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The importance of clustering phenomena in magmas

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Abstract

Currently, clustering phenomena receive considerable attention in stellar systems and in fluid-dynamics problems. Although the whirling motions are regarded as a problem of basic significance for geology, the mechanisms for forming clustering processes in magmas are still poorly understood. Luckily, the discovery of felsic clusters in some basaltic rocks of Western Sicily throws light on this subject. The main aspects of cluster evolution occurring in the magma are described, and a three-stage process is proposed. The coalescing mechanism and the interparticle interactions due to the surface forces are emphasized.

In the author's opinion the conditions now existing in the Earth's upper mantle favour clustering phenomena of this kind.

Introduction

Vortices consist of matter in motion around a common axis. The clustering concept is very important because the whirling motions are observable, for instance, in the galaxies of the Universe, in the spiral arms of the galaxies, in the planetary system, in the Earth's atmosphere and in magmas. In this latter case, clustering processes are of interest in the study of the movement and solidification of magma and lava as well as in evaluating processes of energy extraction.

In the simplest approximation, known as the Boussinesq approximation, a fluid is assumed to be incompressible (except for thermal expansion, which provides the buoyancy) and convective onset in a non-rotating fluid is governed entirely by the Rayleigh number. For a fluid layer of depth H, with constant kinematic viscosity $v = \mu/e$ and thermal diffusivity k, the Rayleigh number is: $Ra = \frac{g\alpha\beta H^4}{vk}$, where g is the gravitational acceleration, α is the coefficient of thermal expansion, and β is the difference between the actual temperature gradient and the adiabatic temperature gradient. The essential idea is that convection in magmas does not begin until the Rayleigh number exceeds some critical value, which depends on the imposed boundary conditions, but is usually in the neighborhood of 1,500 (Spera, 1980). It may be thought of as the ratio of buoyancy forces favoring convection to the viscous forces retarding flow. Normally natural convection will become turbulent when the Rayleigh number exceeds 10^9 (Rohsenow & Choi, 1961). The high Prandtl number $\frac{v}{k}$ in this case, however, tends to raise this critical Rayleigh number turbulent transition point. Hieber and Gebhart (1971) studied this problem and their result for the critical Rayleigh number for the onset of turbulence can be put in this form:

$$Ra_{critical} = 7{,}23 \times 10^4 \left(\frac{v}{k}\right).$$

The critical Rayleigh number determined from this equation for the heat exchanger in basaltic magma is $4{,}5 \times 10^{12}$ (Hardee, 1981). So, according to Hardee (1981), since the actual Rayleigh number ($9{,}9 \times 10^{16}$) is much larger than the critical Rayleigh number ($4{,}5 \times 10^{12}$), the convections is turbulent.

In this paper, on the basis of the remarkable textural characteristics of the Sicilian igneous rocks and taking into account the present state of experimental knowledge concerning the interparticle forces, a three-stage transport mechanism is proposed. It is suggested that this mechanism may play a very important role in upper mantle convection.

Geological setting

The igneous rocks in which felsic clusters occur, are situated near S. Stefano di Quisquina (Sicily). They are located in the Sicano basin that lies along the western Sicily bridge between the Neogene-Pleistocene basins of Caltanissetta on the east and Castelvetrano on the west (Catalano & D'Argenio, 1978). Detailed geologic mapping of the igneous outcrop has been by Broquet (1968). From the Upper Trias to the Miocene the Sicano basin was involved in tensional tectonic activity verifiable in the tectono-sedimentary evolution of the facies and in the characteristics of the fissural magmatism. The E-W fault system and its conjugates allows for the identification of the tectonic directrices which fed this magmatism. Beginning from the Tortonian, in connection with the anticlockwise rotation of the African plate, the Maghrebian compression front affected this area building the chain of the Sicani Mts.

The Sicilian igneous rock was first reported on by Baldacci (1886) who considered it a basaltic dyke. The outcropping rock consists of three small hillocks that crop out upon a probable erosion surface of Palaeocene, Eocene and Oligocene sediments. It generally shows porphyritic texture, but panidiomorphic and ocellar structure are also observed. The fresh basic samples are black, whereas the weathered ones have a greenish colour due to the presence of chlorite and other alteration products. The light clusters fundamentally are subspheroidal or ellipsoidal and form a strong centrast to the basaltic matrix. Lacking geochronological data and because of the poor field-relationships the actual age of the outcrop is uncertain.

Coalescence and clustering in the magma

During research carried out on the Sicilian rocks of the Sicano basin, a mechanism forming silicic segregations from basaltic magma was discovered (see Lucido, 1983). This mechanism suggests that the formation of silicic segregations is a consequence of liquid immiscibility phenomena. In particular, the author found that at the moment of immiscibility much of the magmatic melt was in a state of intensive motion. For instance, Figure 1 shows a vortex of felsic composition in the surrouding basic host-rock. Minerals occurring in the basic host-rock are the same as those in the

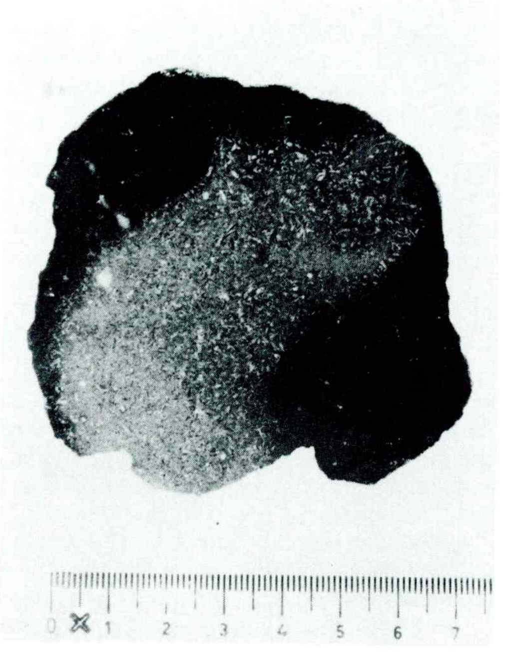

Fig. 1. Hand-specimen showing a whirling felsic portion in the surrounding basaltic magma. Scale in cm

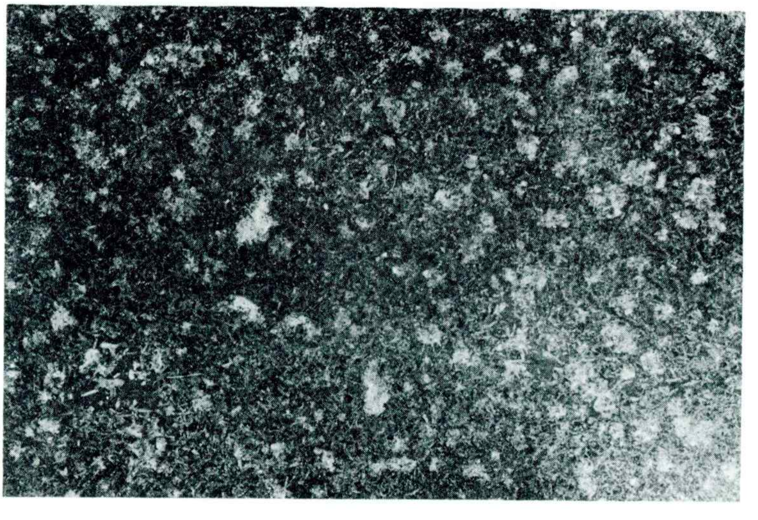

Fig. 2. Felsic clusters, scattered in the basaltis matrix, point that under certain circumstances a phase seration occurred in the Sicilian magma. Magnification, ×6

whirling felsic portions, but they occur in differing proportions, with light and hydrous phases concentrated in the felsic fractions. These felsic aggregates are characterized by the following mineralogical assemblage: K-feldspar, plagioclase, kaersutite, titanaugite, analcite and zeolites. Among the accessory minerals are: ilmenite, titanomagnetite, anorthoclase, biotite, aegirine-augite and apatite (see Lucido, 1981).

During immiscibility, due to liquid phase separation of the basaltic magma, an enormous number of barely visible clusters (having diameter up to 1 mm in size) appear throughout the Sicilian magmatic melt (Fig. 2). This new liquid phase differs sharply in composition from the original magma; at the same time, the matrix also changes its composition. The diffusion from one phase into the other (and vice-versa) is promoted by the fact that the liquid-liquid interface is always mobile (Delitsyn et al., 1974). From the equation of motion of a liquid drop in liquid we know at the liquid-liquid interface, the normal velocity component vanishes, whereas the tangential component is finite (Levich, 1959). Because translational and rotational motions at the instant of immiscibility originate simultaneously, everywhere and point in the most diverse directions, the motion becomes chaotic or turbulent. The turbulence of magmatic liquid produces zones of lower component concentrations in the space between two converging and independently rotating clusters, which then begin to coalesce via a neck (Fig. 3), and become larger. In the emulsion generated in the magma, the clusters tend to become larger so as to decrease the surface energy of the system, while at the same time turbulence tends to deform and shatter the enlarged clusters, and prevents them from growing beyond a certain size. So, such a magma is an unstable system, in which there are constant changes in surface tension, viscosity

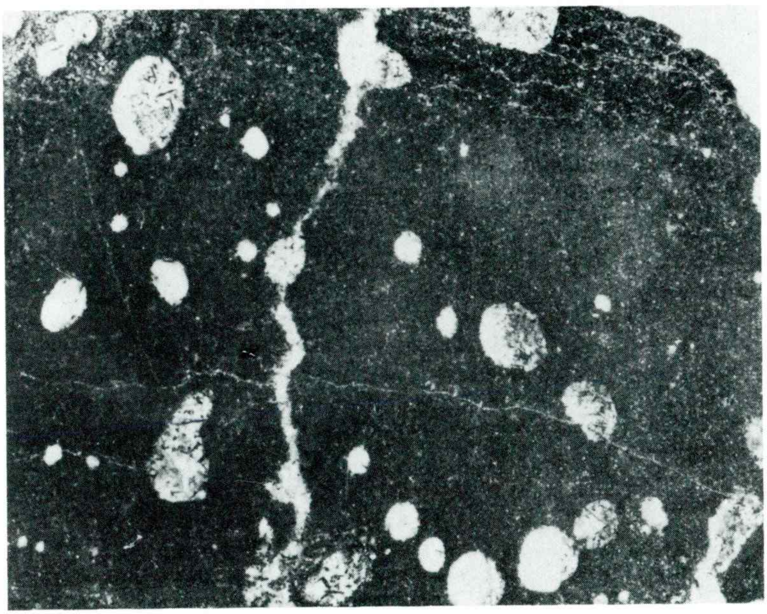

Fig. 3. Photograph showing light clusters coalescing via a neck to form larger clusters. Some clusters clearly show cups point towards the basaltic liquid. Magnification, ×3

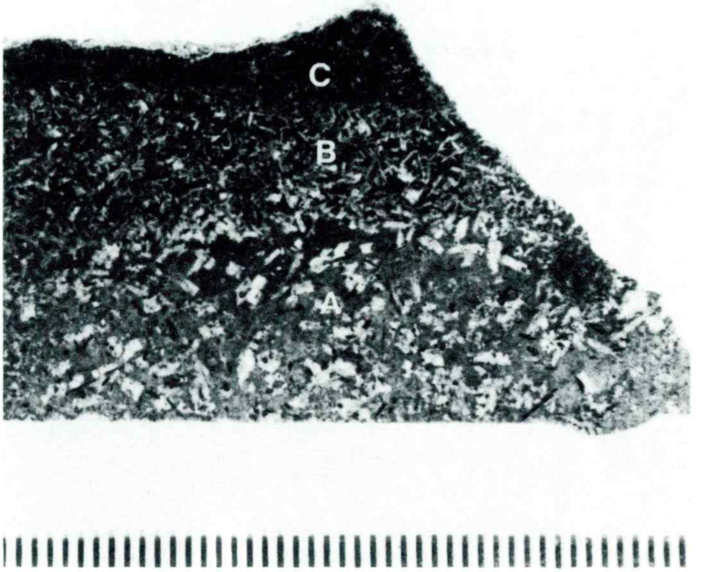

Fig. 4. This Fig. exhibits three different-textured layers. The lower layer (A) shows a phaneritic texture, with common subvariolitic crystallization between inosilicates and feldspars. The middle layer (B) is characterized by subophitic texture with equigranular and elongated feldspar crystals. The upper part (C) is the basaltic portion. Undoubtedly density gradients between the liquid phases must have played an important role. Scale in mm

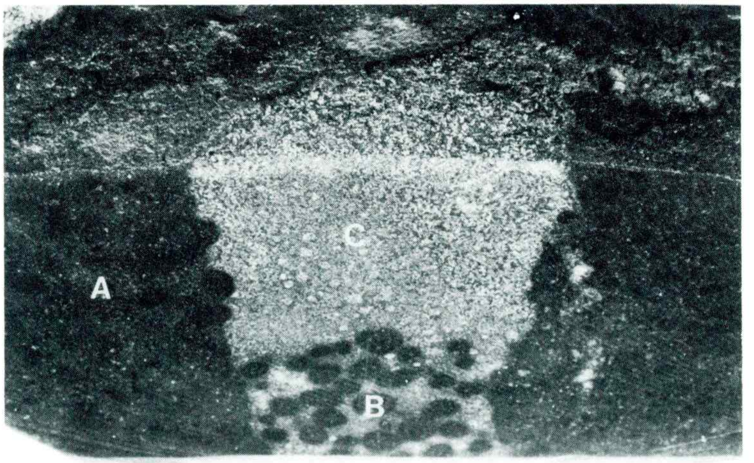

Fig. 5. A single spheroidal whirl forming an independent flow in the Sicilian magma. The view of Figure 5 is three-dimensional: the upper part of it shows half a whirl such as in occurs in outcrop; the lower part of the photograph represents the whirl vertically dissectioned along its diameter. Width of Figure 5 is 25 cm

and density until the melt is stabilized and the motion ceases. Upon separation into immiscible liquids, layering of liquid phases chiefly occurs during the period of turbulence in the melt. After the period of intensive turbulence, there is a further change in the volumes of liquid layers. This process occurs under comparatively quiescent conditions (Fig. 4) and is controlled by the rate of settling (or floating up) of the band of clusters in the corresponding matrices (Delitsyn et al., 1974). Upon separation, the individual whirl forms independent flow in the magma (Fig. 5). In whirls formed in the turbulent liquid, there are zones of denser and less dense emulsion. In emulsion zones that have thickened to a certain density, motion ceases (i. e. viscosity increases), so relative to zones of less dense emulsion these zones behave as solid boundary surfaces, along which the friction is higher than elsewhere in the magmatic liquid. Within the remaining less dense zones, the motion of emulsion will continue to create new zones of denser and less dense emulsion until motion ceases completely. Field and microscopic observations show that clusters with a diameter more than a certain critical value are deformed (markedly elongated) and behave like liquis under gravity. Vice-versa, clusters having a subcritical diameter are not deformed and retain their spherical shape.

Discussion and conclusions

Theoretical considerations

Various magmatic processes (for instance, differentiation, crystallization, polymerization and degasification) occur at the expense of a certain energy reserve (Zavaritskii & Sobolev, 1964) and lead to the formation of colloid systems. Colloids dispersed in the magma can be either magmaphilic or magmaphobic, depending on whether the energy obtained by their solvation by the magma liquid is higher or lower, respectively, than the sum of their aggregation energy and the dissociation energy of the liquid silicate (Yariv & Cross, 1979). It is therefore not surprising that various mineral species are recorded to have been derived through the colloidal state and show spheroidal forms, e.g. the so called colloform structures (Augustithis, 1982).

The present state of experimental knowledge concerning the forces that govern the properties of colloidal dispersions has been recently reviewed (Israelachvili, 1981). According to the Hellman-Feymman theorem all intermolecular forces are strictly electrostatic in origin. Nevertheless, it has been found convenient to classify the major forces between particle surfaces into van der Waals' forces, electrostatic and double-layer forces, polymer and steric forces, structural forces (hydrogen bonding, hydration and hydrophobic interactions), and adhesive forces. These interparticle forces are not as stong as covalent or metallic binding forces and their weakness and very short range (« 1 μm) makes them difficult to measure directly.

The behaviour of a colloidal system is far more complex than any gas-fluid system. Ageing and time-dependent effects often occur for dispersions in electrolyte solutions when equilibrium is attained slowly or not at all. More complex behaviour, such as hysteresis effects, occurs in polymer dispersions. Thus, at present not even an empirical equation of state, similar to the van der Waals' equation, exists for any colloidal system. It is worth reflecting what a force between two particles implies. For large particles with large forces it is realtively simple: if the force is attractive they will stick, if repulsive they will repel. But when the forces are weak and the particles small, entropy effects may not be ignored and only a complete thermodynamic or statistical mechanical treatment can expose all the subtle concentration – and temperature – dependent phase transitions, phase changes, polydispersity, and other diverse properties of a colloidal dispersion (see Israelachvili, 1981). Surface forces that lead to aggregation are primarily van der Waals. Recently, experimental techniques have been developed to measure the van der Waals' forces in liquids, either indirectly (Sabisky & Anderson, 1973; Requena et al., 1975) or directly (Blake, 1975; Israelachvili & Adams, 1978; Derjaguin et al., 1978).

The above surface forces may therefore be responsible for the attraction between particles. In a magma, for example, the coalescence fundamentally depends on these interparticle forces existing in the melt. According to Delitsyn and others. (1974), in fact, these forces cause considerble turbulence during the coalescence, hastening equilibration. An excellent two-dimensional display of this turbulence is observable on the surface of a hot thin soup between fat globules. Haller (1965) has develepoed the mathematical theory of coalescence, based on kinetic studies of the liquid-liquid immiscible microphases in alkali borosilicate melts. As a matter of fact, most researchers (e.g. Philpotts & Hodgson, 1958; Ferguson & Currie, 1971, 1972; Gélinas et al., 1976; Cawthorn et al., 1979; Philpotts, 1979; Furnes et al., 1981) agree that coalescence is involved in the silicate liquid immiscibility origin.

Comparison with theory

The spinodal decomposition is important or potentially important in every system in which we observe a clustering phenomenon. The theory of spinodal decomposition is based on the diffusion equation modified by thermodynamic requirements, which relates a spontaneous flux of matter to a gradient in composition (see, for example, Cahn, 1968). This theory is phenomenological, and each parameter can be measured by independent thermodynamic or diffusion experiments. Consider for instance the dynamics of the various clustering phenomena in a model system composed of two species of mobile individuals in which individuals of the same species have a short-range preferential attraction to each other.

Take first the case that is almost random: preference for like members is small. The individuals execute a nearly random walk, but have a tendency to join favourable clusters and linger there a little longer. Clusters have no permanence, they form and disappear. If we perform a diffusion experiment by setting up a concentration gradient, the gradient would tend to disappear. The attraction to high concentration is not sufficient to prevent normal diffusion down the concentration gradient.

Now consider the other extreme-preference for the like species is so large that in a gradient the flux of the individuals is in the direction in which they are attracted, i.e., up the concentration gradient. This kind of model leads to spontaneous separation into two »phases«. This system is within the spinodal. The individuals in the gradient move toward the cluster of their species and cause its concentration to increase, leaving a depleted zone around it. The outer edge of this depleted zone contains individuals that now also sense a concentration gradient, but away from the original particle. Because of their short-range interactions they sense only the depleted zone and move away from it, building up a new cluster a short distance away from the original one. We thus expect rapid formation of extremely small clusters approximately periodically arrayed in space.

In the case intermediate between these extremes, attractions are not steong enough to cause a flux up a concentration gradient but strong enough that a large cluster will attract and hold the individual. This situation is the case for nucleation and growth. Let's now see what the processes are, and how and when they operate in magmas.

When preference for the clusters is high, phase separations may be initiated by statistical concentration fluctuations not far from the critical, which allow the formation of stable nuclei at an extremely small degree of supersaturation (Cahn, 1961). In particular, if the controlling feature of the phase separation is the charge density, then the temperature decrease brings about a decrease of the fluctuations characterizing the critical behavior. In this manner, the fluctuations involving high charge ions really will be more damped than the others. According to this, the magmatic fluid will tend to separate into two immiscible liquid portions, one enriched in elements like iron, magnesium, calcium, titanium etc., and the other enriched in low charge density ions and elements having the tendency to form polymaric networks.

In the case of the Sicilian rocks, the extreme-preference for light clusters is so large that in a gradient the flux of the clusters is in the direction in which they are attracted. Because of the short-range interactions due to the surface forces, the clusters coalesce to form larger clusters (Fig. 3).

Comparing the foregoing results and evidence to theory, a three-stage process for the Sicilian basaltic magma may be reconstructed.

1. In the first stage cybotaxic zones, the nuclei of future phases, are formed in the Sicilian magma. Indeed, it is possible to see very small light spots in the basic portion (A) of Figure 5. This splitting phenomenon can be explained in two different ways: a) the cybotaxic zones are formed in a homogeneous melt near the binodal point; b) the phase separation occurred by spinodal decomposition. On the basis of the textural characteristics of the Sicilian rocks, and on the basis of the known data on the dynamics of fluids, the writer thinks that the clusters originated by spinodal mechanism.

2. The second stage is the stage of liquid immiscibility (see Lucido, 1981). A well-developed example of this phenomenon occurs in the portion (B) of Figure 5.

3. After unmixing, an agglutination process of clusters which tends to stabilize the turbulence of the magmatic melt occurs. The light portion (C) of Figure 5 represents the result of this agglutination process, and it is a portion richer in alkalic alumino-silicates than the surrounding basic rock.

Application to the upper mantle

Although there is still no indication of a consensus on the nature of mantle dynamics, importance of whirling motions in the thermal evolution of the Earth is clearly established. It may be expected that convection is the dominant heat transfer mechanism in the Earth's upper mantle. In this regard, in fact, magmatic vortexes under the Earth's crust were long ago hypothesized (e. g. Hopkins, 1939; Bull, 1921; Holmes, 1931; Pekeris, 1935; Hales, 1936).

Convection in magma at liquidus and subliquidus temperature has been analyzed using Bingham plastic and power-law rheology models (see, for instance, Hardee & Dunn, 1981). It is interesting to note that a noticeable change in the trend of the convective heat flux data has been observed in the vicinity of the liquidus temperature. Above the liquidus, magma (or molten lava) behaves as a Newtonian fluid (Shaw et al., 1968). At temperatures below the liquidus, non-Newtonian behavior begins to appear (Shaw et al., 1968; Pinkerton & Sparks, 1978). It is generally believed that this deviation from Newtonian behavior is due to time-dependent structural changes in the melt.

The application of the Sicilian phase separation mechanism to the upper mantle at first leads to the formation of very small clusters (new silicate-liquid nuclei) having more acidic composition than the surrounding melt and of course pseudoplastic behavior.As the phase separation proceeds the magmatic fluid will comprise immiscible portions in a state of dispersion. After unmixing, in the long run the interparticle surface forces decidedly prevail and are resonsible for an attraction process of colloidal clusters (mushes) which tends to stabilize the turbulence of the melt. In this attraction process, finite compressibility and viscous dissipation tend, of course, to decrease the flow speed for a given Ra, and so contribute to the melt stabilization.

Recently, Arzi (1978) suggested that most of the upper-mantle low velocity zone (L V Z), if partially melted, is likely to be stabilized somewhat below a non-zero rheological critical melt percentage (R C M P). For most rocks, the R C M P is probably within the range of $20 \pm 10\%$. It is a drastic transition with viscosities lowered by several decades due to breakdown of the solid and interlocked crystalline skeleton, and onset of rapid convective heat transfer. In agreement with Arzi (1978), much is explained if the original suggestion by Press (1959) that the L V Z is at »a state nar the melting point« is revised to »near the R C M P«. According to this, and assuming that in proximity to the lithosphere-asthenosphere boundary magma tends to occur at near the liquidus temperature, »the writer proposes that the L V Z is to be considered as the transition zone from Newtonian to non-Newtonian behavior«.

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