

# OAK RIDGE NATIONAL LABORATORY

MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

Post Office Box 2008  
Oak Ridge, TN 37831-6243  
Phone: (865) 574-4934  
Fax: (865) 574-6872  
E-mail: jubinrt@ornl.gov

November 24, 2010

To: Terry Todd

From: Robert T. Jubin

**Subject: Completion of the ORNL Fuel Cycle Research and Development (FCR&D) Level 3 Milestone – Off-Gas Sigma Team - FTOR11SW0309, MS# M31SW030904, “Sigma Team Coordination Meeting,” due 11/30/2010**

This letter documents the completion of the FCR&D Level 3 milestone for the Off-Gas Sigma Team - ORNL work package (FTOR11SW0309), “Sigma Team Coordination Meeting” (M31SW030904), due 30 November 10. The subject meeting was held at Idaho National Laboratory on October 12 and 13, 2010. The agenda and meeting minutes are attached to this memo.

If you have any questions, please contact me at (865) 574-4934.

cc: C. V. Bates (INL)  
E. D. Collins  
L. K. Felker  
D. T. Ingersoll  
D. K. Jensen (INL)  
D. W. Ramey  
B. B. Spencer

Minutes of the Off-Gas Sigma Team Meeting  
Oct 12-13, 2010  
Idaho National Laboratory

Attached to these minutes is the agenda of the meeting

Attendees:

Jim Bresee (DOE – Via Phone)  
Kristi Christensen (INL)  
Jeff Fortner (ANL)  
Steve Frank (INL)  
Troy Garn (INL)  
Mitch Greenhalgh (INL)  
Bob Jubin (ORNL)  
Jack Law (INL)  
Tina Nenoff (SNL – Via Phone)  
Veronica Rutledge (INL)  
Nick Soelberg (INL)  
Denis Strachan (PNNL)  
Terry Todd (INL)

Introduction of new team members:

The meeting began with introductions. There were a number of new (and some not so new) faces in the room from INL. These folks will be working on the following tasks:

Nick Soelberg – PI on iodine work  
Mitch Greenhalgh – PI on Krypton work  
Troy Garn – Kr capture  
Veronica Rutledge – Iodine and modeling  
Kristi Christenson – Kr capture

Quick review of FY 11 technical scope:

*ORNL - Iodine*

Continue “Single Pellet” or thin bed organic iodide or elemental iodine loading of dry air aged hydrogen-reduced silver exchanged mordenite to determine impact of aging and aging conditions on sorbent performance.  
Perform microscopy and other analysis to determine changes to structure resulting from aging and aging environments.  
Procure synthetic highly crystalline mordenite in either engineered (pellet) form or powder form and provide to others as appropriate.  
Prepare samples of silver exchanged mordenites loaded at difference rates and to different extents of loading to initiate the determination of the iodine loading sites within the media and how this changes with increased loading.  
Initiate long term (~6 month) aging studies with NO<sub>x</sub> as a constituent in the gas stream.  
Conduct modeling of data to derive rate parameters and to support other associated modeling efforts.

Provide aged and fresh baseline and other materials to SNL and INL.  
Conduct iodine loading tests on alternate sorbents from PNNL and SNL. This provides common loading and performance measurements.  
Conduct caustic absorption and silver nitrate absorption / precipitation tests and provide bulk iodine samples to SNL to support waste form development activities.

*INL - Iodine*

Deep bed loading tests to determine mass transfer zone length and overall DF as a function of flow rate and temperature.  
Observed in FY10 that the bed effluent contained elemental iodine during loading tests using methyl iodide – further understanding of mechanism is needed.  
One possible test idea offered was to repeat run using methyl iodide and a bed of mordenite containing no silver – helps to determine if silver plays any role.  
Catalytic reaction with mordenite?  
Testing of alternate media (NC-77, light phase of silver exchanged mordenite (AgZ) [synthetic mordenite], others.

*PNNL – Iodine*

Continue to develop silver functionalized silica aerogels  
Determine loading capacity  
Provide to ORNL for comparative tests  
Chalogels – iodine loading capacity and retention of these non-silver containing materials

*SNL - Iodine*

Mechanistic studies on reduction of Ag and loading of iodine – planning additional tests with ORNL aged materials  
Loading of iodine on MOF  
Low temperature glass iodine waste forms

*ANL – Iodine*

Designing experiment to help determine what happens to the iodine during electrolysis for drawdown – large data gap here.  
Working with INL on planning to make iodine measure on actual materials

*INL – Krypton work*

Key questions:  
How is selectivity impacted by nitrogen / air?  
What materials should be tested? AgZ, MOF, other  
Need vendor for hydrogen mordenite – ORNL to provide info from CETE supplier.  
Work with a newly developed nanostructured titania material  
Selectivity / function of temperature  
Comparison of several materials for Kr recovery / selectivity and thermal stability  
Regeneration – thermal swing  
Current bed is 2 inch diameter and 7 inch tall  
Could go as small as 1/8 inch diameter

*PNNL Krypton work*

Room temperature selection of Kr and Xe from air  
Disposal of Kr in MOF filled containers  
Isolation of Rb  
Work with ZIF8 MOF

Provide MOF to INL for testing  
Radiation stability of materials

*ORNL - Tritium*

Conduct “Single Pellet” or thin bed tritium adsorption / desorption tests on Type 3A molecular sieves.

Derivation of tritiated water adsorption / desorption rate data from thin bed studies

Modeling of deep bed performance from data obtained in thin bed studies and from the literature

Determine “Single Pellet” or thin bed iodine adsorption / desorption rates and capacity for Type 3A molecular sieves.

Evaluation of tritium co-adsorption on silver-exchanged mordenite between 25 and 200°C.

Peer Review:

Purpose of the peer review is to help steer scope of work and provide an outside perspective on what we are doing. Not to specifically direct funding of work but to improve our progress in solving the technical and scientific challenges. Proposed themes to be presented:

Iodine Sorption:

Key Technical Challenges:

Mechanism of iodine capture

Rate limiting steps

Iodine Waste Form

Key Technical Challenges:

How to control release rate to acceptable limits.

How does waste form interact with water

Effects of radiation and radiolysis

Krypton

Key Technical Challenges:

Krypton selectivity and mechanism of selectivity on specific sorbents

Rb in-growth issues

Capacity and heat generation – what are limits

Tritium

Key Technical Challenges:

Better understanding of source terms

Impact of head-end processing

Development of isotherm data at very low concentrations

Recovery / immobilization from aqueous systems

Role of modeling

Fundamental vs. applied

Data needs and timing

Peer review should be identified early as possible. Sigma Team members should make suggestions.

Peer review panel should be asked to provide questions that they want Team to answer during review process.

Discussion on analysis of HIPed Kr Samples located at INL

A series of slides were presented to the team that showed the samples in detail and laid out preliminary information that had been located on the samples.

These samples were identified and preserved prior to D&D of building CPP-603 at INL. Discussions with researchers who performed the work and review of literature resulting from the project indicate that the metal capsules contained <sup>85</sup>Kr encapsulated in a 5A zeolite in a glass matrix that was HIPed at about 5000 psi. Samples have now experienced ~3.5 half lives of the <sup>85</sup>Kr. It was suggested looking at the photos that one capsule may not have been HIPed.

Preliminary thoughts on analysis needing include:

To obtain information on:	Possible methods could be utilized
Radiation damage to zeolite matrix	XRF
Structure of capsule and waste form	Neutron radiography
Chemical state of Rb	
Container state / Waste form state	XRD, SEM, TEM

Test plan for the analysis of the samples to be issued by INL on or before 24 Dec, 2010. ORNL and other labs to provide input.

Combined waste mass/volume and C-14 / Kr-85 management option study report.

The FY 11 Off-Gas Sigma Team efforts also included two closely related studies that targeted two separate audiences. The first requested report is to provide estimates of the off-gas related waste / waste form volumes. The purpose of this study is to support systems engineering studies being performed by several groups (e.g., Systems Analysis and Used Fuel Disposition). These estimates would include: estimates of the source terms associated with several processing options and splits within the off-gas treatment and waste processing steps. Based on these splits and the waste forms selected the mass and volume of the waste forms would be estimated. Ideally these could be ultimately reduced to a set of equations that would be used in options studies. Special attention should be put on the order of separation (e.g., I before or after <sup>3</sup>H). The second report is to examine the requirements for capture of <sup>14</sup>C and <sup>85</sup>Kr during reprocessing operations and the options for Kr and CO<sub>2</sub> removal and/or release. The technical issues should be the focus of this analysis, rather than the regulatory aspects. This second study would support ongoing efforts by EPA and other regulatory bodies.

There appeared to be significant overlap between these two studies and a single dual purpose analysis and report was proposed. The preliminary report outline was drafted and is attached to these minutes along with the proposed schedule to prepare the report.

Iodine distribution in Pyroprocess

Steve Frank provided a review of the pyroprocess and some limited data on the distribution and pathways throughout the process. He is looking for help/input on how to make measurements to confirm / determine iodine releases at key points in the process. LiveWeb meeting scheduled for

Nov 12 to continue this discussion. Some of the questions to be answered include: How much iodine is lost during the ceramic waste form processing, salt distillation, and from the electrorefiner. To date they have not been able to directly measure iodine in the ceramic waste and need leaching data on the ceramic waste form.

Schedule for upcoming meetings:

Joint discussions on the iodine mass balances in the e-chem processes. Sigma Team / Steve Frank and others. Format: LiveMeeting web conference – Nov 12, 2010.

Review / coordination of Sigma Team report(s) on Waste volumes/masses and Kr/C management. Format: Face to Face meeting – SNL – April 19-20, 2011.

Review of Technical Progress. Format: Face to Face meeting – Combine with two day report writing/review meeting. SNL - April 21, 2011.

Dry Run of Peer Review Presentations. Format: LiveMeeting web conference – two to three weeks before Peer Review (June 1, 2011?).

Work Package Preplanning meeting. Format: Face to Face meeting – same week as peer review meeting (mid-June).

Joint NEAMS off-gas modeling and Sigma Team meeting. Format: Face to Face meeting – same week as peer review meeting (mid-June).

## Report Outline

### Introduction

- Purpose of the document
- Need for this document

### Scope Statement

- Some version of what is above with additional input
- What is in scope and what is not

### Assumptions and Caveats

- Source Terms
  - Fuels types
  - Fuel burn-up
  - Decay time
- Discussion of waste classification
- Current requirements / drivers
- DFs needed vs DFs needed to perform the analyses in this document

### Part I – Generation and Capture Technologies

- Estimation of stream compositions
  - Generation rates
  - Concentrations
    - Radionuclides
    - Other key constituents
  - Splits into various streams
- Applicable capture technologies
  - Capture efficiency
  - Products
    - Form
    - Composition
    - Volumes
- Variations

### Part II – Waste

- Waste forms
- Conversion processes
  - Complexity
  - Requirements
- Waste estimations
  - Waste loadings
  - Waste form volumes / mass



Part III

Management options

Conclusions

**Proposed schedule for the combined waste mass/volume and C-14 / Kr-85 management option study**

Annotated Outline	12/15/2010
Draft of intro and scope sections	1/31/2011
Source Terms / Basis of estimates / Stream definitions	2/28/2011
Draft of capture / management requirements section	2/28/2011
Draft of capture technologies sections complete	3/30/2011
Industrial Practice for C / Kr capture	3/30/2011
Estimations of Splits	4/30/2011
Draft of text for Waste forms and processes	5/15/2011
Waste Form Volumes	5/30/2011
Finalize Draft	7/31/2011
To Tech Editor	8/1/2011
Final review	8/10/2011
Final comments due	8/15/2011
Document Clearance	8/22/2011
Issue document	8/31/2011