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(G*) (POS-58) Fabrication and surface chemistry of uranium oxide films for α -irradiation experiments

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Canada plans to use a deep geological repository (DGR) system consisting of corrosion-resistant used fuel containers (UFCs) and other barriers to store spent nuclear fuel safely. Concerns arise regarding potential fuel exposure to groundwater, leading to fuel oxidation and dissolution due to water radiolysis by residual fuel radioactivity. Radionuclides within spent fuel, primarily located in UO_2 grains, can be released based on fuel corrosion rates. Unlike that of β - and γ -radiations, the α -radiation dose rate will remain high for extended periods and make the α -radiolysis of water the primary source of oxidants.

This study aims to explore α -particle interactions with fuel surfaces to figure out their impact on UO_2 dissolution rates. The methodology is to conduct in-situ α -irradiation-electrochemistry experiments using the Rutherford backscattering beamline on the Western Tandatron accelerator and sealed radiation sources to provide a constant flux of high-energy α -particles. As direct usage of UO_2 fuel pellets was impractical due to the specific experimental setup, uranium oxide thin films were grown on metallic foil substrates via electrodeposition, using an aqueous electrolyte containing uranyl nitrate. Films were grown using current densities of 5-30 mA/cm², pH = 7.5 to 8.5, and 76 ± 1 °C temperature.

To make sure the composition of the deposited films matched the composition of the used fuel, a detailed characterization of the films was performed. Films showed a cauliflower-like morphology in SEM analysis, with uranium and oxygen presence confirmed through EDX. RBS measurements indicated the film thicknesses in the 1-5 μm range. XRD showed that as-deposited films were amorphous, turning into UO_2 polycrystalline films after annealing at 600 °C in 10⁻⁶ Torr H₂. Raman analysis detected U_4O_9 and U_3O_8 phases in the as-deposited films, while UO_2 phases emerged in the annealed samples' spectra. Further characterization of the films, as well as preparation for in-situ α -irradiation-electrochemistry experiments, is currently underway.

Keyword-1

Uranium oxides

Keyword-2

α -irradiation experiment

Keyword-3

electrodeposition

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