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MRS 2023 – 47<sup>th</sup> Scientific Basis for Nuclear Waste Management (SBNWM)

## In situ Raman monitoring of studtite formation under alpha radiolysis with <sup>18</sup>O isotopic labeling

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### Abstract

During interim storage of fuel assemblies in pools the prospect of a through-wall cladding defect must be taken into consideration. The pool water is subjected to a strong radiation field producing oxidizing species, the main molecular species of which is hydrogen peroxide. When brought into contact with the fuel, hydrogen peroxide generated by water radiolysis leads to the precipitation of studtite (UO<sub>2</sub>)(O<sub>2</sub>)(H<sub>2</sub>O)<sub>2</sub> at the surface of the fuel. This phase having a density lower than that of the fuel can induce a worsening of the defect and thus impact the mechanical strength of the rods. It is therefore important to understand their formation mechanisms at the surface of the spent fuel.

In order to assess this problem, an experimental approach aiming to understand its formation mechanism in the presence of alpha radiolysis of water was conducted. Thus, an <sup>18</sup>O-labeled solution was irradiated with He<sup>2+</sup> ions through a UO<sub>x</sub> target, itself leached by the radiolysed solution. An in-situ Raman monitoring of the studtite precipitation kinetics was carried out. The use of <sup>18</sup>O-labeled water was chosen, in order to distinguish the air oxygen dissolved in the solution from that contained in the water.

The absence of the presence of <sup>16</sup>O in the peroxide bond added to the results of the Chemsimul software, reveals that contrary to gamma radiolysis, the oxygen dissolved in the water does not intervene in the mechanism of production of studtite in alpha radiolysis. We deduce that the process of H<sub>2</sub>O<sub>2</sub> formation is the result of the primary radiolytic yield during the heterogeneous chemistry step by the OH<sup>o</sup>+ OH<sup>o</sup> recombination. This work teaches us that the mechanisms of H<sub>2</sub>O<sub>2</sub> production in aerated water, leading to the peroxide bonds of studtites, are dependent on TEL effects.