr and Turner (1982) developed well-defined layers by growing crystals from tanks of fluid initially with stratified layers with different compositions. In a typical experiment, sodium sulphate solutions were overlain by copper sulphate solutions. Freezing from below caused layers of the two sulphates to form sequentially, but not repetitive layers were formed. Subtle layering in the final solid was observed in these experiments due to slight changes in conditions such as the change in laboratory temperature overnight or a sudden increase in cooling rate. Some other examples of layering due to different nucleation conditions are described in Part II.

Despite these studies, the role of double-diffusive fluid layering in the origin of igneous layering is still unclear. Irvine et al. (1983) developed the most specific ideas in their concept of downdip accretion in the Stillwater Complex. They maintain that rock layers of different mineralogy and composition form by crystallization from adjacent magma layers separated by double-diffusive interfaces. The rock layers grow laterally into the magma layers. This concept requires that each magma layer evolves progressively by release of buoyant residual melt at the margins of the accreting front. It requires the layers to dip inwards, and Irvine et al. (1983) suggested that the synformal shape of many layered intrusions is a consequence of this process. The scale of rock layering is metres to tens of metres and smaller-scale rhythmic layering is not of concern. The concept requires that the major layers be discordant to the position of the solid-ification front.

The downdip accretion concept is an elegant attempt to establish a relationship between liquid and rock layering, but many aspects, both on the geology and fluid dynamics, await critical evaluation. The experiments described by Huppert et al. (1986b) and in Part II involve crystallization of aqueous systems along an inclined floor. These usually also display double-diffusive layering. Some of the features postulated by Irvine et al. (1983) have been confirmed by the experiments, such as cusp-shaped solidification fronts and compositional boundary layers moving up slope. A possible consequence of the down-dip accretion concept is that rock layers may become discordant to the solidification front. However, we have failed so far to form solid layers with a marked discordance to the solidification front. Weak layering was sometimes observed to develop in the solid which was parallel to the cooling boundary and solidification front. These results may merely indicate that we have failed to find the right experimental conditions to reproduce layers as envisaged for the downdip accretion concept. Cryptic layering (surfaces of constant solid composition) on the other hand can be strongly discordant to the solidification front (see Part II) and mimics the discordances between cryptic and rhythmic layering in some intrusions such as that found in the Fongen-Hyllingen intrusive series by Wilson and Larsen (1985). The geological evidence for downdip accretion is not yet fully convincing, because there are several other processes that could be invoked to explain the geological relationships, but it is an interesting hypothesis that warrants further investigation.

Perhaps the most convincing application of tank experiments to the origin of layering concerns those experiments investigating the fluid dynamics of magma chamber replenishment. Experimental studies (Huppert and Turner, 1981b; Huppert et al., 1982) have shown that replenishment by dense hot fluid results in a temporary liquid layering in which the new input fluid crystallizes on the floor of the tank. A physical theory has been developed for this process (Huppert and Sparks, 1980) and the process has been widely accepted as a satisfactory explanation of large cyclic or macrorhythmic layering (Irvine, 1980; Wilson, 1982; Raedeke and McCallum 1984; Campbell et al., 1983; Tait, 1985).

Replenishment by light inputs has also been investigated by Sparks et al.(1980) and by Campbell et al.(1983), and in tank experiments by Huppert et al. (1986b). Mixing of magmas in plumes can cause precipitation of crystals, which then settle to form discrete layers. Mixing of magmas can cause temporary crystallization of unexpected phases or combinations of phases, because of the complicated topology of phase diagrams. Thus some monomineralic layers may well form by mixing processes during replenishment (Irvine, 1977). Layering in chromite seams has been attributed to magmatic stratification formed during replenishment (Turner and Campbell, 1986).

4.   CONCLUSIONS

Tank experiments with aqueous solutions offer a simple and effective approach to the understanding convective and solidification processes in multicomponent fluid systems. Many of the processes observed in these experiments are likely to occur in magmatic systems. However, the physical theories are not yet developed for many of the newly identified phenomena, so extrapolation to geological situations requires caution. Layering in the liquid is a prominent feature of many experiments with aqueous solutions and can be caused by marginal crystallization and compositional convection, and by replenishment processes. Weak layering has also been formed in the solid products of experiments. The relationships between solid and liquid layering are still not well understood, and there is still only limited evidence about how and if the various kinds of layering in intrusions are related to liquid layering. Replenishment of magma chambers by new influxes, however, does provide a convincing explanation for large-scale cyclic layering. We suggest that further progress might emphasise two points. First, there might be increasing efforts to give petrologists a more complete background in fluid dynamics so that they are able to evaluate the various models in an informed and critical way. Some of the present difficulties and controverises have arisen either from total uncritical acceptance of fluid dynamic models or from a complete rejection based on lack of knowledge. Second the development of sound physical theories are essential. The recognition of new phenomena in experiments and the sophistication of some geological models are running ahead of understanding of the basic physics.

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