

Experimental constraints on H diffusion in clinopyroxene at low temperatures (195 – 400 °C): Implications for re-equilibration at shallow depths

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Hydrogen in nominally anhydrous minerals (NAMs) is of significant interest due to its substantial impact on various properties even when present in trace amounts (e.g., melting and transport properties). Previous studies [1,2,3] investigated H diffusion in clinopyroxene at temperatures > 600 °C, but fast diffusion of hydrogen (compared to other elements) might allow re-equilibration of H contents at lower temperatures, as well. This would have major consequences for applications such as ascent chronometry or the interpretation of H contents of NAM as a mantle signature. Experimental data at temperatures < 600 °C have been lacking due to experimental and analytical difficulties. We overcame these by implanting hydrogen in single crystals of natural diopside to induce concentration gradients and combined this with the non-destructive analytical tool of Nuclear Resonance Reaction Analysis to measure nanoscale diffusion profiles that result from low temperature experiments (195, 270, and 400 °C) at non-oxidizing conditions. Concentration profiles were fitted by solving the diffusion equation using a finite difference scheme (fig.1). The Arrhenius relation obtained from the extracted diffusion coefficients is $D_H = 5.47 (\pm 13.98) \cdot 10^{-8} \exp(-115.64 (\pm 11.5) \text{ kJ mol}^{-1} / RT) \text{ m}^2 \text{ s}^{-1}$. Our experimental results are in agreement with extrapolations from HT-experiments in previous studies (fig.2). We simulated the ascent of crystals of different sizes (0.5, 1.0, and 2.0 mm) along two geotherms (an oceanic and a continental type) from 600 to 100 °C in a non-isothermal diffusion model using the newly obtained diffusion rates. The boundary conditions were set to consider the changing solubility of OH⁻ in clinopyroxene as a function of P-T/ fO_2 . The model results indicate that a fast ascent (e.g., kimberlitic magma) leaves original hydrogen contents in the core of even small crystals (0.5 mm) unaltered but residence at low temperatures (e.g., serpentinization) for thousands of years substantially modifies initial H contents even in large crystals (2.0 mm). Thus, clinopyroxene water contents are susceptible to modification by long-lived low-temperature geologic processes.

[1] Ferriss, E.; Plank, T.; Walker, D. (2016) Contrib Mineral Petrol 171

[2] Hercule, S.; Ingrin, J. (1999) AmMin 84

[3] Woods, S.C.; Mackwell, S.; Dyar, D. (2000) AmMin 85

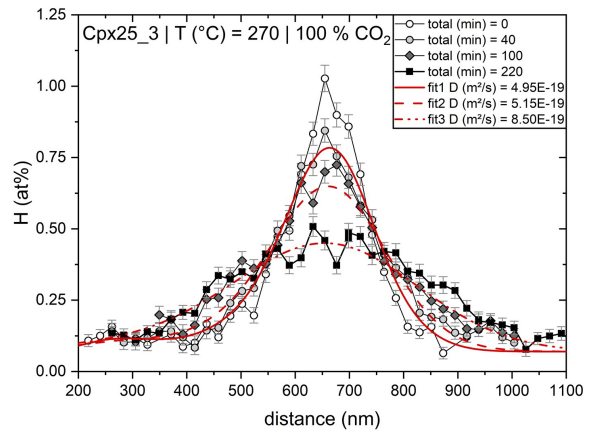


Figure 1: Taking advantage of the non-destructive nature of Nuclear Resonance Reaction Analysis allowed us to perform time-series diffusion experiments within the same sample (Cpx25_3). Obtained diffusion coefficients from best fits are consistent over time.

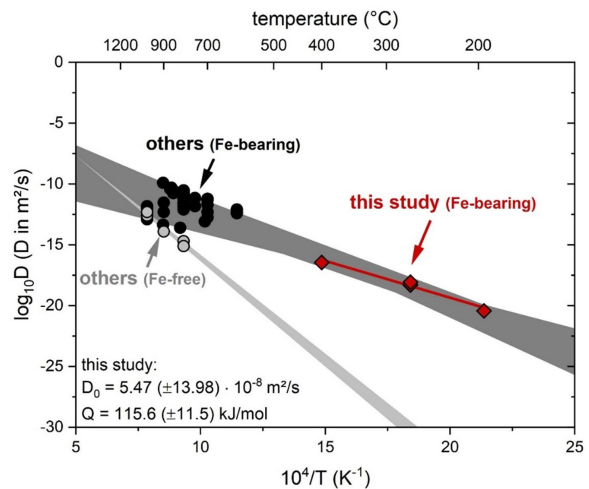


Figure 2: Our data (red diamonds, 5 experiments) lie well within the range that is predicted by extrapolation of diffusion coefficients of H in Fe-bearing clinopyroxene from high temperatures ($T \geq 600$ °C, black filled circles).