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View Abstract

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ABSTRACT

TITLE: Gamma radiation effect on UO_2: set-up flow-through dissolution experiments.

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ABSTRACT BODY:

Abstract Body: In most countries, the long-term disposal of spent nuclear fuel (SNF) into a deep geological repository (DGR) is a radioactive waste management policy. Assuming a container failure, once groundwater contact with SNF, the potential radionuclide release is a key issue to understanding. Solubility and dissolution rate of radionuclides are strongly dependent on UO_2(s) matrix dissolution since upon water exposure. The SNF solubility depends on a variety of factors (intrinsic and extrinsic): redox, pH, temperature, radiolysis, aqueous system composition among others. Equally, the U dissolution behavior is clearly determined by its redox state. In this scenario, oxidized U in aqueous systems will be stabilized as UO_2^{2+} (hexavalent U), as a consequence of tetravalent U oxidation, *e.g.* oxidizing species as radiolytic H_2O_2. Uranyl cationic species in different speciation forms are expected to be found at a wide pH range. The importance of solubilized UO_2^{2+} lies in its potential incorporation of trace radioelements onto secondary uranyl phases. The influence of radiation emitted by the fuel is an additional parameter that directly affects the dissolution rate of the fuel matrix. When groundwater is subjected to radiolysis by ionizing radiation particles (α-particles, β and γ) from nuclear fuel, generate H_2O_2 and H_2 at much higher rate than that of other radiolysis byproducts [1]. Moreover, the yield of the various radiolytic products varies with the radiation type, its energy, and time [2]. Gamma radiation emitted from the fission and neutron activation products will go across long distances from the fuel surface and even go through the cladding and the inner surface of cask [2]. In young spent fuel, γ radiation plays a significant role in the total dose rate under temporary or final disposal because of short-lived decay products of irradiated SNF.

In order to know the effect of this radiation in the fuel stability at DGR conditions, leaching experiments were performed on UO_2 pellets in the presence of an external gamma irradiation source using well-controlled flow-through dissolution experiments at different low flow velocities under oxidizing conditions. Firstly, in order to verify the set-up (the design of the flow-through reactor) and different flow rates (3.2 ± 0. 2; 30 ± 10 and 90 ± 20 $\mu L x min^{-1}$) and residence times of the leachants in the flow cell, single-pass flow-through experiments are performed at room temperature in the absence of a radiation at I = 0.02 M of ionic strength as a function of bicarbonate concentration (0, 0.001M $NaHCO_3$ and 0.019 M $NaHCO_3$). Ionic strength was stablished by using $NaClO_4$ (no bicarbonate) or NaCl (bicarbonate). A good agreement of data and trends (U concentration *vs* pH, $[HCO_3^-]$, residence time and flow rates) is obtained for the experimental set-up and procedure. The results show the expected impact of carbonates on the dissolution rate of U, *i.e.* the concentration of dissolved U(VI) increased with increasing $[HCO_3^-]$ at oxic conditions [3]. Then, experiment in presence of intense γ-radiation field (dose rate of 3.9 $mGy x s^{-1}$), equivalent to a cooling time of 30 years for a 40 $MWd x kg^{-1}$ fuel, was carried out in absence of bicarbonates (I = 0.02 M, $NaClO_4$ solution) at 95 ± 20 $\mu L x min^{-1}$ flow velocity. It was observed the formation of H_2O_2 in the leachate as well as the formation of a secondary phase identified as metastudtite, on the surface of the UO_2 pellet. This uranyl secondary phase was identified by both, XRD and Raman spectroscopy.

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KEYWORDS: Properties/Radiation/radiation effects, Composition & Microstructure/Chemical Element/U, Composition & Microstructure/Material Type/oxide.