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Mineral alteration in ammonia-water solutions

AMBER ZANDANEL[1], ROLAND HELLMANN[2] AND LAURENT TRUCHE[3]

[1]Université Grenoble Alpes
[2]ISTerre
[3]ISTerre, Université Grenoble Alpes

Presenting Author: azandanel@gmail.com

Introduction: The identification of a liquid ocean beneath the ice crust on Saturn's small moon Enceladus has kindled interest in low-temperature water-rock interactions as a driver in developing habitable worlds [1], [2]. The preservation of NH_3 and salts that depress the freezing point of water in small, cold satellites suggest that the heat generated from exothermic mineral dissolution at below-0 °C could even initiate the ice melting processes that create liquid oceans [3]. However, numerical models of mineral dissolution in these environments often rely on thermodynamic and kinetic data extrapolated from 25 °C or above.

Methods: Batch experiments combined San Carlos olivine ($Mg_{1.83}Fe_{0.17}SiO_2$) with NH_3-H_2O solutions, reacted at -20, 4, and 22 °C for up to 442 days. Solution chemistry changes were monitored at intervals to evaluate mineral alteration over time. The ice-fluid-mineral interface was characterized using Raman spectroscopy and geochemical modelling. In addition, olivine surfaces were analyzed at the nanoscale with TEM microscopy.

Results: Changes to fluid chemistry over time show that olivine dissolution occurs even in partially frozen solutions. Initial evolution of concentrations of Si and Mg show a surprisingly weak dependence on temperature, pH, and NH_3, while long-term rates show an apparent weak inverse relationship with temperature. Our findings imply that olivine dissolution is not significantly retarded at -20 °C compared to 22 °C. In addition, high-resolution TEM analysis of the olivine surface after 442 days of reaction shows a thin (< 1 nm) altered layer at the olivine surface, unambiguously demonstrating that secondary reaction products can form in partially frozen solutions even at experimental timescales. These findings are applicable to evaluating mineral weathering processes in frozen or partially-frozen icy worlds such as Enceladus, (1) Ceres, or Uranus' moon Ariel.

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References:

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