

Growth mechanisms of giant crystals in pegmatites: Insights from intra-crystalline trace-element zoning

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Giant crystals (>1m long) are one of the most stunning but enigmatic unique features of pegmatites. They are thought to nucleate heterogeneously with a low nucleation density, but extremely large growth rate, from undercooled hydrous silicate magmas. Despite decades of field-based and experimental investigations, the development of large to megacrystic single crystals and their role in creating valuable mineral resources remain poorly understood. In this study we interrogate elemental zonation patterns of spodumene ($\text{LiAlSi}_2\text{O}_6$) megacrystals, to assess whether they formed at constant volumetric growth rate, as suggested by our previous experimental results. The samples were collected from lithium-rich, internally-zoned granitic pegmatites mined in South Dakota and Western Maine, USA. Regardless of provenance, spodumene was subhedral and interpreted to be among the first minerals to nucleate and grow in the pegmatite inner zones, from a highly fractionated magma.

Trace-element distribution in crystallographically-oriented sliced spodumene was evaluated using portable-XRF (p-XRF) 2D compositional mapping at 5 mm pixel size and high-resolution benchtop micro-XRF (μ -XRF) scanning at ≥ 35 mm pixel size. P-XRF concentrations of Mn, Fe, Ga, Ge, and Sn were calibrated using a matrix-matched set of reference spodumene samples, independently analyzed with LA-ICP-MS. Crystals are zoned with Fe-rich, Mn-poor, nearly homogeneous core zone (Z1), surrounded by relatively thin, Fe-poor, Mn-rich rim zone (Z2). The Z1-Z2 boundary is a skeletal, stepwise combination of (010), (100), and (110) growth faces. Z2 displays cm to mm thin, rhythmic, oscillatory growth zoning with concomitant sector zoning (Fig. 1). Typically, Fe, \pm Ti, and Ga correlate positively, and are inversely correlated to Mn and Ge. In Z1 Sn is coupled with Mn, but, in Z2 it decouples from Mn and it couples with Fe, \pm Ti, and Ga. This consistent pattern in crystals from various localities suggests a sudden change in the crystallization mechanism, possibly from undersaturated to fluid saturated magma or aqueous fluid. The relative homogeneity of cores suggests an uninterrupted proportional growth mechanism from a viscous melt. At late stages, crystallization from a medium with lower viscosity and higher diffusivity is envisaged, with coeval Fe-Mn-Sn minerals controlling the rhythmic, oscillatory behavior of trace elements.

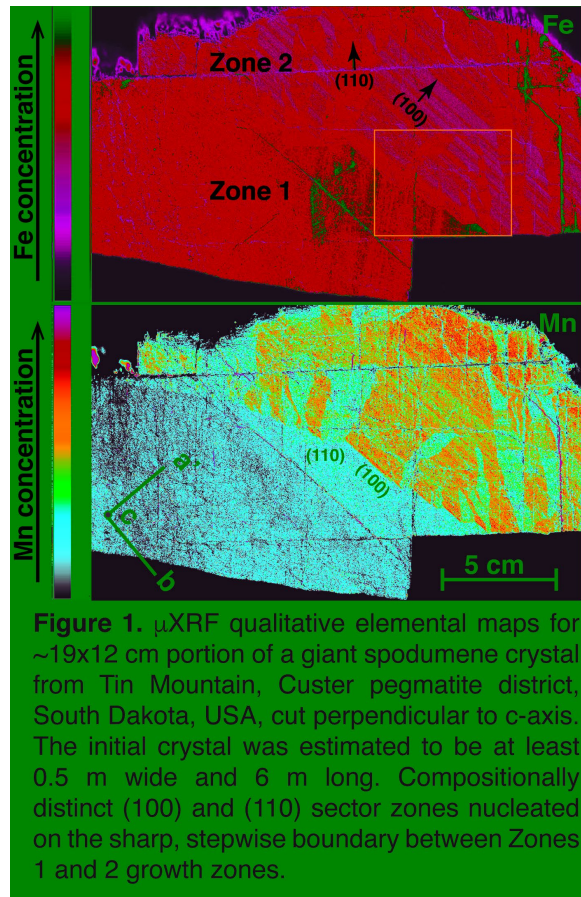


Figure 1. μ XRF qualitative elemental maps for $\sim 19 \times 12$ cm portion of a giant spodumene crystal from Tin Mountain, Custer pegmatite district, South Dakota, USA, cut perpendicular to *c*-axis. The initial crystal was estimated to be at least 0.5 m wide and 6 m long. Compositionally distinct (100) and (110) sector zones nucleated on the sharp, stepwise boundary between Zones 1 and 2 growth zones.