

## **Lithium isotopic systematics of granites and pegmatites from the Black Hills, South Dakota**

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### **ABSTRACT**

To study Li isotopic fractionation during granite differentiation and late-stage pegmatite evolution, Li isotopic compositions and concentrations have been measured for the S-type Harney Peak Granite, the spatially associated Tin Mountain pegmatite, and possible metasedimentary source rocks in the Black Hills, South Dakota. The Harney Peak Granite is isotopically heterogeneous, with  $\delta^7\text{Li}$  varying from  $-3.1$  to  $+6.6$ . The  $\delta^7\text{Li}$  values of Proterozoic metasedimentary rocks that are possible sources of the Harney Peak Granite range from  $-3.1$  to  $+2.5$  and overlap with post-Archean shales and the Harney Peak Granite. For the granite suite, there is no correlation between  $\delta^7\text{Li}$  and elements indicative of degrees of granite differentiation ( $\text{SiO}_2$ , Li, Rb, etc.). The Li isotopic composition of the Harney Peak Granite, therefore, appears to reflect the source composition.

Minerals from the zoned Tin Mountain pegmatite have extremely high Li contents and heavier Li isotopic compositions than the granite or surrounding Black Hills metasedimentary rocks. The heavier compositions may reflect Li isotopic fractionation resulting from extensive crystal-melt fractionation. Lithium concentrations decrease in the order: spodumene ( $\sim 3.7$  wt%), muscovite (0.2 to 2.0 wt%), plagioclase (100–1100 ppm), quartz (30–140 ppm). Plagioclase, muscovite, and spodumene in all zones display a relatively narrow range in  $\delta^7\text{Li}$  of  $+7.9$  to  $+11.4$ . In contrast, quartz is isotopically heavier and more variable ( $+14.7$  to  $+21.3$ ), with  $\delta^7\text{Li}$  showing an inverse correlation with Li concentration. This correlation reflects the mixing of isotopically heavy Li in quartz and lighter Li in fluid inclusions, as documented by fluid inclusion compositions ( $\delta^7\text{Li} = +8.1$  to  $+13.4$  and Li of 280 to 3960 ppm). Extrapolation of this trend to an estimated intrinsic Li concentration in quartz of  $<30$  ppm, yields an inferred  $\delta^7\text{Li}$  for fluid inclusion-free quartz of  $>+21$ . The large difference in  $\delta^7\text{Li}$  between quartz and other minerals may reflect  $^7\text{Li}$  preference for less highly coordinated sites, which have higher bond-energies (i.e., the two- or fourfold site in quartz vs. higher coordination number sites in other minerals). Comparison of the Li isotopic composition of fluid inclusions with that of the wall zone of the Tin Mountain pegmatite suggests  $\sim 4\%$  isotopic fractionation during fluid exsolution, which agrees with the results derived from studies of hydrothermal alteration of basalts.

**Keywords:** Stable isotopes, igneous petrology, pegmatites, fluid phase, lithium, Harney Peak Granite, Tin Mountain pegmatite, isotope fractionation