SHORT COMMUNICATIONS

A comment on crystal shapes resulting from dissolution in magmas

MUCH of the textural interpretation of igneous rocks that has evolved during the last hundred years, rather than being rooted in experiment and theory, is based on inference and handed-down opinion. This situation is changing as petrologists become more familiar with the literature of crystal growth and materials science, and as they examine the crystallization of silicate melts from the view points of nucleation, crystal growth, and textural development (see e.g. Kirkpatrick, 1975, 1980; Dowty, 1980; Lofgren, 1980, 1983). The purpose of this note is to comment on two characteristics of the shapes of phenocrysts, the interpretations of which have come to be regarded as virtually self-evident but which experiments bring into question: (1) that facets on crystals only form during growth and (2) that crystals whose surface includes curved 'faces' have experienced dissolution.

The note was provoked by a recent paper in which Nicolas and Prinzhofer (1983) concluded that 'Idiomorphism probably reflects only equilibrium between a crystal and a fluid whether the crystal is growing or dissolving'. Their evidence for breaking with petrographic convention and concluding that facets can develop during dissolution comes from partial melting experiments on a lherzolite in which reaction of the melt with residual olivine produced 'euhedral grain boundaries by oriented corrosion'. As discussed below, there is some support for this conclusion in the crystal-growth literature, where there are descriptions of experiments in which minerals have produced facets during dissolution in non-silicate melts.

(1) *Facet formation during dissolution.* In a series of papers, Franke and co-workers (Heimann *et al.*, 1970, 1971; Lacmann *et al.*, 1974; Siesmayer *et al.*, 1975) reported on investigations of the morphological effects of dissolution on various crystalline materials, among them quartz, rutile, and corundum, in a variety of non-silicate, inorganic melts. All the experiments used mineral spheres as the initial crystal form.* Depending on the composi-

tion of the melt used, crystals either remain spherical during dissolution or develop facets that range from curved (both concave and convex) to flat, the facets becoming flatter with duration of dissolution. For example, Heimann (1973) found that β-quartz spheres dissolving in $NaF-K_2Cr_2O_7$ melt at 800 °C produce a combination of hexagonal prisms and hexagonal bipyramids in one hour; on the other hand, in a KHF_2 melt the dissolution morphology remains a sphere, free of facets.

Heimann (1973) and Lacmann *et al.* (1974) show that partially dissolved spheres that develop facets have a morphology that is simply related to the growth morphology; thus the faceted dissolution morphology may be predicted by truncating the corners of the growth morphology to produce new faces (e.g. fig. 1), or put another way, the corners of a dissolving crystal correspond with the poles of faces on the crystal during growth (cf. Goldschmidt and Wright, 1903, 1904; and Schnorr, 1915). Furthermore, directions of rapid and slow *growth* on a crystal are, respectively, directions of slow and rapid *dissolution* (Bagdasarov *et al.*, 1974) which may result in dissolution habits differing substantially from the growth habits. The dissolution habit is also dependent on the melt in which dissolution occurs, probably as a result of adsorption of 'foreign' ions on faces (Heimann, 1973; Buckley, 1951).

The factor that determines whether dissolution results in a faceted morphology or a facetless sphere is the kinetics of the reaction. The rate of dissolution in any given crystallographic direction is governed by one of two processes, either (*a*) by the rate at which ions are released from the crystal lattice and incorporated in the melt, i.e. 'interface (detachment) kinetics', or (*b*) by the rate at which these ions are transported away from the crystal-melt interface and replaced by components in the bulk melt. The tacit (or unappreciated) supposition behind the conventional petrographic interpretation of crystals with curved outlines as dissolution shapes is that it is transport of components in the melt (specifically diffusion) rather than interface kinetics that governs mineral dissolution rates in silicate melts. Under such circumstances geometry

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* Use of spheres merely makes it easier to detect a change in morphology resulting from dissolution. If faceted crystals were used these would evolve to the dissolution morphology.
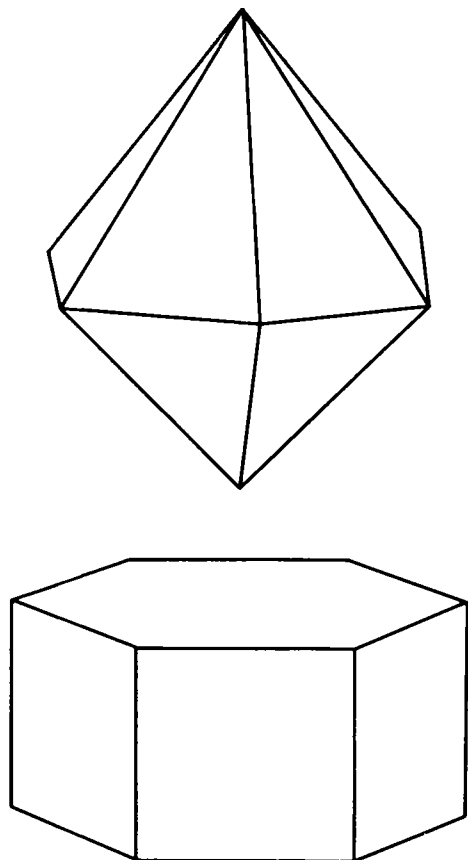
FIG. 1. Comparison of a growth morphology (upper) and dissolution morphology (lower) of β-quartz (after Heimann, 1973). The two are simply related by truncation of corners on the growth morphology.

dictates that there is more rapid dissolution at and adjacent to crystal corners and edges than in the middle of planar faces and the crystal shape evolves into one of curved outline in thin section (Wells, 1946; Smith, 1963; Correns, 1969). On the other hand, if transport in the melt is rapid compared to interface kinetics and the interface kinetics are anisotropic in different crystallographic directions, then a faceted dissolution morphology can result.

A few experiments of the kind conducted by Franke and his colleagues, but using anhydrous basalt melts as the solvent, have been reported in the petrological literature (Scarfe *et al.*, 1980; Watson, 1982; Donaldson, in prep.). In all cases, the originally spheroidal morphology of the crystals is retained as dissolution progresses (e.g. fig. 2; see also Scarfe *et al.*, 1980, fig. 18). In the author's experience, if faceted crystals with sharply angular

edges and corners are used instead of spheres, then the edges and corners tend to become rounded, though development of curvature on original faces is very variable. These results are consistent with conventional petrographic wisdom about the effect of dissolution on crystal morphology and with melt transport as the step limiting dissolution rate.

Though data on mineral dissolution rates in rock melts and simple silicate melts are scarce, those which exist support this conclusion. Thus Kuo and Kirkpatrick (1983) report that dissolution of quartz, forsterite, diopside, and enstatite in the system Mg_2SiO_4-$CaMgSi_2O_6$-SiO_2 at atmospheric pressure is governed by melt transport (both convection and diffusion); the same inference applies to data obtained on quartz, olivine, and plagioclase dissolution in a basalt (Donaldson, in prep.); likewise Watson (1982) found the rate of dissolution of quartz in a basalt melt to be limited by diffusion in the melt.

On the other hand, it is conceivable that, in rock melts in which transport of components is rapid, dissolution rate can be dictated by interface kinetics, and therefore that faceted dissolution morphologies will arise. It is in very fluid magmas that these conditions would be most likely to occur (e.g. picrites and komatiites, basalts at high pressure, and water-rich melts such as pegmatites). In this connection it may be significant that in the experiments reported and illustrated by Nicolas and Prinzhofer (1983, fig. 8*a*), in which a small proportion of faceted olivines formed in partially melted lherzolite, the melt is extremely magnesian (MgO 18–25 wt. %, unpubl. data of A. Nicholas), i.e. komatiitic. Furthermore, during melting the sample was deformed in the Grigg's apparatus and some melt migrated to the boundary between the sample and its container (platinum foil): conceivably this flux of melt facilitated removal of dissolved olivine components from crystal–liquid interfaces, thereby encouraging facets to form.

Another possible circumstance favouring dissolution facets is that in which the bulk melt is only slightly undersaturated with respect to the dissolving mineral; the driving force for dissolution will then be small and hence the rate of ion detachment from the lattice is minimal. Working with crystals of halite dissolving in water, Schnorr (1915) showed that, whereas large undersaturation conditions result in the dissolution velocities of individual faces approaching one another in value (and hence promoting spherical and elliptical crystals), small undersaturation accentuates the relative differences between the velocities and thus encourages faceting. Clearly, long-duration dissolution experiments at very small degrees of undersaturation are needed in rock melts and simple systems to test whether
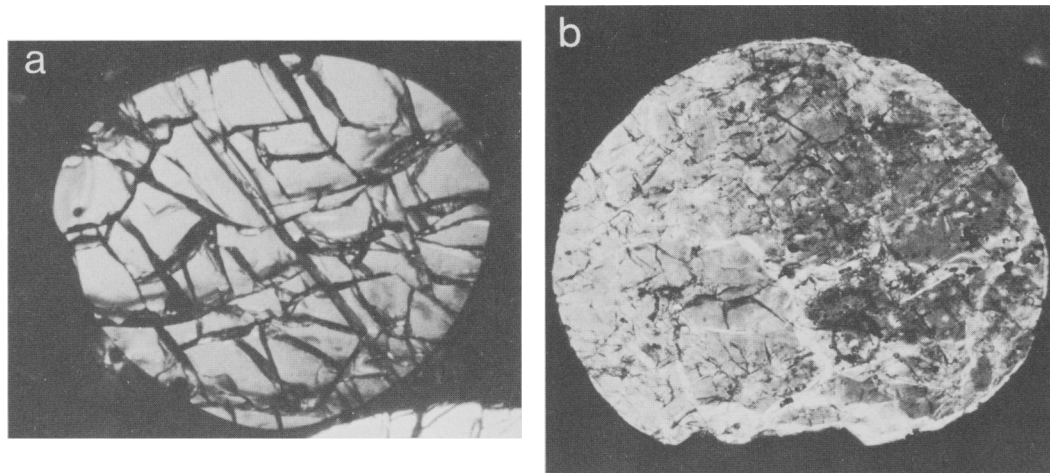
FIG. 2. Retention of spherical morphology on crystals of (a) olivine and (b) quartz following dissolution in superheated basalt melt ($+\Delta T = 52\,°C$). In the experiments a mineral spheroid of ~ 1 mm diameter was allowed to partially dissolve for 4 and 1 hours, respectively, before quenching; dissolution rates are approximately 25 and 50 μm/hr, respectively (Donaldson, in prep.). Cross-polarized light.

this finding applies to silicate minerals dissolving in silicate melts.

(2) *Curved faces formed by growth.* In the interests of balance, it is worth recalling that curved 'faces' can arise during growth of a crystal (e.g. Holmes, 1921). That this is so is most spectacularly demonstrated by Tsukamoto *et al.* (1983, fig. 4a, b). Using a new experimental technique whereby crystals may be observed in silicate melt, they have watched combinations of flat faces and (convex) 'faces' on diopside crystals growing from (a) pure diopside melt, and (b) anorthite–diopside eutectic melt, at 10–30° of undercooling. Curved and flat faces were also observed growing on anorthite crystals in anorthite–diopside melts.

The curved and flat facets must have different surface micro-topographies and probably also different growth mechanisms. The curved faces will have 'rough' surfaces and probably grew by a continuous mechanism, whereas the flat faces are more likely to have smooth surfaces and probably grew by a layer-spreading mechanism. The exact combination of planar and curved faces probably reflects the degree of supercooling during growth and the effect of impurities adsorbed on to the interface (e.g. Wells, 1946; Kohli and Ives, 1972) that disrupted layer spreading and encouraged continuous growth. It will be interesting to see whether there are silicate systems and circumstances in which crystals may grow entirely surrounded by curved 'faces'.

Conclusion. As more is learned about the growth and dissolution of crystals in silicate melts, attitudes to the petrographic interpretation of curved and planar facets on phenocrysts must become more flexible. At the same time, experiments and development of theory should allow more quantitative information to be gleaned from rock textures.

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Co-ordination of boron in sillimanite

ION-MICROPROBE analyses of six sillimanites associated with kornerupine show that the sillimanite can incorporate from 0.035 to 0.43 wt. % B_2O_3 (Grew and Hinthorne, 1983). Boron appears to substitute for silicon concomitantly with Mg substitution for Al such that the atomic Mg/B ratio is close to 0.5. This substitution results in a deficiency of cationic charge, which Grew and Hinthorne (1983) attributed to a submicroscopic rearrangement of the sillimanite structure involving loss of oxygen. A possible substitution scheme is $2(B + xMg) \rightarrow 2(Si + xAl) + (1 + x)O$, where $x \simeq 0.5$. In the present study, we have addressed the question of co-ordination of boron in sillimanite. As boron can occur in trigonal or tetrahedral co-ordination with oxygen, there is no compelling reason that B substitution for Si implies tetrahedral co-ordination for B.

Two of the boron-bearing sillimanites analysed by Grew and Hinthorne (1983), samples E 2724 and 3083D from South India (these authors' samples no. 2 and 4), and a sillimanite from a pegmatite lacking borosilicate minerals from Antarctica, sample 556 (no. 8, see also Grew, 1980; Rossman *et al.*, 1982), are in fragments suitable for single crystal infra-red studies. Spectra were obtained on (010) cleavage fragments $// c$ and $// a$ with a Perkin Elmer 180 spectrometer (fig. 1). Prominent absorption bands in the B-rich sillimanite E 2724 (0.42 % B_2O_3) are centred at 1372, 1327, 1281, 1244, and 1128

cm^{-1} in the $E // c$ polarization and 1317 cm^{-1} in $E // a$. These bands, which are less conspicuous in the boron-poor sillimanite 3083D (0.035 % B_2O_3) and are weak or absent in the Antarctic sillimanite, lie in the range 1100 to 1450 cm^{-1} in which BO_3 groups absorb (Moenke, 1974; Weir and Schroeder, 1964; Weir, 1966). Absorption due to BO_4 groups lies in the 1180 to 800 cm^{-1} region. Unfortunately, this region is largely inaccessible under our experimental conditions due to the opacity of the samples at wave numbers below 1050 cm^{-1}. We suggest that the observed spectral features between 1372 and 1128 cm^{-1} in the two boron-bearing sillimanites are due to B in threefold co-ordination. The B–O polyhedron could be either a triangle in which the B is coplanar with the three oxygens, or a highly distorted tetrahedron, in which the B lies slightly out of the plane of the three closest oxygens and is weakly bonded to a fourth oxygen extended from the boron: BO_3–O.

The absorption features appear not to be caused by submicroscopic inclusions in sillimanite of borosilicates with BO_3 groups such as grandidierite (which is present as a constituent in E 2724), tourmaline (present in 3083D), or dumortierite. M. A. Carpenter (written communication, 1983) has examined a small sample of sillimanite E 2724 with the transmitting electron microscope (TEM) and did not observe any submicroscopic features that could represent incipient exsolution of a B-rich