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Does silicate-borosilicate melt immiscibility occur in natural settings? An assessment based on experimentally synthesized tourmaline nodules.

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Introduction

Silicate-borosilicate (S-B) liquid immiscibility has been invoked to explain, perhaps most notably, the local concentrations of tourmaline in granites and pegmatites, as well as other phenomena in highly evolved melts.

The strongest evidence in favor of S-B immiscibility is the orbicular or nodular form that tourmaline sometimes takes within a host granite (e.g., in the Třebíč Granite, Fig. 1). These tourmaline nodules are hypothesized to form when a highly evolved melt cools and unmixes into two melts, one much richer in boron but volumetrically minor and thus tending to form spheres suspended within the relatively B-poor host melt. The B-rich melt spheres crystallise to predominantly tourmaline (e.g., Drivenes et al., 2015). However, tourmaline nodules are not strictly—nor even typically—spherical; rather, they can take a variety forms less consistent with liquid-liquid unmixing. Thus, they have alternatively been interpreted as magmatic fluid related (Sinclair and Richardson, 1992), a consequence of hydrothermal replacement (Rozendaal and Bruwer, 1995), or as primary magmatic crystallisation of tourmaline with their shape controlled by nutrient supply (Perugini and Poli, 2007).

Experimental evidence thus far has not resolved the issue. While immiscibility has been demonstrated in B-rich silicate melts (e.g., Veksler et al., 2002), these synthetic melts are also doped with other incompatible elements to unnaturally high levels, so that the effects of B alone—as well as the relevance to natural B-rich melts—are unclear. In experiments that do not include these other fluxes (i.e., where melt compositions are closer to natural granitic melts), Pichavant (1981) and van Hinsberg (2011) do not observe liquid immiscibility in B-rich melts up to 18 wt% B_2O_3 which exceeds the B-content of common natural granitic melts. This suggests B alone does not induce unmixing, yet does not explain nodular textures.

Here, we resolve these ambiguities by showing, via experimental run textures, that natural B-rich melt compositions *do not* exhibit S-B immiscibility but can exhibit nodular textures due to other mechanisms.

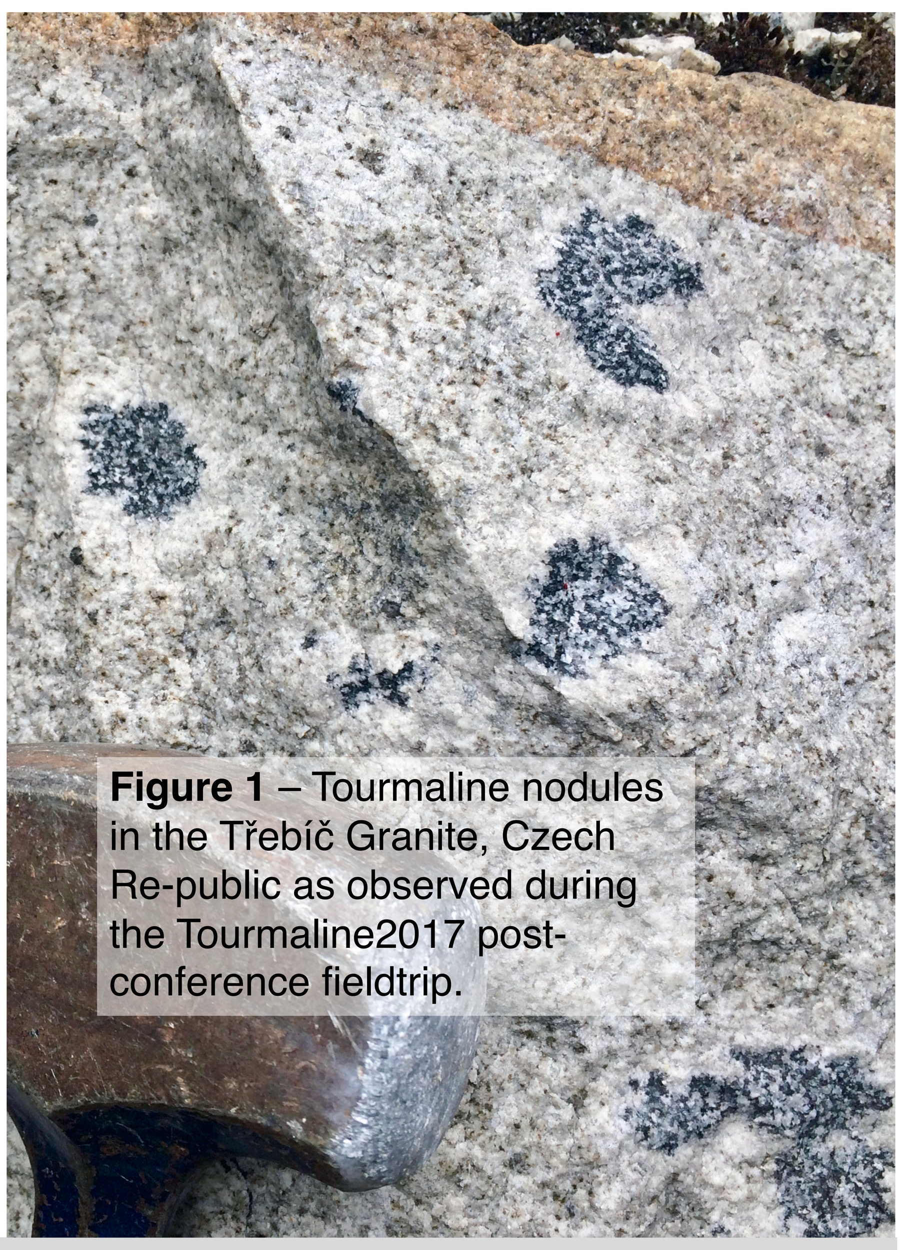


Figure 1 – Tourmaline nodules in the Třebíč Granite, Czech Re-public as observed during the Tourmaline2017 post-conference fieldtrip.

Methods

Two sets of experiments were conducted. One set used a tourmaline-free granite doped with up to 50 wt% B_2O_3 and was conducted dry (H_2O-free) at 1 atm in sealed quartz tubes in a box furnace. The second set used mixtures of nodular tourmaline and host granite from the Třebíč Granite (Fig. 1) up to 5 wt% B_2O_3 and was run H_2O-saturated at 1 kbar in sealed gold capsules in a rapid-quench cold-seal pressure vessel. In all cases, experimental charges were first homogenized to a single melt at 1200°C (dry, 1 atm) or 900°C (wet, 1 kbar). They were then cooled slowly over several days to a dwell temperature between 400 and 700°C where they were held for at least three days, and then finally quenched rapidly. Run products were imaged in BSE mode on an SEM.

Results

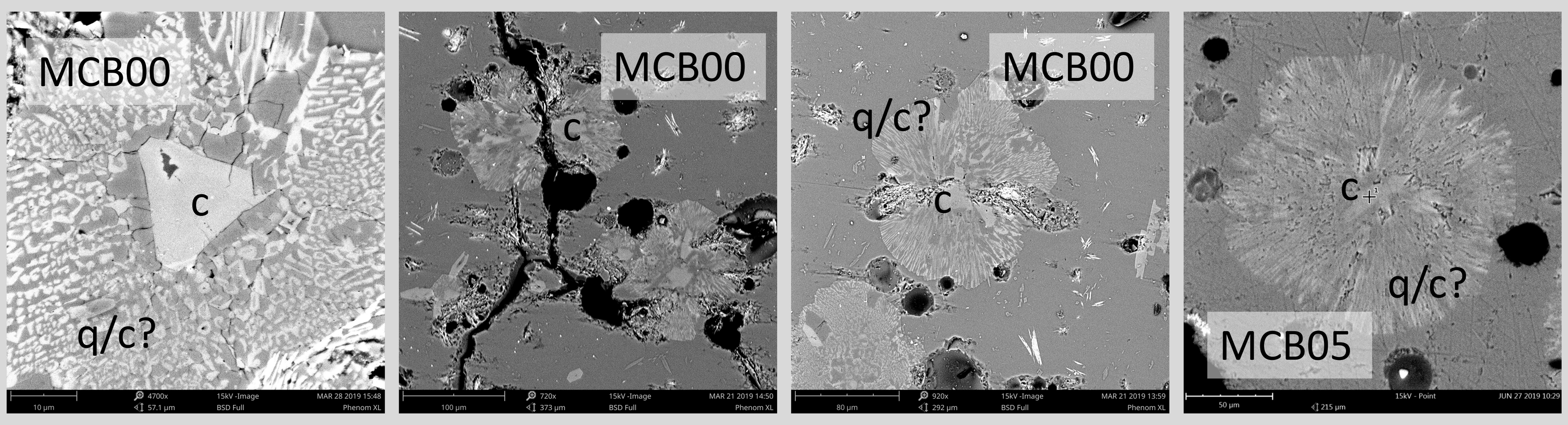
Run products from the dry 1-atm experiments exhibited no textures other than homogeneous glass (sometimes with small minor crystallites), which we do not present images of here.

Run products from the H_2O-saturated 1-kbar experiments displayed a wide variety of textures. In all cases, we observed only one glass, never two. A further key observation is the occurrence of ~100-µm nodular/orbicular structures (Fig. 2). These micronodules have symplectitic quartz and tourmaline—or cordierite for lower B_2O_3 runs—intergrown at the scale of ~2 µm, and coexisting with quench fluid, glass, and biotite. Where cordierite is present, the textures are much more equant and nodular; where tourmaline is present, a wide variety of intergrowths with quartz are observed. We also note the common spatial association of nodules and other intergrowth textures with vesicles left by the exolved vapor phase.

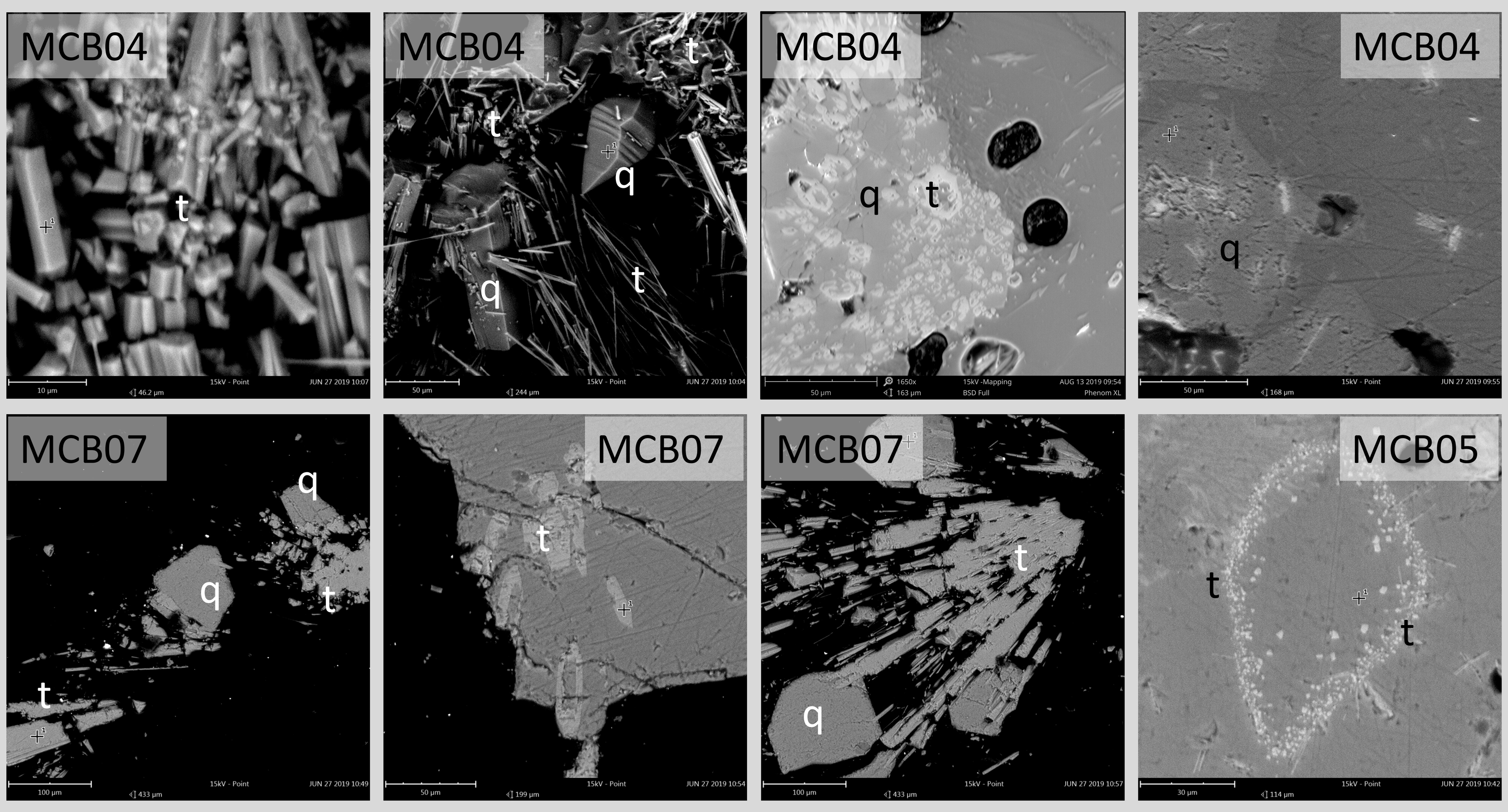
Exp	P (kbar)	T_{final} (°C)	Cooling history	B_2O_3 tot w%	H_2O tot w%	B_2O_3 glass w%	H_2O glass w%	Run products
MCB00	1	600	slow	2.22	11.53	0.79333	7.2	q, c, b
MCB04	1	625	slow, ⇅	7.29	7.85	—	12.6	q, c, t, b
MCB05	1	625	slow, ⇅	2.38	4.91	—	5.8	q, c, t, b
MCB07	1	648	fast	8.09	8.23	10.3701	7.6	q, t, b

Table 1 – Run table for the experiments the textures of which we exhibit here. Run products are: q = quartz, c = cordierite, t = tourmaline, b = biotite. Glass and fluid were also observed in all experiments, corresponding to melt and vapor at run conditions. The symbol ⇅ in cooling history indicates temperature cycling.

Figure 2 – Back-scattered electron images of mineral textures in the experimental run products. Symbols q, c, t are quartz, cordierite, and tourmaline.



Nodular textures consisting of a large cordierite grain surrounded by symplectitic/graphic intergrowths of quartz and another mineral (presumably cordierite or tourmaline?) formed in MCB00 and MCB05.



Tourmaline and quartz formed a variety of other textures as well, not as typically nodular as the cordierite/quartz nodules. In particular, large tourmaline and quartz grew freely in the vapor phase in MCB04 and MCB07 (higher B runs). Tourmaline "paisleys" were found in MCB05, although it is unclear what the three-dimensional structure of this shape was and thus difficult to speculate what it means.

Discussion

Dry 1-atm experiments showed no textural nor compositional evidence for S-B immiscibility (e.g., two glasses, one forming spherical inclusions), even at unnaturally high 50 wt% B_2O_3 contents, thus we infer that B alone (i.e., without other volatile and incompatible elements) cannot induce liquid immiscibility. This is consistent with experiments by Pichavant (1981). Textural evidence for S-B immiscibility was similarly absent in the wet 1-kbar experiments. Nodules and other crystalline textures should not be inferred to represent crystallisation of a borosilicate melt given that evidence of such a melt was not observed in higher temperature experiments either—if such a melt exists, we should be able to quench it. Furthermore, because of the presence of cordierite-quartz nodules in some runs, it is unlikely that a precursor of these B-free nodules could possibly have been an immiscible B-rich melt phase. The close physical association of nodules and other intergrowth textures with the vapor phase suggests that H_2O plays some role in their formation as well.

Thus, we hypothesize that nodules and other intergrowths formed by the following steps: (1) Temperature drops and the vapor phase exolves from the melt phase. (2) As a result, the melt (perhaps especially melt surrounding the site of vapor exolution) becomes saturated in tourmaline/cordierite, and tourmaline/cordierite begins to crystallize. (3) Growth of these mafic minerals removes mafic components from the immediate surroundings, saturating the melt with respect to quartz. (4) Quartz crystallizes, inducing resaturation of tourmaline/cordierite.

Length scales of the intergrowth could be related to differential transport rates of mineral nutrients, similar to how the nodules on Elba were interpreted by Perugini and Poli (2007). We further speculate that overall nodule size would then be related to the dwell time prior to quench, where longer times would allow for larger nodules, although this prediction would need to be tested.

At the microscopic scale, miscibility would imply that B is tetrahedrally coordinated (a network former that substitutes readily for Si) in silicate melts, while immiscibility would imply that B is trigonally coordinated (a network breaker that leads to non-bridging oxygens). Our results suggest that tetrahedral B is dominant.

We note that natural tourmaline nodules can be surrounded by a zone devoid of mafic minerals, visible in Fig. 1 as white rims around the dark nodules. If nodule growth occurs as we suggest, it would deplete the melt of mafic components immediately surrounding the nodule. By contrast, an exolved borosilicate melt is unlikely to be able to do this because transport of melt components across the higher surface energy interface would be more difficult.

Conclusion

We have demonstrated that nodular textures do not require immiscibility to form, and therefore should not be considered evidence for immiscibility. As tourmaline nodules were the strongest evidence in support of borosilicate immiscibility, our experiments lead us to conclude that B does not induce immiscibility in natural granitic systems. Tourmaline nodules are better explained by the alternative hypotheses, either as the result of a magmatic crystallisation process with local B-enrichment, or as a byproduct of the transition from magmatic to hydrothermal conditions and formation from a fluid in the cooling magmatic body.

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