

Distribution of partial melt in a felsic system: the importance of surface energy

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Abstract. The texture, distribution, and infiltration tendency of a quartz-albite melt in equilibrium with a synthetic, texturally-equilibrated quartzite was examined in a series of distribution and infiltration experiments at 1,250° C and 8 kbar hydrostatic pressure. Wetting angle measurements from melt distribution experiments show a dihedral angle (θ) of 60 degrees, implying a quartz/quartz interfacial energy approximately 1.7 times the quartz/melt value. Because of this specific relationship between interfacial energies, the system can achieve its lowest surface free energy state with the melt either in pools or along grain edge intersections, possibly forming some interconnected channels. Stability of melt in pockets and along grain edge intersections was observed in a fourteen-day, dispersed-melt experiment, yet melt pools failed to disperse into the quartzite during infiltration experiments. Comparison of the observed dihedral angle with previously measured surface energy values for the melt and quartz shows excellent agreement, and also demonstrates that an aggregate of randomly orientated anisotropic grains acts approximately isotropically.

While these experiments are not strictly applicable to real crustal systems, they do indicate that, at least in some felsic systems, the melt has no preference for uniform grain-edge wetting relative to collection at grain corners or in large pools. This “ambivalent” behavior is attributable to the 60-degree wetting angle, which has been shown to separate systems in which melt tends to disperse in interconnected channels ($\theta < 60^\circ$) from those in which melt tends to become isolated at grain corners ($\theta > 60^\circ$).

Introduction

The origin of granitic magmas is a complex problem to which there is no single solution, but it is now generally agreed that melting is involved to some extent. While classically we interpret granites as crystallizing from a body of mostly liquid magma, recent theories tend to support the view that some granitic magmas are composed of nearly equal amounts of suspended solids and liquid (White and Chappell 1977; Winkler and Breitbart 1978). The application of trace element geochemistry to granitoid bodies reinforces this theory, and suggests that partial melting is involved in the formation not only of migmatites but also of some gneisses (Martin 1979; Price and Taylor 1977; Rapela and Shaw 1979).

Experimental studies have shown that partial melting

of gneisses will give rise to a granitic liquid and a less-fusible residue (Mehnert et al. 1973). It has also been suggested that removal of this granitic liquid will cause dehydration of the residue, and that a mechanism of this type may be responsible for the existence of some anhydrous granulites (Nesbitt 1980). However, many questions remain unanswered regarding the nature and distribution of partial melts in crustal magmatic systems. Unfortunately, these questions cannot be answered on the basis of petrography due to a lack of preserved evidence in natural systems.

In the past few years, experimental work has provided considerable insight into the textural aspects of melt distribution in partially-molten mafic systems under hydrostatic pressure (Cooper and Kohlstedt 1982; Waff and Bulau 1979; Watson 1982). The results of these studies show that a basalt melt will wet the grain edge lines of a texturally-equilibrated peridotite, forming interconnected channels. However, while this behavior is well established for the mafic system studied, we cannot assume it will also be observed in felsic systems, because wetting characteristics are critically dependent upon specific crystal/crystal and crystal/liquid interfacial energies. For this reason, we undertook a series of experiments in the quartz-albite system in order to 1) determine the equilibrium melt distribution and wetting angle in samples consisting of quartz plus (1, 3, 5, and 15%) quartz-albite melt, and 2) observe whether pools of quartz-albite melt will disperse by infiltrating the dry grain edges in a texturally-equilibrated quartzite with which it is in chemical equilibrium.

Theory

The equilibrium arrangement of grain edge lines is governed by the necessity that all surface forces acting on the grain edge lines be balanced. This balance can be expressed by the relation;

$$\frac{\gamma_{jk}}{\sin \theta_i} = \frac{\gamma_{ik}}{\sin \theta_j} = \frac{\gamma_{ij}}{\sin \theta_k} \quad (1)$$

where γ_{jk} is the surface energy of interface jk and θ_i is the contact angle (Bulau et al. 1979; Smith 1964). If we consider a two phase system consisting of a crystal aggregate and a melt, only one dihedral wetting angle is observed (assuming a random orientation of grains) and is given by the equation:

$$2 \cos \frac{\theta}{2} = \frac{\gamma_{ss}}{\gamma_{sl}} \quad (2)$$

where θ is the wetting angle.

Any system of molten or solid phases will tend, through textural adjustments, to minimize its total surface free energy. Under conditions of hydrostatic pressure, the specific texture or melt distribution that is approached will be determined by the interplay between the free energies of the solid/solid and solid/liquid interfaces. Depending on the relative value of these quantities, four distinctly different types of behavior can be expected (Kingery et al. 1976):

1) The melt may wet all grain surfaces. This case is realized when γ_{sl} is small ($\gamma_{ss}/\gamma_{sl} > 2$) and the wetting angle θ , therefore, is zero (see Eq. 2). Judging from near-equilibrium textural studies of partially-molten rocks and rock analogs, however, this situation is not generally relevant to natural silicate systems.

2) The melt may be distributed along grain edge intersections only, still forming an interconnected network in three dimensions. This behavior is exhibited by systems in which the ratio γ_{ss}/γ_{sl} is less than 2 and greater than $\sqrt{3}$, so that the wetting angle is between zero and 60 degrees. Such systems are characterized by a lower value of total interfacial energy in a wet grain edge region than in a dry one of the same size (Watson 1982); this characteristic, along with the three-dimensional continuity of grain edges in a rock, allows for the by infiltrating the grain edges of the host material (see Watson 1982, for a demonstration of this process).

3) The melt may be generally restricted to grain corners, thus lacking three-dimensional continuity, yet still show no tendency to collect into larger melt bodies. This type of behavior can be expected of systems in which the ratio γ_{ss}/γ_{sl} is between 1 and $\sqrt{3}$, which implies a wetting angle between 60 and 120 degrees.

4) The melt may tend to collect in large pools or pockets so as to eliminate wetted grain boundaries altogether. Such a tendency would be observed in systems in which the ratio γ_{ss}/γ_{sl} is less than one and the wetting angle will be greater than 120 degrees.

It is thus apparent that knowledge of the wetting angle alone is sufficient to predict the melt distribution characteristics (including dispersion/infiltration or pooling tendencies) of a given system.

This conclusion and the foregoing discussion provide sufficient background and context for presentation of our primary experimental approach; for a more complete explanation of surface energy/texture interrelationships in par-

tially molten materials, the reader is referred to Bulau et al. (1979), Kingery (1976), Smith (1964), and Watson (1982).

Experimental methods

The system $\text{NaAlSi}_3\text{O}_8 - \text{SiO}_2$ was selected for study because it is consistent with the dominance of quartz and feldspar in natural felsic systems. A melt composition of $\text{Ab}_{50} - \text{Qtz}_{50}$ by weight was found to be in chemical equilibrium with a synthetic quartzite at conditions of 1,250°C and 8 kbar hydrostatic pressure. These conditions are just below the quartz liquidus of the melt, insuring saturation with respect to quartz during a run and thus eliminating all driving forces for textural rearrangement other than surface energy. The temperature is, of course, unrealistically high for crustal fusion events, but in this initial study the attainment of textural equilibrium (promoted by high temperature) was considered of primary importance.

Our initial experiments were of the (attempted) "infiltration" variety, in which a pool of quartz-albite melt occupied one end of an experimental capsule at the start of the run, and a synthetic, texturally-equilibrated, non-porous quartzite occupied the other end.

The synthetic quartzite was prepared by powdering optically pure rock crystal and sieving it to obtain an average grain size of approximately 50 microns. The material was then loaded into a graphite capsule and annealed for 10 h at 1,300°C and 10 kbars in a piston cylinder apparatus. The end product was a texturally-equilibrated quartzite, which is shown in Fig. 1. The quartz-albite glass was prepared by fusing a mixture of appropriate oxides inside three-millimeter platinum capsules for two hours at the same conditions as the quartzite. Both the glass and the quartzite were synthesized as three-millimeter diameter slugs, which were subsequently ground to cylindrical form.

The assembly for the infiltration experiments was made by placing together 3 × 3 millimeter cylinders of quartzite and glass inside a platinum tube. Optimum contact was insured by polishing contacting faces. Once completed, the assembly was placed vertically, melt side down, in a 3/4-inch NaCl/Pyrex/crushable Al_2O_3 piston cylinder assembly, as shown in Fig. 2.

The second series of experiments were of the melt distribution type, in which the assemblage for the experiments was made by grinding the quartz-albite glass to a very fine powder and then mixing it with the appropriate amount

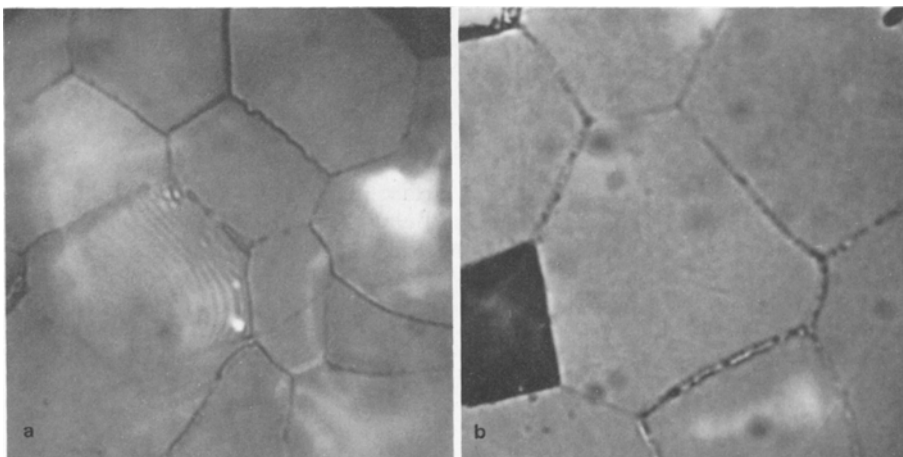


Fig. 1 a, b. The texture achieved in the synthetic quartzite at (a) × 625 and (b) × 1,250. Grain boundaries appear as dark lines and dark polygonal patches represent grains plucked out during polishing. The average grain size is 50 microns

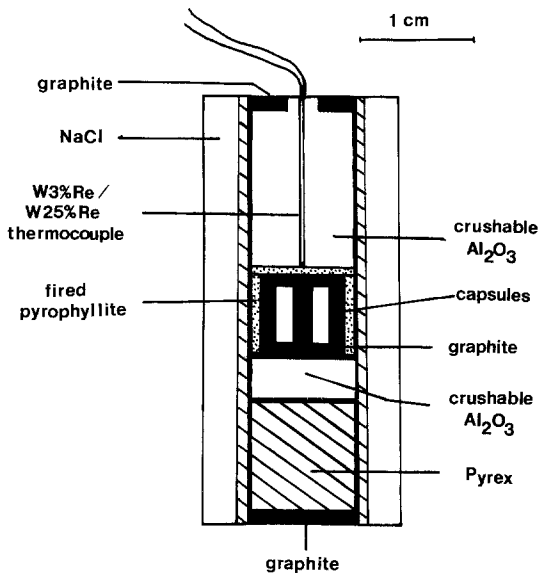


Fig. 2. The piston cylinder assembly used for both infiltration and distribution experiments. The assembly is capable of maintaining stable, hydrostatic, dry conditions for 14 days or longer. The Pyrex bottom filler piece is soft at run conditions

of 50-micron quartz grains until the mix was homogeneous. Mixes were made containing 1, 3, 5, and 15% melt (glass), which were then sealed and in platinum capsules and run in groups of three.

The infiltration and distribution experiments were sub-

jected to the same run conditions of 1,250° C and 8 kbars. Run durations for the infiltration experiments were 24 h, while the distribution experiments were at run conditions for 1, 6, and 14 days. This long duration was found necessary to approach textural equilibrium.

Experimental results

Both the infiltration and distribution runs were examined under reflected light after etching with dilute HF. The quartzite in the infiltration experiment was very friable, and upon visual inspection revealed no infiltration of the melt. A typical section of the quartzite/melt interface is shown in Fig. 3.

The products of the melt distribution runs were not nearly as friable as the glass-free quartzite, due to the general presence of glass along grain edges. As judged by the method described below, runs shorter than six days were not texturally equilibrated. The six-day run showed some approach toward textural equilibrium, but the 14-day run yielded the best results. Some small melt pockets (Fig. 4) are present in all melt distribution runs. We interpret these as having developed during initial pressurization of the sample, probably from the force filling of spaces between grains. Wetting angles on the edges of pools and on grain edge lines revealed the true dihedral angle to be approximately 60 degrees.

Evidence for the approach to equilibrium was based upon the Riegger and Van Vlack method for extracting true dihedral angles from randomly cut, two dimensional

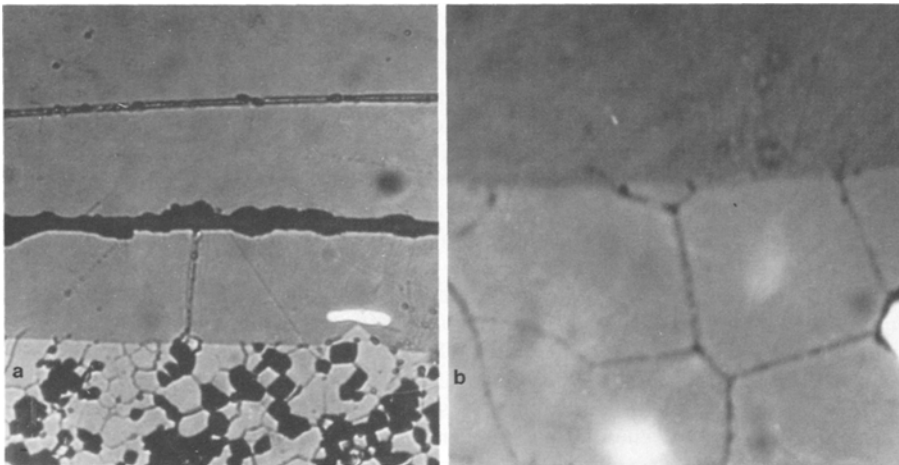


Fig. 3a, b. The quartzite/melt interface at (a) $\times 125$ and (b) $\times 1,250$. Dark polygons represent plucked grains; their occurrence right up to the interface is indicative of the absence of penetrating melt, which is confirmed in the higher magnification photograph. Dark horizontal bands above the interface in (a) are dilational cracks in the quenched glass, produced upon cold depressurization

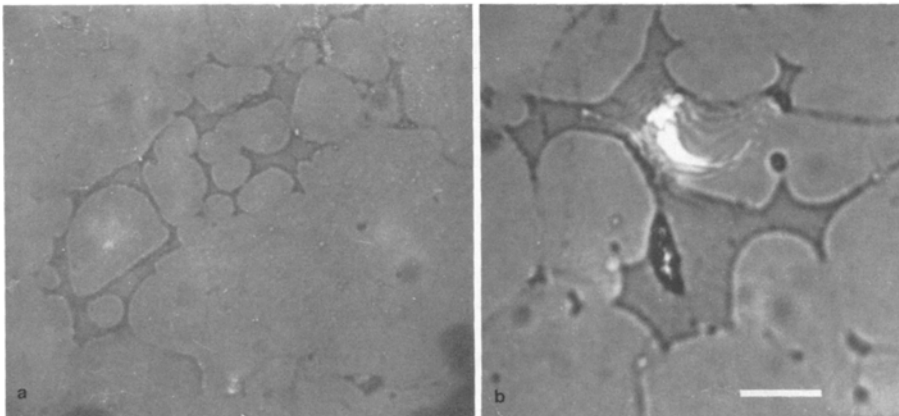


Fig. 4a, b. Melt pools from the 14-day melt distribution experiment with 1% melt.

a Dark, cusped patches are glass, defining an irregular, large pool in which grains do not appear to make contact.

b Smaller pockets also occur, in which fewer "swimming" grains are found, and around which the dihedral angle is roughly constant. The white bar in the photograph is approximately 20 microns

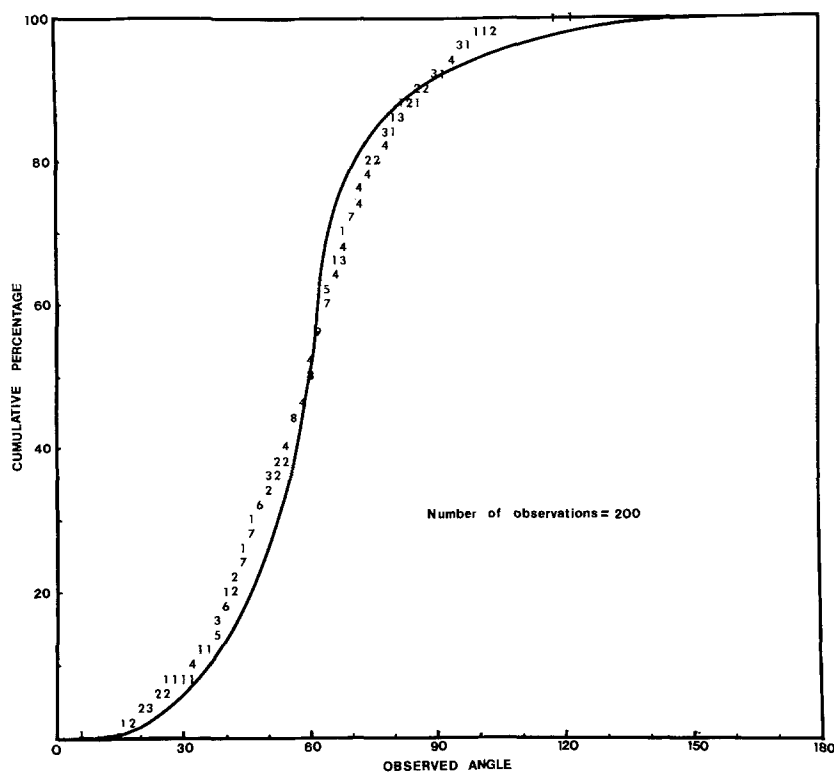


Fig. 5. A plot of cumulative percentage vs. observed dihedral angle. Numbers represent angle measurements from the 14-day distribution experiment; the dark line is the predicted theoretical distribution of observed angles from a two dimensional plane cut through a three dimensional aggregate with a true dihedral angle of 60 degrees (Riegger and Van Vlack 1960)

surfaces through a three dimensional aggregate (Riegger and Van Vlack 1960). This involves statistically correcting the two dimensional plane by constructing a cumulative frequency plot of observed dihedral angles, and comparing that plot with the expected theoretical frequency. Figure 5 shows the observed curves for the 14 day experiment in relation to the theoretical equilibrium curve for a dihedral angle of 60 degrees. Of the two hundred measurements, 100 were taken from melt pools, and the rest from grain edge intersections. If the system were acting anisotropically with respect to surface energies, then the cumulative frequency plot would deviate significantly from the predicted theoretical value for a 60 degree dihedral angle (Riegger and Van Vlack 1960). Figure 5 shows only a slight deviation, which is probably due to difficulties in measuring observed angles greater than 120 degrees. This apparent isotropic behavior is due to the random orientation of quartz grains in the sample and the very poor cleavage of quartz.

Discussion

Given the dihedral wetting angle of 60 degrees obtained in the experiments described above, some simple theoretical considerations, in combination with existing data from the literature, can be used to compute actual values for the quartz/quartz and quartz/melt interfacial energies in the experimental system. These values can subsequently be used to check for consistency between our results and previous, direct measurements of surface energies in related systems.

The first task is to estimate γ_{ss} in a quartzite, which by application of Eq. (2) allows direct estimation of γ_{st} . We are not aware of any measurements of the interfacial energy between quartz grains, but an average value can be obtained in the following way from the quartz surface energy values reported by Brace and Walsh (1962). First,

it must be recognized that the energy of the interface between two perfect crystals of exactly the same orientation and surface energy is zero. However, the interfacial energy rises rapidly with the first 10 degrees of tilt or twist along the boundary and approaches the crystal surface energy at relatively high "mismatch" angles (Dunn and Lionetti 1949; Moment and Gordon 1964; Readey and Jech 1968; Hondros and McLean 1976). In a randomly-organized aggregate of isotropic grains, less than 5% of all possible boundaries have low energy orientations (Smith 1964). In a random aggregate of *anisotropic* crystal grains with poor cleavage, the probability of finding two contacting surfaces of nearly the same crystallographic orientation is even further reduced. Therefore, to a rough approximation, the energy of the quartz/quartz interface is equal to the surface energy of quartz. This is, in fact, a quite meaningful estimate when regarded with the knowledge that most direct measurements of solid/solid interfacial and solid surface energies have uncertainties as large as ± 200 ergs/cm².

Brace and Walsh (1962) reported surface energy values of 760, 410, 500, and 1,030 ergs/cm², respectively, for the (1120), (10 $\bar{1}$ 1), (1011), and (10 $\bar{1}$ 0) crystallographic faces of quartz. Assigning equal weight to the probability of facial contact for the different faces and a random distribution of orientations, we compute an average value for the quartz surface energy of approximately 675 ergs/cm². Taking this value as the quartz/quartz interfacial energy, we see from Eq. (1) and the observed dihedral angle of 60 degrees that the quartz/melt interfacial energy (γ_{st}) is about 390 ergs/cm².

Because we have assumed that the quartz surface energy is approximately equal to the quartz/quartz interfacial energy, it must follow that the sessile-drop wetting angle ϕ , measured between the melt/vapor interface and the crystal/melt interface when a drop of melt is placed on a crystal surface exposed to a vapor or atmosphere, is approximately 1/2 the observed dihedral angle, i.e. 30 degrees. Now using

the relation:

$$\gamma_{lv} \cos \varphi = \gamma_{sv} - \gamma_{sl} \quad (3)$$

we can calculate the surface energy of the melt to be about 330 ergs/cm². This value compares favorably with estimated surface energies of chemically-similar melts. For example, in Kingery et al. (1976), we find a surface energy for a [0.80 SiO₂–0.20 Na₂O] melt at 1,350° C of 380 ergs/cm², while fused SiO₂ at the same temperature has a value of 300 ergs/cm². Assuming only a slight change in surface energy with temperature (Walker and Mullins 1981) and a linear dependence upon composition, we obtain a value for our quartz-albite melt [0.94 (SiO₂+Al₂O₃)–0.06 Na₂O] of 320 ergs/cm². This is very similar to the predicted value of 330 ergs/cm², and we consider the agreement to be excellent.

Conclusions and geologic implications

These experimental results demonstrate that a Ab₅₀Qtz₅₀ melt in chemical equilibrium with a texturally-equilibrated quartzite will not penetrate the dry grain edge lines, and that the stable configuration of this partially-molten system is one in which the melt remains in its site of generation, either in small pockets at grain corners or at grain edge intersections.

In view of the rather small dependence of all surface energies upon temperature, this result raises some questions about the relationship between equilibrium melt distribution and magma genesis in natural felsic systems. Consider, for example, a situation in which the temperature of a felsic, high-grade metamorphic rock is raised to the point where melt begins to form. The initial melt is stable as a dispersed phase at grain edge intersections or in small pockets, but not as a thin film wetting all grain boundaries. It thus appears that “external” (physical) driving forces such as gravity and non-hydrostatic tectonic stress are required to induce collection of melt into larger bodies capable of migrating toward the surface. Note, however, that once coalescence into large melt bodies has occurred, there should be no tendency toward re-dispersal by infiltration into the host rock. This behavior contrasts with that expected of mafic systems, in which wetting angles are significantly lower and infiltration can occur (Waff and Bulau 1982; Watson 1982). The end products of crustal melting anticipated on the basis of this study include both segregated felsic magma bodies (in cases where melt collection is expedited by physical influences and a high melt fraction), and migmatites (in which a low fraction of melt and residue remain closely intermingled). This duplicitous behavior is due simply to a balance of surface forces that produce an intermediate case in which there is no strong tendency for melt dispersal or for melt coalescence.

We emphasize, in conclusion, that the type of behavior just described can be regarded as strictly applicable only to the analog quartzite system studied. Melt distribution and infiltration tendency in other systems and in nature can be expected to vary with rock type, water content, impurity content, non-hydrostatic stress, oxygen fugacity, and the temperature-pressure regime involved. Nevertheless, the study has demonstrated the importance of experiments of this type, and underscores the probable contrast between the behavior of felsic, crustal magmatic systems and pre-

viously-studied mafic systems relevant to basalt generation in the upper mantle.

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