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Solving the SQUI (spodumene-quartz intergrowth) enigma

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Spodumene-quartz intergrowths, or SQUI, impact negatively on lithium recoveries from LCT (lithium-cesium-tantalum) pegmatites. The origin of SQUI has been long debated and, except for petalite-bearing pegmatites, has not been adequately explained. Here we report the results of an investigation of SQUI in the North American Lithium pegmatites, Québec, Canada, and provide explanations for its development. The pegmatites have a simple mineralogy comprising spodumene + quartz + albite + K-feldspar ± garnet ± muscovite/biotite ± beryl. The spodumene contains: (1) an oscillatory zoned core (bright in cathodoluminescence), with an irregular margin indicating an interruption in crystallisation; (2) an overgrowth (dark in cathodoluminescence); and (3) a SQUI zone in the overgrowth. SQUI occurs as coarse-grained or fine-grained (bright in cathodoluminescence) varieties, the latter developed at the outer edge of the spodumene overgrowth; the contact between the two SQUI types is variably abrupt or gradational. Significantly, SQUI occurs exclusively where spodumene is in contact with feldspar (K-feldspar or albite), i.e., it is absent where spodumene is in contact with quartz. We propose that these textures record the evolution of the magma to progressively higher SiO_2 activity, i.e., from a state marked by the crystallisation of the oscillatory zoned spodumene cores to one in which both spodumene and quartz were saturated and growth was rapid. We further propose that SQUI only developed in contact with feldspar because spodumene is a relatively basic mineral (low SiO_2) and the SiO_2 activity of the magma was too high for the feldspar growing adjacent to the spodumene to accommodate the excess SiO_2. This SiO_2 was therefore accommodated by co-crystallisation of spodumene and quartz (SQUI). Adjacent to quartz, however, the excess SiO_2 was taken up by additional quartz growth as this would be favoured energetically over nucleation and, thus, SQUI development would be inhibited. Finally, we attribute the fine-grained SQUI to alteration of the feldspar to spodumene by an exsolved Li-bearing hydrothermal fluid via a reaction in which Li in the fluid replaced K or Na in the feldspar, and the excess SiO_2 crystallised as quartz.