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Kinetic Fractionation of Lithium Isotopes During Diffusion in Water

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The mass-dependent fractionation of isotopes can occur during a number of processes, only some of which are well understood theoretically and well investigated experimentally. In particular, the kinetic fractionation of isotopes during diffusion, though understood for gaseous systems, is not well understood for condensed systems such as liquid water or silicate melts. As a compliment to diffusion experiments in silicate melts [1], we undertook experiments in aqueous solutions to better understand the phenomenon of kinetic fractionation during diffusion.

Diffusion experiments consisted of a small volume glass flask containing a LiCl solution immersed in a larger (~300x) container of high purity water. The flask communicates with the larger surrounding volume via a thin tube. Flask-container pairs were set up and allowed to exchange for times ranging from 31 to 99 days. At the end of an experiment, the concentrations and isotopic compositions of Li remaining in the flask and built up in the container were measured. ⁷Li/⁶Li measurements were made by MC-ICPMS (IsoProbe).

The longest duration experiment left 1.5% of the original amount of Li in the flask, producing a change of 9 per mil in the residual Li relative to the starting solution. Taking all of the experimental runs together, a coherent relationship is observed between ⁷Li/⁶Li and F, the fraction of Li remaining in the flask. In kinetic fractionation theory, the ratio of the diffusivities of two isotopes is proportional to the inverse ratio of the isotopic masses raised to the power of a factor, β . Observed ⁷Li/⁶Li fractionation during diffusion in silicate melts was ~10x greater [1]. Though the aqueous data can be modeled with a β much less than the β (=0.215) for Li diffusion in silicate melt, an alternative is to consider that Li⁺ diffuses as a hydrated complex. With a mean hydration sphere of four water molecules [2], a β of 0.2 can be used to model the data. Isotopic fractionation during diffusion of Li across a membrane [3] is also significantly larger than diffusion in water.

References

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- [3] Fritz S.J. (1992) *GCA* **56**, 3781-3787.