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Christophe Ballouard, Yannick Branquet, Romain Tartèse, Marc Poujol, Philippe Boulvais, et al.. Nb-Ta fractionation in peraluminous granites: A marker of the magmatic-hydrothermal transition: REPLY. Geology, 2016, 44 (7), pp.e395. 10.1130/G38169Y.1 . insu-01339792

HAL Id: insu-01339792

https://insu.hal.science/insu-01339792

Submitted on 30 Jun 2016

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Forum Reply

doi:10.1130/ G38169Y.1

Nb-Ta fractionation in peraluminous granites: A marker of the magmatic-hydrothermal transition

Christophe Ballouard[1], Yannick Branquet[1,2], Romain Tartèse[3], Marc Poujol[1], Philippe Boulvais[1], and Jean-Louis Vigneresse[4]

[1]Géosciences Rennes, UMR CNRS 6118, OSUR, Université Rennes 1, 35042 Rennes Cedex, France

[2]Institut des Sciences de la Terre d'Orléans (ISTO), UMR 6113 CNRS, Université d'Orléans, BRGM, Campus Géosciences, F-45071 Orléans Cedex 2, France

[3]Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, Muséum National d'Histoire Naturelle, Sorbonne Universités, CNRS, UPMC, IRD, 75005 Paris, France

[4]Université de Lorraine, UMR 7539 GéoRessources, BP 23, F-54501 Vandoeuvre Cedex, France

We thank A. Stepanov and co-authors (Stepanov et al., 2016) for giving us the opportunity to clarify some important points made in our original manuscript (Ballouard et al., 2016) and to discuss the issues raised in their Comment. In Ballouard et al. (2016), we propose that the decrease of the Nb/Ta ratios to <~5 in peraluminous granites "is the consequence of both fractional crystallization and sub-solidus hydrothermal alteration," an interpretation challenged by Stepanov et al. (2016) who argue that low Nb/Ta ratios in peraluminous granites are better explained by magmatic fractionation and that the role of magmatic-hydrothermal processes is not significant.

In their Comment, Stepanov et al. (2016) repeatedly mention "post-magmatic" alteration, implying that the fluid-rock interaction processes we propose as responsible for the decrease in the Nb/Ta ratios occurred when the bulk of the peraluminous magmas were fully crystallized. This is not the case, and we clearly define in the first sentence of our introduction ("the magmatic-hydrothermal transition separates a purely magmatic system dominated by a crystal-melt interaction from a system dominated by a crystal–melt–magmatic fluid phase interaction") that we consider sub-solidus alteration with primary magmatic fluids/brines exsolved from crystallizing melts. Also, Stepanov et al. (2016) frequently refer to mineralized pegmatites in their Comment, which are complex objects involving, for example, undercooling processes. As stated in Ballouard et al. (2016), we took care not to include pegmatite/aplite in our database and only focused on pervasive hydrothermal activity in peraluminous granites.

Stepanov et al. (2014) demonstrated, on the basis of fractional crystallization modeling, that the fractionation of biotite and muscovite will induce a decrease of the Nb/Ta ratios in granitic melts. In Ballouard et al. (2016), we did not overlook the important role of mica fractionation in the decrease of the Nb/Ta ratios, but we show that fractional crystallization alone is not sufficient to explain the behavior of Nb-Ta in most peraluminous granites. In their Comment, Stepanov et al. (2016) argue that low Nb/Ta granites contain insufficient ilmenite to counteract the decrease in Nb/Ta due to the fractionation of micas. This is likely true. However, the authors missed the major point of our argument. The results of our modeling suggest that, even during magmatic fractionation of an ilmenite-free cumulate composed of quartz, feldspar, biotite and muscovite (micas accounting for 20% of the cumulate), unrealistic amounts of fractional crystallization (>90 wt%) are needed to significantly decrease the Nb/Ta ratio from ~8 to 2. As detailed by Ballouard et al. (2016), such amounts of fractionation cannot be reached in most of the granites compiled in our study. Moreover, we would like to underline again that small magmatic bodies like pegmatite or aplite, which can reach an extreme rate of fractional crystallization, were not included in our compilation.

Stepanov et al. (2016) also point out that low abundances of "immobile elements," such as Ti, Zr, and rare earth elements (REEs), in low-Nb/Ta granites likely resulted from fractional crystallization process. We agree with this point but we would like to specify once again that in our paper we suggested that the decrease of Nb/Ta in peraluminous granites is the consequence of both fractional crystallization and sub-solidus hydrothermal alteration.

Stepanov et al. (2016) argue that fluid-rock interactions decrease Sn concentrations in granites and that a high Sn enrichment cannot be used as a marker of magmatic-hydrothermal alteration. The simple fact that greisens (i.e., granites resulting from extreme sub-solidus hydrothermal alteration) are highly enriched in Sn (up to 1 wt% Sn; see Ballouard et al., 2016, our figure 3A) refutes this assumption.

Mineralogical evidence for Ta hydrothermal enrichment exists in ongonites, but also in several rare metal granites of Southern China where hydrothermal overgrowths of tantalite are commonly observed on magmatic columbite (e.g., Zhu et al., 2015; Xie et al., 2016). For example, Zhu et al. (2015, their figure 4a) show columbo-tantalite crystals (CGM) with a CGM-I core with a columbite composition surrounded by a CGM-II rim with a tantalite composition (including some CGM-II veinlets within the CGM-I). Few experimental studies exist on Nb and Ta solubility in aqueous solution, but Ta seems to be less soluble than Nb (e.g., Zaraisky et al., 2010). Therefore, we suggest that during magmatic-hydrothermal alteration, Ta will preferentially form overgrowths around Nb-Ta–bearing minerals, whereas Nb will be carried away by fluids, resulting in a decrease of the whole-rock Nb/Ta ratios.

At the magmatic-hydrothermal transition, granitic melts exsolve large amounts of fluids with variable compositions, ranging from aqueous to hydrosaline, having a variable capacity to transport economic lithophile elements such as Nb and Ta. Many unknowns remain regarding the behavior of Nb and Ta in such systems, warranting further experimental work on this topic. However, we maintain that it is hard to account for a decrease of the Nb/Ta ratios < ~5 in peraluminous granites by magmatic fractionation alone, and that it is likely the consequence of both fractional crystallization and magmatic-hydrothermal alteration processes.

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