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Reoxidation/Stabilization of pyrophoric ISOLDE targets after irradiation

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Pyrophoric metal carbides such as uranium carbides (UCx), thorium carbides (ThCx) and lanthanum carbides (LaCx) are used as target materials in CERN-ISOLDE to produce radioisotopes due to their high cross-section, thermal stability and porous structure. After irradiating these materials by proton beams, they become pyrophoric radioactive waste and require controlled oxidation prior to disposal to eliminate the risk of thermal runaway or ignition upon contact with oxidants (mainly oxygen). The aim is to develop a controlled oxidation process for these materials in ISOLDE hot cells by taking the necessary safety precautions and considering environmental impacts, for example capturing the formed radioactive volatile species as a result of radionuclei, produced by proton irradiation, and oxygen reactions. Then, they would be ready for long-term disposal in deep geological repositories.

The project started with non-irradiated micro-structured lanthanum carbide to foresee potential safety risks and practice the process development without radioactivity. The oxidation characteristics of core material (metal carbides) and structural materials (graphite sleeve and tantalum container) of ISOLDE target unit are being investigated using characterization techniques such as thermogravimetric analysis (TGA), differential thermal analysis (DTA), X-ray powder diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and scanning electron microscopy (SEM). Our strategy is to oxidize the core material at the lowest possible temperature under the appropriate oxygen-containing atmosphere to convert it into a thermally stable compound and characterize it by analytical techniques.

Two oxidation pathways were identified for lanthanum carbide in dry and wet atmospheres. In the dry atmosphere (1% O2-Ar mixture), 1074 J/g of heat was generated at 380 oC to reach a thermally stable material (unknown phase). In the moist atmosphere (humid air), the reaction started at room temperature and effectively continued at 50 oC producing 1485 J/g of heat, resulting in a thermally stable lanthanum hydroxide-graphite mixture. Scale-up studies for both methods are ongoing, considering their advantages and disadvantages. Various methods for monitoring the oxidation reaction and determining its completion are also under consideration. The most prominent of these are monitoring of outgases, oxygen concentration, pressure differential and mass change.

Keywords: radioactive waste, pyrophoricity, uranium carbide, thermal runaway, controlled oxidation, dry oxidation, wet oxidation

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