Contract No:

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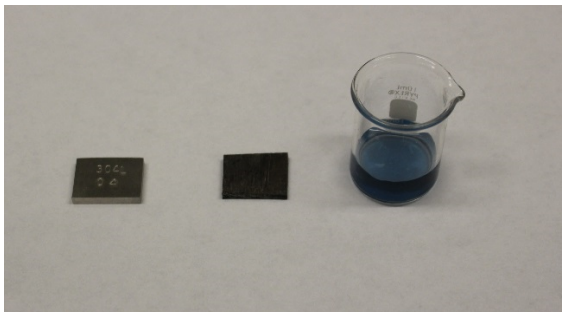
Demonstration of Chemical Decladding and Disposition Options for Non-Al Clad Fuels in H-Canyon

Project highlight. Two dissolution methods were identified and tested on the lab scale that would allow decladding of non-Al clad fuels in H-Canyon. This is an additional disposition option for the non-AL clad fuels in L-Basin beside shipment offsite to be processed or dry storage onsite. For dissolution on a large scale, H-Canyon, work still needs to be done to address waste streams but on a small scale some L-Basin fuels could now be dissolved in hot cells and sent to the waste tanks.



Dissolved stainless steel cladding

Awards and Recognition

NA

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publically published in its current form.

SRNL Legal Signature

[redacted] 10/1/18

Signature Date

Demonstration of Chemical Decladding and Disposition options for Non-Al clad Fuels in H-Canyon

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Subcontractor: SRNL

Thrust Area: NMM

Project Start Date: October 1, 2017
Project End Date: September 30, 2018

Aqueous disposition of non-Al clad fuels was studied in order to determine if H-Canyon would be a viable disposition option for this type of L-Basin fuel. A review of the L-Basin fuel types and its condition was done suggesting two major cladding types, Zircaloy and 304L stainless steel needed to be studied. Dissolution flowsheets options were reviewed. Sulfex and Zirflex processes were chosen as best for H-Canyon integration. Both process flowsheets were demonstrated on a lab scale to determine decladding rate, and off-gas and waste production. The resulting data suggested decladding in H-Canyon was an option but dissolver corrosion as well as off-gas and waste generation needed to be addressed. Small scale dissolution in a hot cell of smaller damaged fuel is a viable option with the flowsheets tested.

FY2018 Objectives

- Review of L-Basin non-Al clad fuels
- Review of aqueous decladding flowsheets
- Scoping experiments to evaluate flowsheets identified as promising for SRS use, include dissolution rate, off-gas, and waste produced
- Training of new scientists on process chemistry

Introduction

L-Basin inventory includes stainless steel and Zircaloy clad fuels that need to be dispositioned. A disposition path of these fuels is uncertain or "TBD" with some being corroded, damaged, or at risk of degrading with time. A possible path, utilizing H-Canyon would need development as H-Canyon flowsheets don't address non-Al clad fuels. Technologies exist to declad stainless steel and Zircoly-clad fuels such as Darex, Sulfex, and Zirflex. These technologies are decades old and will likely result in effective decladding allowing fissile material recovery or disposal. However, the question of whether they can utilized at SRS in H-Canyon processing equipment or hot cells, with the resulting waste streams dispositioned needs study.

The project was significant for SRS in that it identified decladding technologies that with future work would be most useful for disposition of non -AL clad L-Basin fuels. The review of the L-Basin fuel cladding and its condition suggested the fuels most in need of disposition. The scoping dissolution rate tests run for this project pointed out that on a small scale both Sulfex and Zirflex were viable technologies for SRS, but on a larger H-Canyon scale Sulfex was the one that would be easiest to implement but waste streams and equipment corrosion need to be addressed. Dissolution of small "boutique" fuels in the SRNL hot cells now has options.

Approach

The objectives of the project were achieved through running lab scale dissolution experiments on non-Al cladding at typical H-Canyon temperatures for the dissolution technologies that were identified. Initial beaker tests were run prior to testing in the reaction vessel. Waste output and off-gas produced during dissolution were also monitored. The results would be used determine applicability for running at larger scale in H-Canyon. Lab scale results could also be used for smaller scale dissolution in hot cells. If our tests fit the older test data we assumed all of the older data could be used for assessing dissolutions in H-Canyon.

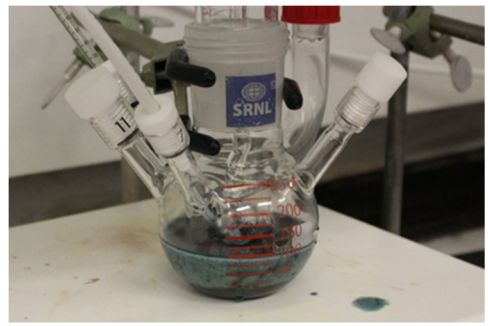

Figure 1. Reaction vessel showing 304L metal cladding dissolution rate experiment.

Results/Discussion

The non-Al clad L-Basin fuel inventory was reviewed and binned for undamaged verses damaged cladding and fuel type. Three cladding types, Zircaloy 2, Zircaloy 4, and 304L stainless steel were found to be the majority in the Basin. Our dissolution technologies were thus constrained to stainless steel and Zircaloy cladding. A literature search for flowsheets was completed with three (Sulfex, Zirflex and Darex) flowsheets selected for study. The initial check of downstream effects for H Canyon disqualified Darex as it contained 3M HCl which would be problematic to fully remove. Chloride is well known cause crevice corrosion and produce pinholes in stainless steel. Only Sulfex and Zirflex would be tested.

Two Sulfex dissolution solution concentrations 4M and 6M H_2SO_4 were tested on stainless steel. Dissolution rates of matched earlier work. The dissolution gives off only H_2 in a 1:1, stainless steel : H_2 molar ratio. Thus air sparging will need to be run with this process in H-Canyon as well as using a dissolver insert or corrosion resistant dissolver. Considerable sulfate solids are produced so waste issues may be a problem with large scale processing.

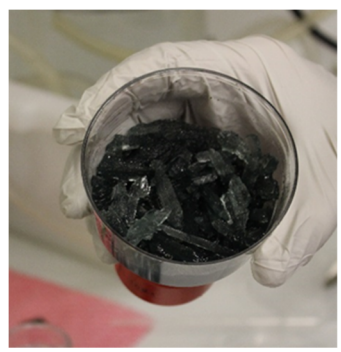

Figure 2. Na_2SO_4 crystals produced from Sulfex waste solutions.

Two Zirflex dissolution solution concentrations 4M NH_4F + 0.5M NH_4NO_3 and 6M NH_4F + 1M NH_4NO_3 were tested on Zircaloy cladding. Dissolution rates were matched those of earlier work. The dissolution gave off a substantial amount of gas. Both H_2 and NH_3 are given off with the majority being NH_3. Dealing with the large amount of NH_3 gas with the nitric acid used in H – Canyon is an issue that would need to be addressed. The solubility of the waste product $(NH_4)_2ZrF_6$ would also have to be dealt with.

FY2018 Accomplishments

- Discovered the majority of non-Al clad L-Basin Fuels is clad in Zircaloy 2 and 304 L stainless steel so fewer dissolution flowsheets are needed.
- Sulfex and Zirflex dissolutions are consistent with earlier data allowing for this older data to be used for dissolution system design – less experimental development is needed

- Sulfex and Zirflex dissolutions produce substantial waste so large-scale processing in H-Canyon would need to address this in the SRS waste disposition processes.
- Sulfex and Zirflex dissolutions worked well which adds an aqueous decladding tool to small-scale dissolutions in Hot cells viable
- Two newer scientists were trained in experimentation needed for process chemistry

Future Directions

- Apply for DOE funding for dissolution of small damaged fuels in hot cells
- Corrosion testing to determine ideal dissolver material

FY 2018 Publications/Presentations

1. "Demonstration of Sulfex and Zirflex Dissolution Processes" D.D. Dick, A.B. Thompson, W.E. Daniel, M.G. Bronikowski. and T.S. Rudisill, SRNL-STI-2018-00509, Sept. 2018.
2. "Application of Chemical Decladding to Non - aluminum Clad fuels in L-Area Basin", T.S.Rudisill
3. "Demonstration of chemical decladding and disposition options for non-Al clad nuclear fuels at the Savannah River Site" A. Thompson, D. Dick, G. Daniel, T. Rudisill, M. DeVore, and M. Bronikowski, Poster presentation, 70th SERMACS2018, Nov.1, 2018

References

Acronyms

Intellectual Property

NA

Total Number of Post-Doctoral Researchers

DeWayne Dick