Appendix 4. Extended discussion

Epitaxy and twins

Epitaxy in its original meaning describes crystallographically-controlled growth of a guest crystal on a host crystal of a different mineral species (Sunagawa 2005). Homoepitaxy describes this relation between two crystals of the same mineral (e.g., quartz on quartz). Epitaxial relations are often observed in hydrothermal quartz and can result in a mutual orientation between the guest and substrate or host (e.g., scepter quartz). This is usually associated with cessation of growth and regrowth after either the supply of a new solution with different impurity concentrations, or the precipitation of a foreign coating of other minerals on the substrate or host surface (Sunagawa 2005). Both processes are associated with a significant change in the crystal growth environment. Grouped parallel quartz units in Toba rhyolites do not possess characteristics consistent with either of these processes. Cathodoluminescent images do not reveal significant differences in CL intensity (a proxy for Ti concentration) between grouped parallel units, consistent with quartz trace element data (**Table 5 of App.**[1]). Furthermore, we do not observe frequent unit attachments above old dissolution surfaces within the crystal zoning pattern. Thus, dissolution and regrowth did not promote epitaxy. Additionally, tomograms and serial sections of quartz clusters perpendicular to [0001] reveal doubly terminated habits, implying that clusters likely formed suspended in the magma, and were not derived from a larger substrate. As iterated in the main text, we suggest that if homoepitaxial growth was at all operating in the construction of quartz clusters, it must have been initiated by preferential nucleation at crystal corners or edges or by dislocations, all generated by supersaturation.

Assuming all non-parallel unit orientations are generated by simple, repeated or multi-component twinning, then a predominance of Esterel and Verespatak twins is related to their low-index twin planes, which allow them a greater ability to form (Sunagawa 2005). Even if truly "random" unit orientations exist, a predominance of twinned units is consistent with their more energetically favored positions (Buerger 1945). This may also explain a predominance of Esterel twins over Verespatak twins, as they contain the lower-index twin plane $(10\bar{1}1)$. Esterel twins have been found to occur twice as frequently as Verespatak twins in other environments (Drugman 1927).

Subordinate textural transformations

Exotic growth (anhedral → skeletal). A minority of spongy, skeletal morphologies composed of the brightest CL quartz we observe are structurally similar to 'late' growth protuberances (**Fig. 10l; Fig. 4n, s and 5l of App.**[3]). However, most are underlain by rounded, dark CL zones that evidence precursory dissolution of pre-existing crystals. Such stratigraphy indicates exposure to a quartz-undersaturated melt and subsequent growth in a melt not only quartz-supersaturated, but either richer in compatible trace elements, or at conditions promoting increased trace element solubility in quartz. Arborescent CL zones that occasionally overlie the dissolution surface signify an initial 'pulse' at the onset of renewed growth, which evolves from skeletal or fine-dendritic to spongy morphology.

Granophyric intergrowth. Quartz-alkali feldspar intergrowths plausibly represent the highest ΔT texture in the Toba rhyolites (MacLellan and Trembath 1991) (**Fig. 5j, k of App.**[3]). Optically continuous quartz transitions from a coarse to radiating texture outward from intergrowth centers. Quartz zoning can also change from complex interior zoning (with some feathery features) to non-zoned rims or rods of moderate CL intensity. Skeletal cavities and polyhedral terminations of radiating rods in contact with the matrix glass indicate growth from the host melt (**Fig. 5j, k of App.**[3]). Intergrowth characteristics are generally consistent with those produced in prolonged isothermal experiments (Fenn 1986; MacLellan and Trembath 1991), despite that those here enclose many euhedral-subhedral plagioclases and few mafic minerals.

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