## Investigating and quantifying natural H<sub>2</sub> emissions within the Fe-rich Kansas Precambrian granitoid crust, through high resolution microscopy and tomography

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Natural hydrogen ( $H_2$ ) generation has long been believed to only occur during serpentinization of ultramafic to mafic rocks at mid-oceanic ridges. Nevertheless, several studies have shown that  $H_2$  could also be emitted in the middle of continents, within ancient cratonic basements. Here we report the study of drill cores from the  $H_2$ -emitting DR1-A well (Kansas, USA). In this area, the Precambrian granitoid basement rich in ferromagnesian minerals produces high levels of  $H_2$ .

Petrographic observations of samples from a depth of 452 m within the basement show fractured olivine distributed in a matrix of amphibole, pyroxene, feldspar, quartz, and oxides. As revealed by SEM, these Fe-rich olivines (fayalite, Fe<sub>2</sub>SiO<sub>4</sub>) are crosscut by veins filled with two types of phyllosilicates, associated with iron oxides. XANES spectroscopy at the Fe Ledge showed that these phases contain both ferrous and ferric iron: the external part of the veins exhibits almost 30% of ferric iron versus only 20% for the center part. Transmission electron microscopy investigations conducted on ultrathin FIB sections extracted across these veins show Fe-phyllosilicates structurally related to chlorites. Although rare in such geological context, the presence of fayalite and chlorite with a heterogeneous but significant ferric iron content strongly suggests water-rock interactions leading to the local generation of H<sub>2</sub>.

However, an alternative explanation would be the trapping of  $H_2$  by the Fe-rich phyllosilicates composing the external veins and having crystallized at high temperature. In fact, as shown by XCT, SEM-EDS,  $\mu$ Raman and NanoSIMS, the external veins are restricted to fayalites crystals while the central veins propagate

throughout the entire matrix, strongly suggesting a two-stages history. The further interactions of the external phyllosilicates with subsequent infiltrating fluids could have been responsible for the release of molecular hydrogen. Post-processing of XCT data combined with XANES data allow to estimate the volume fractions of those Fe(III)-rich phases, and thus to quantify the potential H<sub>2</sub> production associated to those phyllosilicates. The present findings have important implications for exploring similar Precambrian granitoids present in every continent that might correspond to potential natural H<sub>2</sub> production sites.

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