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(G*) Effects of irradiation on surface oxidation states of UO(2+x)

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γ and β radiation emitted from fission and activation products in the UO_2-fuel matrix decay to insignificant levels after 1000 years, leaving particles as the primary source of radiation. radiation induces radiolysis of water, a well-known key contributor to the oxidative dissolution of the UO_2-fuel matrix. Extensive studies have been conducted to investigate the effect of water radiolysis on fuel dissolution in the unlikely event of used fuel container failure. In contrast, this study explores the direct impact of residual radiation on the solubility of uranium fuel in solid state. Controlled doses of radiation (at 40 keV and 3000 keV) are applied to uranium fuel to investigate nuclear and electronic interactions near the surface and bulk for varying irradiation damage (in DPA). The goal is to replicate the hypothetical tailed radiation dose rate expected for uranium fuel in deep geological repositories (DGR) simulated for 1000 years; and investigate possible effects of the irradiation damage on the uranium fuel in solid state. X-ray photoelectron spectroscopy (XPS) analysis was used to track changes in $UO_{(2+x)}$ oxidation states before and after irradiation. The results reveal a reduction of $UO_{(2+x)}$, with an increased percentage of U(IV) states alongside reduced percentages of U(V) and U(VI) states. Our findings suggest that prolonged exposure of uranium fuel to radiation in simulated DGR conditions, without container failure, decreases the availability of U(VI), the soluble form of uranium (in the $U^{VI}O_2{}^{2+}$ state). This outcome does not raise additional safety concerns regarding nuclear waste containment. Changes in the oxidation states after irradiation in vacuum will be compared to the changes induced by irradiation in the aqueous environment in the next steps.

Keyword-1

 radiation

Keyword-2

Uranium Fuel

Keyword-3

XPS

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