DOE-ID NEPA CX DETERMINATION
Idaho National Laboratory

CX Posting No.: DOE-ID-INL-21-012

SECTION A. Project Title: A Novel Head-End Process for Used ATR Fuels

SECTION B. Project Description and Purpose:

The objective of the proposed project is to expose surrogate materials (aluminides of zirconium, molybdenum and gadolinium) to pure hydrogen, both under ambient conditions and at elevated temperatures to study their hydriding behavior. Hydriding and dehydriding are the means to separate the bulk aluminum from the metallic uranium fuel. The novelty of the proposed process lies in replacing highly reactive (and corrosive) process gas with a clean (and highly selective) chemical agent. The efficiency of the new process will be tested under a variety of experimental conditions. If successful, the developed process will prove to be an elegant reprocessing method with many superior features, such as less number of unit operations, absence of structural material's corrosion, potential applicability for reprocessing of other used alloy fuels and comparatively less expensive.

Research Plan:

Overview: In the conventional aqueous processing, the ATR fuel assembly is dissolved in caustic soda or an acid to remove the aluminum cladding. For some spent fuels, such a process runs the risk of causing explosion, during the dissolution step. Another problem arises because of the corrosion of the aluminum cladding by way of formation of a surface oxide/hydroxide (of aluminum) layer. Presence of these surface layers will impede the cladding dissolution kinetics. This situation will persist even when a dry chlorine gas is used (because chlorine will not effectively react with oxides/hydroxide of aluminum) to volatilize out aluminum in the form of aluminum trichloride (AlCl3) prior to uranium electrorefining. Also, chlorine can react with uranium to form uranium trichloride (UCl3). The intent of the proposed process is to circumvent these process limitations by way of developing a non-aqueous (pyrometallurgical) reprocessing scheme. The research focus will be centered specifically on the decladding step.

Hypothesis: Hydrogen is not only a clean chemical agent but also is highly selective in the way it reacts. Certain elements (some transition metals, platinum group metals, rare-earth elements and actinides) react with hydrogen to form hydrides whereas others (aluminum, copper, nickel, cobalt etc.) do not without the application of very high hydrogen pressures. The highly selective nature of hydrogen will break the bond between aluminum and uranium and result in the formation of uranium hydrides (UHx). Thus, the hydriding step will release chemically bonded uranium while leaving the aluminum matrix and aluminum cladding plates chemically unaltered. Subsequently, the aluminum cladding plates can be physically separated from the uranium hydride powder. In the second stage, the mixture of aluminum and uranium hydride will be treated with a molten metal (lithium, magnesium) to preferentially dissolve aluminum (to form aluminum alloys). These molten alloys do not chemically react with uranium/uranium hydride. Separated uranium hydride is vacuum treated to remove the bulk of the hydrogen prior to electrorefining in order to elevate its ultimate purity.

The experimental work will involve surrogate aluminides (zirconium aluminide, titanium aluminide, molybdenum aluminide and gadolinium aluminide) for developing the proof-of-the concept studies. The entire gamut of the experimental research will consist of the following unit operations:

1. Configuring the experimental set up: A horizontal furnace (available at INL Research Center) will be configured to perform the hydriding tests. The furnace will be equipped with the (i) gas inlet and outlet arrangements (ii) temperature control systems and (iii) vacuum connections to perform the hydriding tests in the absence of air/moisture.

2. Hydriding of the surrogate materials: The surrogate aluminides will be exposed to flowing hydrogen in the horizontal reactor for different durations and at different temperatures (room temperature up to ~9000°C). The reaction products will be stored in a dedicated desiccator prior to their analyses and characterization. Phase and morphological analyses of the products will be carried out by powder x-ray diffraction (XRD) and scanning electron microscope-energy dispersive spectroscopy (SEM-EDS) techniques. Metallic elemental analysis and determination of hydrogen content will be performed by Inductively Coupled Plasma -Atomic Emission Spectroscopy/Optical Emission Spectroscopy (ICP-AES/OES) technique and in an oxygen analyzer respectively. The specific objective will be to optimize the process parameters for quantitatively unlocking the aluminum from the metal hydrides and maximizing the formation of the metal hydrides (MxHy).

3. Treatment with molten metal: These experiments will be performed inside an argon-atmosphere glove box. The hydrided mixture will be placed in a perforated metallic basket and the basket will be lowered into the molten metal pool to preferentially allow the aluminum to form the liquid alloy and concentrate in the molten metal pool.

4. Dehydriding of the metal hydride: The metal hydrides, obtained after the treatment with molten metal, will be vacuum-heated in the furnace (that will be used to perform the hydriding experiment) to remove the bulk of the hydrogen, which will then become a relatively clean anode feed for the subsequent electrorefining tests. Vacuum-treated metal hydride will be analyzed by x-ray diffraction, chemical analysis (ICP-AES/OES) and determination of residual hydrogen (by a LECO analyzer). Order of (dynamic) vacuum, temperature and time will be optimized to maximize hydrogen removal from the metal hydrides.

5. Electrorefining: Electrorefining test runs will be carried out in the laboratories at MFC and Energy Innovation Laboratory (EIL). A eutectic salt mixture, consisting of lithium chloride and potassium chloride (LiCl-KCl), will be used to perform the electrorefining test runs at 500-6000°C. Respective metal chlorides (zirconium tetrachloride, ZrCl4, molybdenum pentachloride, MoCl5 and gadolinium trichloride, GdCl3), as the functional electrolyte, will be dissolved into the eutectic melt to prepare the electrolyte. The electrorefining test runs will be carried out by varying the process parameters (electrolyte composition, current density, operating temperature, duration, etc.). The electro-refined products will be evaluated and characterized by microscopic and chemical analyses. The objective of this final step will be to determine the optimum conditions for maximizing metal purity.

Specific tasks to be performed at the University of Utah

1. Hydriding of surrogate materials (aluminides of zirconium/molybdenum/gadolinium) under hydrogen flow by varying temperature and duration. A constant hydrogen flow can be maintained while performing the hydriding runs, preferably in the presence of a getter to avoid oxygen contamination.

2. Optimization of the hydriding parameters (temperature, duration).

3. Nature of the aluminide (plates are preferred but rods and/or wires can be used).

4. Depending on the surface conditions, a surface prep. (etching, sand blasting, polishing, etc.) may be incorporated prior to the exposure of materials/alloys to hydrogen.

5. Evaluation and characterization (by X-Ray Diffraction and ICP-AES/OES) of materials (before and after the treatment with hydrogen).

6. Generation of materials/research data for the publication of one/two journal papers.

7. Depending on the progress, additional work will be performed, as necessary.

8. Preparation of project reports (one/year).

The materials will include metallic alloy specimens (10-20g each) and hydrogen gas. The total quantity of the alloys will be between 100-200g. These are not radioactive samples. The alloys include titanium aluminide, zirconium aluminide and gadolinium aluminide.

SECTION C. Environmental Aspects or Potential Sources of Impact:

Air Emissions

Emissions will be minor and covered by existing APAD's for the individual facilities.

Discharging to Surface-, Storm-, or Ground Water

N/A

Disturbing Cultural or Biological Resources

N/A

Generating and Managing Waste

The project will generate waste, including office waste, industrial waste (e.g., gloves, non-hazardous hardware, ceramic-type pellets, machining scrap, lab pipettes, wipes, etc.) Some acidic liquids may be generated from analysis of samples.

Releasing Contaminants

When chemicals are used, there is the potential the chemicals could be spilled to air, water, or soil.

Using, Reusing, and Conserving Natural Resources

All materials will be reused and recycled where economically practicable. All applicable waste will be diverted from disposal in the landfill where conditions allow. Project description indicates materials will need to be purchased or used that require sourcing materials from the environment. Being conscientious about the types of materials used could reduce the impact to our natural resources Project activities may release known greenhouse gases (GHGs) to the atmosphere and increase INL's energy use.

SECTION D. Determine Recommended Level of Environmental Review, Identify Reference(s), and State Justification: Identify the applicable categorical exclusion from 10 Code of Federal Regulation (CFR) 1021, Appendix B, give the appropriate justification, and the approval date.

For Categorical Exclusions (CXs), the proposed action must not: (1) threaten a violation of applicable statutory, regulatory, or permit requirements for environmental, safety, and health, or similar requirements of Department of Energy (DOE) or Executive Orders; (2) require siting and construction or major expansion of waste storage, disposal, recovery, or treatment or facilities; (3) disturb hazardous substances, pollutants, contaminants, or Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)-excluded petroleum and natural gas products that pre-exist in the environment such that there would be uncontrolled or unpermitted releases; (4) have the potential to cause significant impacts on environmentally sensitive resources (see 10 CFR 1021). In addition, no extraordinary circumstances related to the proposal exist that would affect the significance of the action. In addition, the action is not “connected” to other action actions (40 CFR 1508.25(a)(1) and is not related to other actions with individually insignificant but cumulatively significant impacts (40 CFR 1608.27(b)(7)).

References: 10 CFR 1021 Appendix B to subpart D, Item B3.6, "Small-scale research and development, laboratory operations, and pilot projects" and B1.24 "Property Transfers"

Justification: The proposed R&D activities are consistent with CX B3.6 "Siting, construction, modification, operation, and decommissioning of facilities for small-scale research and development projects; conventional laboratory operations (such as preparation of chemical standards and sample analysis); small-scale pilot projects (generally less than 2 years) frequently conducted to verify a concept before demonstration actions, provided that construction or modification would be within or contiguous to a previously disturbed area (where active utilities and currently used roads are readily accessible). Not included in this category are demonstration actions, meaning actions that are undertaken at a scale to show whether a technology would be viable on a larger scale and suitable for commercial deployment.",

B1.24, "Transfer, lease, disposition, or acquisition of interests in personal property (including, but not limited to, equipment and materials) or real property (including, but not limited to, permanent structures and land), provided that under reasonably foreseeable uses (1) there would be no potential for release of substances at a level, or in a form, that could pose a threat to public health or the environment and (2) the covered actions would not have the potential to cause a significant change in impacts from before the transfer, lease, disposition, or acquisition of interests."

Is the project funded by the American Recovery and Reinvestment Act of 2009 (Recovery Act) ☐ Yes ☒ No

Approved by Jason Sturm, DOE-ID NEPA Compliance Officer on: 1/27/2021