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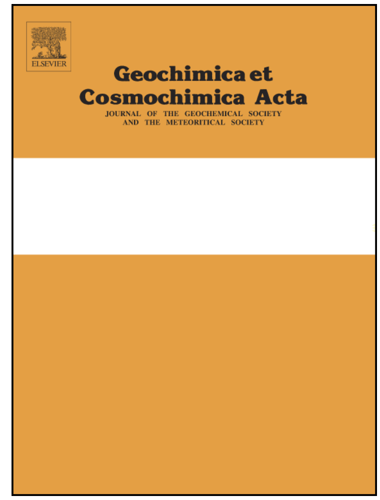

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EXPERIMENTAL CALIBRATION OF CLUMPED ISOTOPE REODERING IN DOLOMITE

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Abstract:

Dolomite clumped isotope compositions are indispensable for determining the temperatures and fluid sources of dolomitizing environments, but can be misleading if they have modified since formation. Carbonate Δ_{47} values are susceptible to resetting by recrystallization during diagenesis, and, even in the absence of dissolution and reprecipitation reactions, alteration by solid-state reordering during prolonged residences at elevated temperatures. In order to understand the potential of dolomite Δ_{47} values to preserve the conditions of dolomitization in ancient sections, the kinetic parameters of solid-state reordering in this phase must be determined. We heated mm-sized crystals of near-stochiometric dolomite in a René-type cold seal apparatus at temperatures between 409 and 717 °C for 0.1 to 450 hours. In order to prevent the decarbonation of dolomite to calcite, periclase, and CO_2 at these conditions, the system was pressurized with CO_2 to 0.45–0.8 kbar. Over the course of 31 temperature-time points and 128 individual Δ_{47} measurements of powdered dolomite crystals from these points, we observed the evolution of dolomite Δ_{47} values from the initial (unheated) composition of the crystals (0.452 ± 0.004‰, corresponding to a formation temperature of ~145 °C) towards high-temperature equilibrium distributions. Complete re-equilbration occurred in the 563 to 717 °C experiments. As with previous heating experiments using calcite and apatite, dolomite Δ_{47} exhibited complex reordering behavior inadequately described by first-order Arrhenian-style models. Instead, we fit the data using two published models for clumped isotope reordering: the transient defect/equilibrium defect model of Henkes et al. (2014), and the exchange-diffusion model of Stolper and Eiler (2015). For both models, we found optimal reordering parameters by using global least-squares minimization algorithms and estimated uncertainties on these fits with a Monte Carlo scheme that resampled individual Δ_{47} measurements and re-fit the dataset of these new mean values. Because the exact Δ_{47}–T relationship between 250 and 800 °C is uncertain, we repeated these fitting exercises using three published high-temperature Δ_{47}–T calibrations. Regardless of calibration choice, dolomite Δ_{47} rate constants determined using both models are resolvably slower than those of calcite and apatite, and predict that high-grade dolomite crystals should preserve apparent equilibrium blocking temperatures of between ~210 and 300 °C during cooling on geologic timescales. Best agreement between model predictions and natural dolomite marbles was found when using the exchange-

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