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Electrochemical Fractionation of Transition Metal Stable Isotopes

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Transition metal stable isotopes are potentially valuable markers of the current state and evolution of the Earth and other planets. However, the interpretation of geochemical signatures requires a quantitative understanding of the physical and chemical mechanisms for isotope fractionation. Here we show that the redox process itself may be responsible for large isotope separations in transition metals, similar to the magnitude observed in some biotic fractionation processes We present a theory based on statistical mechanics of charge transfer reactions that predicts a driving-force dependent stable isotope fractionation in aqueous redox systems. This theory is bolstered by potentiostat-based experimental studies of voltage-dependent isotope fractionation of iron (Kavner et al., Geochim. Cosmochim. Acta 2005) and Zn (Kavner et al., in review). We show new electrochemical and analytical data in both the Fe and Zn systems providing tests of some of the predictions of the electrochemical isotope fractionation theory, including temperature dependence, and mass transport effects.