



HAL
open science

In situ Raman monitoring of studtite formation under alpha radiolysis with ^{18}O isotopic labeling

Aurélien Perrot, Aurélien Canizares, Sandrine Miro, Laurent Claparede, Lola Sarrasin, Renaud Podor, Sylvain Peugot, Christophe Jegou, Nicolas Dacheux

► **To cite this version:**

Aurélien Perrot, Aurélien Canizares, Sandrine Miro, Laurent Claparede, Lola Sarrasin, et al.. In situ Raman monitoring of studtite formation under alpha radiolysis with ^{18}O isotopic labeling. MRS 2023 – 47th Scientific Basis for Nuclear Waste Management (SBNWM), Nov 2023, Cologne, Germany. cea-04650923

HAL Id: cea-04650923

<https://cea.hal.science/cea-04650923>

Submitted on 17 Jul 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

MRS 2023 – 47th Scientific Basis for Nuclear Waste Management (SBNWM)

In situ Raman monitoring of studtite formation under alpha radiolysis with ¹⁸O isotopic labeling

Aurélien Perrot^{1,*}, Aurélien Canizares², Sandrine Miro¹, Christophe Jegou¹, Laurent Claparede³, Renaud Podor³, Sylvain Peugeot¹, Nicolas Dacheux³

¹CEA, DES, ISEC, DPME, Univ Montpellier, Marcoule, France

²CEMHTI, Univ d'Orléans, Orléans, France

³CEA, CNRS, ENSCM, ICSM, Univ Montpellier, Marcoule, France

*aurelien.perrot@cea.fr

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 (UO₂)(O₂)(H₂O)₂ 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 ¹⁸O-labeled solution was irradiated with He²⁺ ions through a UO_x target, itself leached by the radiolysed solution. An in-situ Raman monitoring of the studtite precipitation kinetics was carried out. The use of ¹⁸O-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 ¹⁶O 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 H₂O₂ formation is the result of the primary radiolytic yield during the heterogeneous chemistry step by the OH^o+ OH^o recombination. This work teaches us that the mechanisms of H₂O₂ production in aerated water, leading to the peroxide bonds of studtites, are dependent on TEL effects.