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Integrated Fe and Li isotopes constrain fractional crystallization and fluid-magma interaction in the formation of unzoned Li-mineralized pegmatites

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Due to the increasing usage of lithium (Li) for batteries, Li-mineralized pegmatites, which are one of the primary sources of Li, have recently attracted significant attention. These pegmatites are generally considered to have formed from highly evolved, fluid-rich granitic melts. However, the roles of fractional crystallization and aqueous fluid–magma interactions in the genesis of unzoned Li-mineralized pegmatites remain debated. Because Fe and Li isotopes fractionate differently among melts, crystals, and aqueous fluids, their combined systematics offer a powerful means to discriminate between these processes. In this study, we report Fe and Li isotopic data for the mineralized and barren pegmatites and associated leucogranites in the Ke'eryin field, Songpan–Ganzi orogenic belt, eastern Tibet. These pegmatites and leucogranites have comparable Fe and Li isotopic compositions with δ^{56}Fe values of 0.00–0.28‰ and 0.07–0.32‰, and δ^{7}Li values of -1.5–5.4‰ and -2.0–3.3‰, respectively. In contrast to these similar isotopic compositions, pegmatites have lower Zr/Hf (< 26) and Nb/Ta (< 5) and higher $TE_{1,3}$ values (> 1.1, REE tetrad effect) than leucogranites. In addition, a negative correlation between Fe and Li isotopic compositions is observed in pegmatites from the Ke'eryin and Jiajika fields in Tibet and the Little Nahanni in Canada. These observations cannot be explained by fractional crystallization alone. We propose a two-stage evolutionary model: (1) fractional crystallization of granitic melts increases δ^{56}Fe and decreases δ^{7}Li in evolved melts, accompanied by decreases in Nb/Ta and Zr/Hf; (2) fluid–magma interaction after the emplacement of pegmatitic melts decreases δ^{56}Fe and increases δ^{7}Li, driving Zr/Hf, Nb/Ta, and $TE_{1,3}$ toward values typical of magmatic-hydrothermal systems. Fluid-magma interaction therefore counteracts isotopic fractionation induced by fractional crystallization, resulting in the similar Fe–Li isotopic compositions between pegmatites and leucogranites. The negative Fe–Li isotopic correlation in pegmatites is consistent with a quantitatively constrained mixing model governed by fluid-magma interaction. Overall, this study shows that coupled Fe-Li isotopic systematics effectively distinguish the effect of fluid-magma interaction from fractional crystallization during the formation of unzoned pegmatites.