

Experimental investigations of the N₂ production from NH₃-bearing hydrothermal fluids in Titan and other large icy worlds

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Abstract

In order to test the hypothesis of an enrichment in N₂ of the atmospheres of large icy worlds (Titan, Pluto or Triton) from hydrothermal processes involving NH₃-rich fluids, experiments on chondritic-like materials are carried out at 400, 600 and 800°C and 1GPa in piston cylinder in presence of NH₃-rich fluids. Preliminary results highlight the production of N₂ as well as the possible existence of a maximum of this production for fluids moderately enriched in NH₃ (around 5 wt.%). Concomitant mineral transformations are dominated by the production of talc. Experiments to better constrain N₂ production and detailed investigations on formed products are in progress and will be presented at the conference.

1. Introduction

The low ³⁶Ar/N₂ ratio in Titan's atmosphere as measured by the Huygens GCMS (Niemann et al. 2010) indicates that nitrogen was not brought in the form of molecular N₂ on Titan but rather in the form of more condensable N-bearing molecules, such as NH₃ and N-bearing organics (Tobie et al. 2012, Miller et al. 2019), and was then converted *in situ*. A variety of exogenous processes has been proposed in the literature, but they have difficulty to explain the amount of N₂ observed in Titan's atmosphere today (e.g. Marounina et al. 2015, 2018). An alternative scenario could be that N₂ was converted from NH₃ or N-bearing organics in the interior (Tobie et al. 2012, Glein et al. 2015, Miller et al. 2018). Similar internal process could also be invoked to explain Pluto and Triton's N₂-dominated atmospheres. In order to constrain the possible production of N₂ from the interaction of NH₃-enriched fluids with rock minerals in the context of large icy worlds (Titan, Pluto, Triton), we carried out hydrothermal alteration experiments of olivine and orthopyroxene (main

constituents of ordinary chondrites) in the presence of NH₃-rich aqueous fluids.

2. Experimental methods

The experiments are conducted in piston-cylinder apparatus at 400, 600 and 800°C and 1GPa for approximately 96 hours. The initial material is a mixture in equal mass proportions of natural samples of olivine (Fo₉₁, from San Carlos xenolith, US-AZ) and orthopyroxene (En₉₀, from Maar de Borée xenolith, France) finely crushed (<50 μm). Starting powders are loaded into Pt capsules with demineralized water (resistivity 15 Ω.cm) enriched in dissolved NH₃ to obtain a fluid/rock mass ratio of 5. Concentrations of 0, 5 and 10 wt.% of NH₃ are investigated at each temperatures. At the end of the experiments, the recovered capsules are perforated, and the released gases are collected and analysed with a gas chromatograph for quantifying the amount of produced N₂. The remaining fluid and solid products are then recovered and separated for further analysis.

3. Experiment results

Preliminary results show a significant production of N₂ from experiments with an initial concentration in NH₃ of 5 wt.%, as well as an increase in this production with temperature. At 600°C a molality (*m*) of 0.287 mol.kg⁻¹ of N₂ is measured. At 800°C, *m*=1.097 mol.kg⁻¹. By determining a maximum amount of N₂ from a total conversion of NH₃ to N₂, we can estimate the N₂ produced corresponds to 18.2% of this maximum at 600°C and 69.8% at 800°C. Unexpectedly, an increase in the initial NH₃ concentration (10 wt.%) results in a drop in N₂ production. Thus at 600°C, the molality of NH₃ only reaches 0.051 mol.kg⁻¹, which corresponds to 1.4% of the maximum that can be expected. This suggests

that there is an optimal in the N₂ production for moderately concentrated NH₃ fluids. Additional experiments are currently conducted to understand the origin of this behavior.

Preliminary Raman spectroscopy analyses of solid products obtained at 600°C mainly show the formation of talc (Mg₃Si₄O₁₀(OH)₂) at the expense of orthopyroxene. This is particularly true in the absence of NH₃, where the talc fraction is high while the orthopyroxene fraction has decreased significantly. For initial NH₃ concentrations of 5 wt.% this process is less marked. A systematic solid phase analysis by Raman spectroscopy, but also by SEM and DRX is currently in progress. Additional results will be presented at the conference.

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