AFCI Separations Activities

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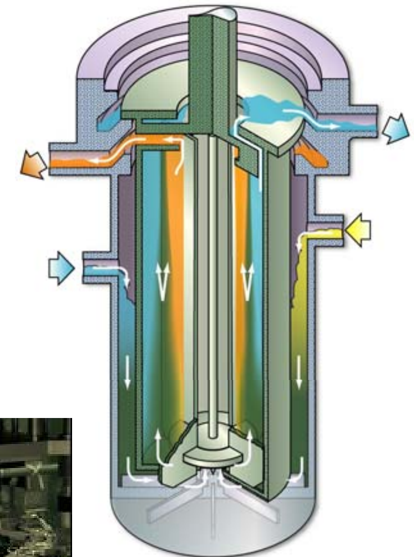
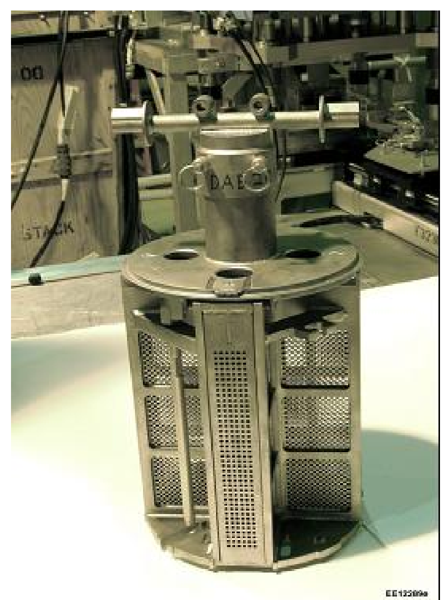
Overview

- Separations Campaign Overview
- Key Campaign Accomplishment / Goals
- Coupled End to End (CETE) Project
- Off-gas Treatment
  - ORNL
  - INL
  - SNL
- Centrifugal Contactors
  - INL

Separations Campaign Program Overview

- **Major program elements**
  - Advanced aqueous separations
  - Advanced electrochemical separations
  - Process equipment scale-up and development
  - Off-gas treatment
  - Process control and monitoring
  - Process modeling and simulation
  - EBR-II spent fuel treatment
  - Separations regulatory and safety crosscut





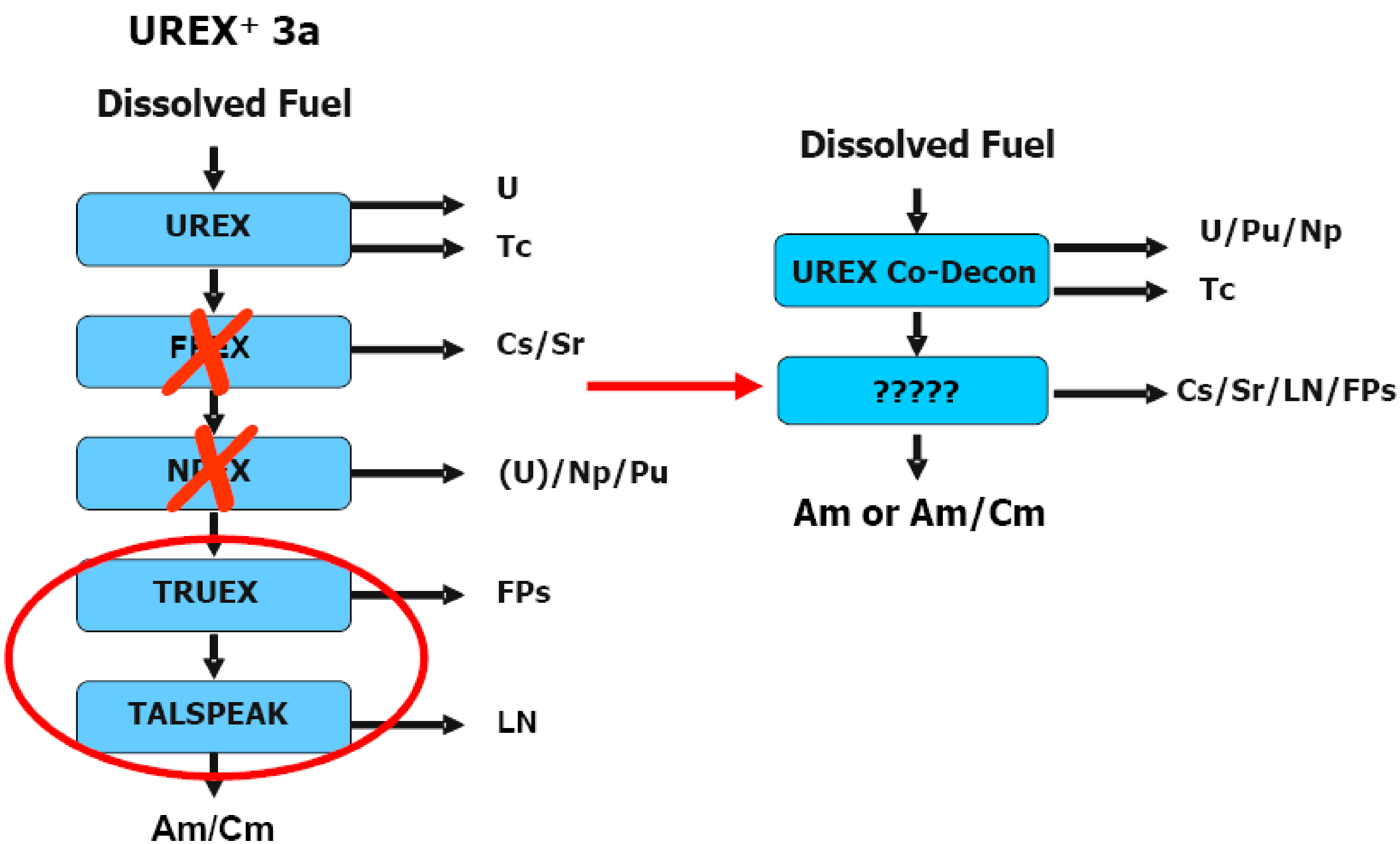
Separations Campaign FY-08 Major Accomplishments

- Revised campaign research strategy for aqueous processing
  - De-emphasize repetitive testing of separation process at lab-scale using small amounts of used nuclear fuel
  - Focus on head-end operations
    - Voloxidation to separate tritium
    - Off-gas capture and immobilization of ^{129}I, ^{85}Kr (and Xe), ^{3}H, ^{14}C
  - Demonstrate product conversion using modified direct denitration
  - Transition development activities from laboratory testing to engineering development
    - Incorporate industry input into campaign development activities
    - Increase Technology Readiness Levels (TRL's) > 6
    - Incorporate solvent recycle effects into testing
  - Integrate Waste Form development with Separations development

Separations Campaign FY-08 Major Accomplishments

- **Revised campaign research strategy for aqueous processing**
  - Focus on ***simplification*** of separation processes
    - **Decay store HLW rather than separate Cs/Sr**
    - **Reduce number of separation steps and separated products**
      - **Separate U/Pu/Np together by manipulating valance states**
    - **Combine TRUEX/TALSPEAK or use alternative technology**
      - **Avoid dependence on rigid pH control**
  - Develop science-based approach to develop alternative Am/Cm separation process
    - **Formation of "*SIGMA TEAM*"**
    - **Multi-lab team focused on development of robust separation process for separation of Am or Am/Cm in a single step**

Separations strategy evolution



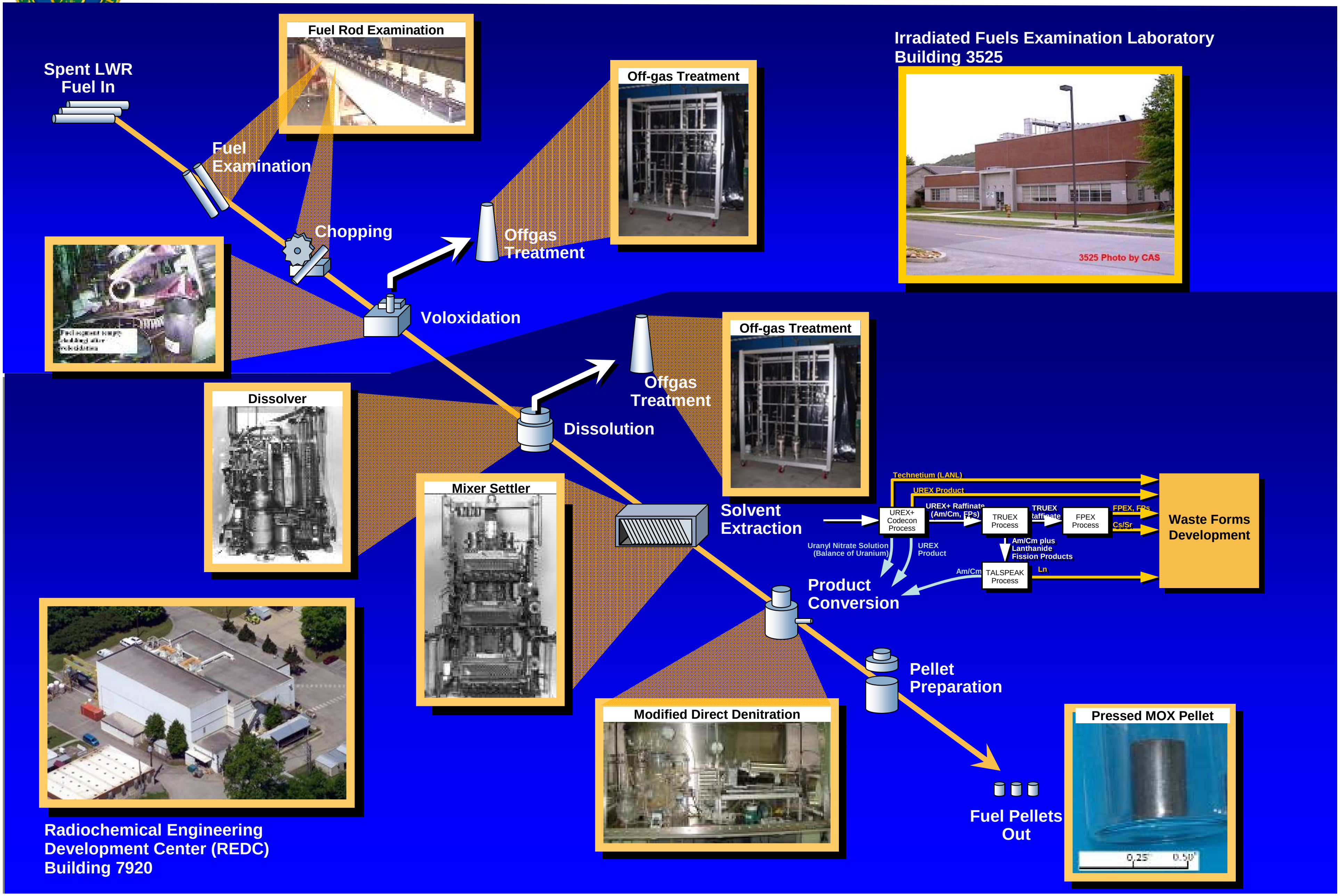
Selected Development Focus Areas for Aqueous Technologies

- **Focus on simplification of process is key objective to improve operation and reliability and reduce cost**
  - Studies have shown that the UREX+ separations options are complicated and costly
  - Largest challenge is separation of Am/Cm or Am alone
    - *Focus area for Sigma Team*
- **CETE Run 2 (focus on off-gas capture and MDD)**
- **Capture and immobilization of volatile off-gas (^{129}I, ^{85}Kr, ^{3}H, ^{14}C?)**
  - Voloxidation to separate tritium
  - Off-gas capture and immobilization
- **Testing at scales larger than laboratory scale, in prototypic equipment w/ solvent recycle (increase TRL's)**
- **Integration of separations and waste form development activities (each affects the other)**
- **Process monitoring and control improvements**
- **Collaboration with industrial teams to identify and resolve design data needs**

Introduction to the CETE Demonstration

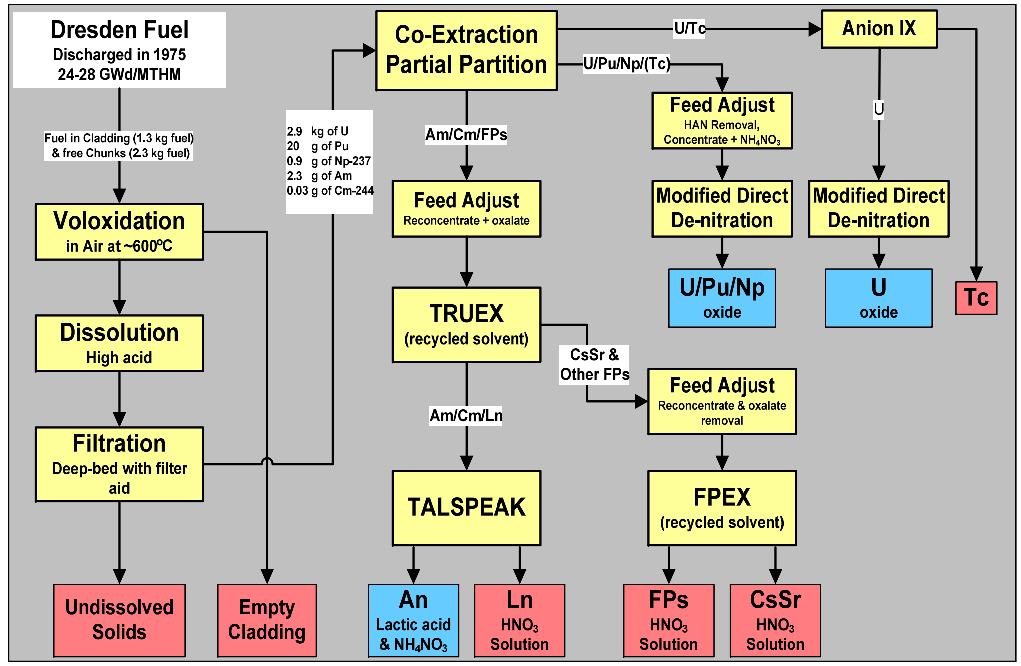
- **Develop/Demonstrate Advanced Recycling Technologies**
- **Multiple process runs ~ 5 - 10 kgs/yr of SNF**
- **Identify and Resolve Scientific & Technical Uncertainties**
  - Interfacial Issues
  - Process Robustness
- **Provide Research Products for Evaluation Across DOE Complex**
  - Actinides
  - FP Waste form
- **FY09: Focus on Head End Processing and Product Conversions**

Coupled-End-to-End Demonstration Overview



Highlights and Lessons Learned from CETE Run 1

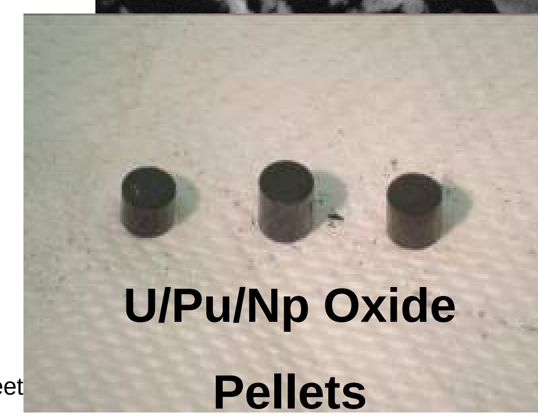
- **Flowsheet Demonstration**
  - Partial partitioning of a U-Pu-Np Product: *No separated Pu*
  - TRUEX-TALSPEAK for Minor Actinide Separation
  - FPEX for Cs/Sr Separation
- **Converted U-Pu-Np and U by Modified Direct Denitration**
- **Demonstrated Fabrication U-Pu-Np co-converted product to pellets.**



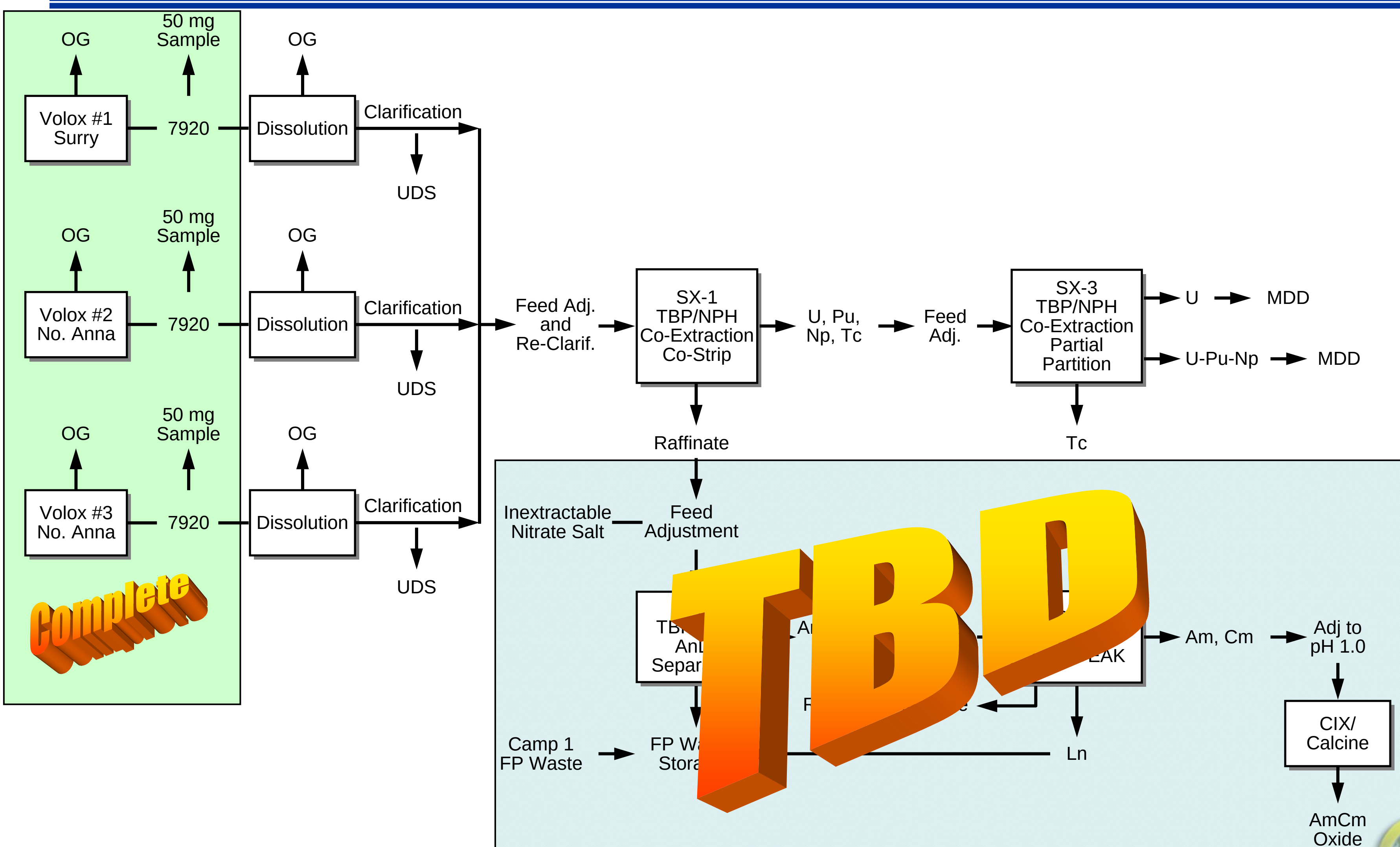
- **Removal and disposal or recycle of the residual chemical complexants (DTPA/Lactic Acid) used in the TRUEX-TALSPEAK processes from the americium-curium product is difficult.**
  - Recovery by re-extraction into HDEHP has been successful

Product Conversions

- **Modified Direct Denitration shows promise for simplifying the interface between separation and fuel fabrication.**
- **Produces a powder with good ceramic properties for pellet fabrication.**
- **Further R&D required**
  - **Process development**
  - **Scaleup**
  - **Qualifying the ceramic product**



CETE Run 2 Plans



Management of Volatile Fission Products

- **Volatile components that must be considered have a wide range of half-lives and disposal requirements:**
  - ^{3}H 12.31 yr
  - ^{14}C 5715 yr
  - Xe/^{85}Kr Stable/10.76 yr
  - ^{129}I 1.57×10^{7} yr
- **Regulatory Drivers, some not written with consideration for full-scale reprocessing facilities, unlikely to be relaxed**
- **Data still needed on process integration and removal process interactions**
  - Description of integrated capture systems for CETE Demo
  - Complemented by cold Bench-scale testing at INL
  - Waste form work for Iodine at SNL

Federal regulations limit radionuclide air emissions in the U.S.

	Dose equivalent to public, mrem/yr	Max fuel cycle emissions per GWyr energy produced	Ambient air conc at site boundary, Ci/m^3
DOE facilities (40CFR61.92)	10		
Nuclear fuel cycle (40CFR190.10)	25 (75 to thyroid, 25 to any other organ)	^{85}Kr: <50,000 Ci ^{129}I: <5 mCi ^{239}Pu: <0.5 mCi	
NRC licensees (10CFR20.1301, .1302, App. B)	100		^{3}H: 1.0E-7 ^{14}C as CO_2: 3.0E-7 ^{85}Kr: 7.0E-7 ^{129}I: 4.0E-11
Others (40CFR61.102)	10 (3 from ^{129}I alone)		

40 CFR 190 Derived Process Requirements

Isotope	Ci/MTIHM	Ci/GW(e)-yr	Min Required DF
^{129}I	0.02648	0.89	178
^{85}Kr (5 year cooled)	7121	239,000	4.77
^{85}Kr (10 year cooled)	5154	173,000	3.45
^{85}Kr (30 year cooled)	1414	47,000	0.95

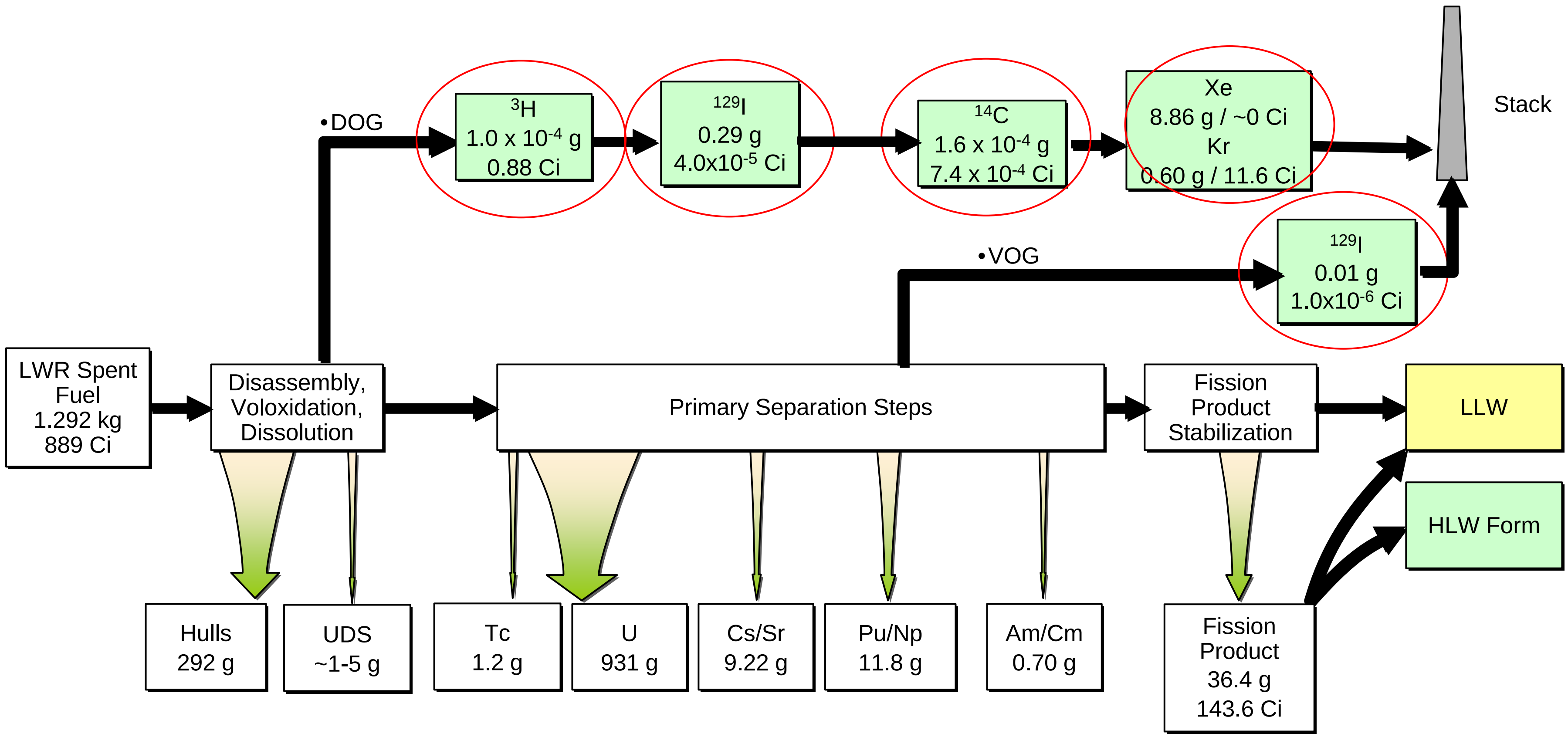
Note: Burn-up: 33 GWd/MTIHM

Isotope	Ci/MTIHM	Ci/GW(e)-yr	Min Required DF
^{129}I	0.04149	0.83	167
^{85}Kr (5 year cooled)	11620	234,000	4.67
^{85}Kr (10 year cooled)	8413	169,000	2.38
^{85}Kr (30 year cooled)	2309	46,000	0.93

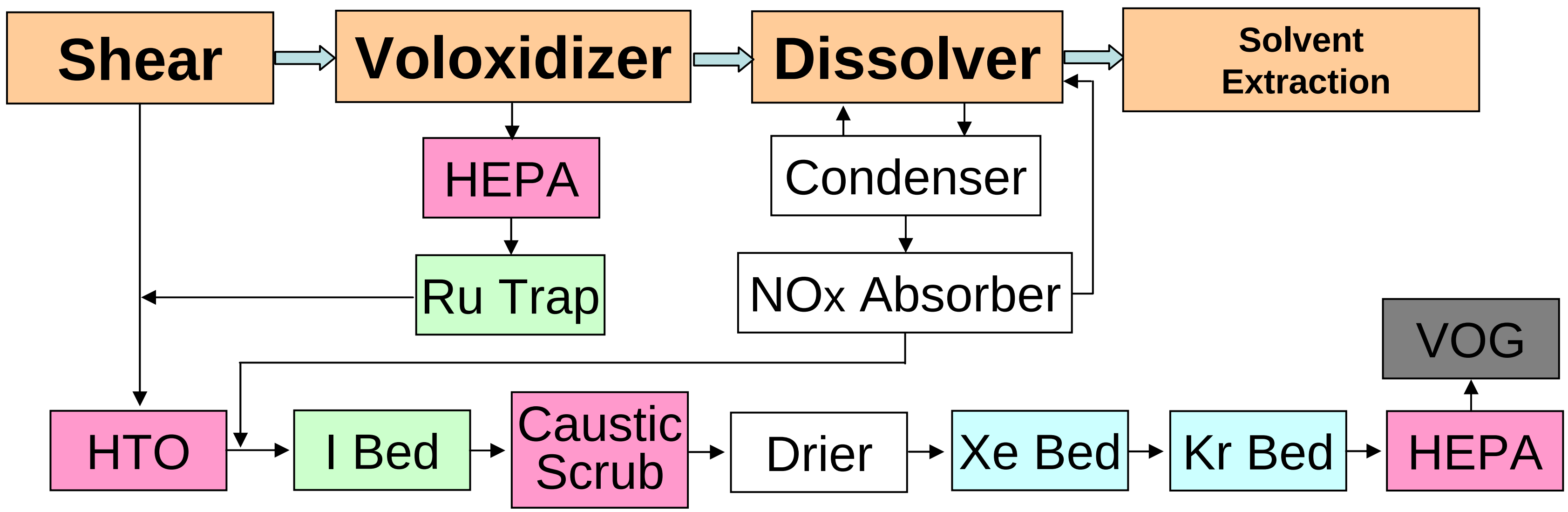
Note: Burn-up: 55 GWd/MTIHM

Simple Reprocessing Demonstration

(Mass Basis: 1 kg SNF; 55 GWD/MTIHM; 5 year Cooling)



Generic Head-End Off-Gas Treatment Concept



HTO	I Bed	Caustic Scrub	Xe Bed	Kr Bed	HEPA
Catalytic oxidizer	*Ag-Z capture*	*NaOH scrub*	*Cryogenic Ag-Z*	*Cryogenic HZ*	*Remove*
MS (3A) capture	*Remove bed*	*Remove*	*Release from Ag-Z*	*Release from HZ*	*Leach*
Release from MS	*Encapsulate*	*Mix in cement*	*Combine w/ Kr*	*Combine w/ Xe*	*Dispose - SLB*
Condense	*Package*	*In-can grouting*	*or Release*	*Pressurize*	
Mix in cement	*Dispose - Rep*	*Dispose - SLB*		*Bottle*	
In-can grouting				*Decay storage*	
Dispose - SLB					

FY 09 CETE Based Tests (ORNL)

- **Hot Integrated Off-gas capture tests on the Voloxidation and Dissolver Off-Gas streams**
  - Objectives
    - *Close volatile component material balances*
      - Voloxidation
      - Dissolution
        - *Analysis for residual iodine in dissolver solution*
      - Shearing of full length fuel – Run 3 or later
    - *Understand impacts of head-end processing conditions on volatile component releases*
    - *Determine capture process interactions*

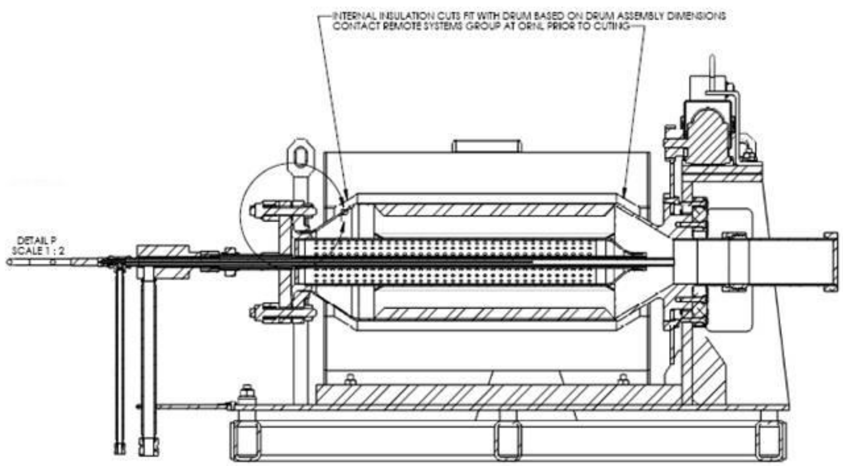
Voloxidation Run 2—Plan and Status

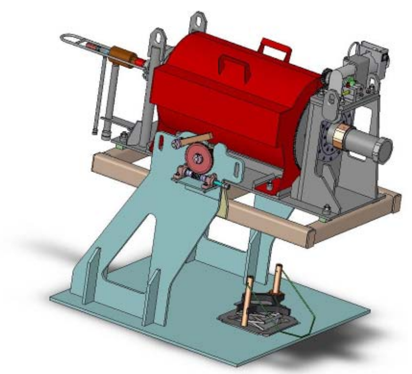
- **Voloxidation processing for Run 2 was planned as three batches**
  - Batch sizes of 1 to 2 kg of spent fuel
  - Total production of at least 5 kg to support separations activities
  - Different conditions were planned for each batch to obtain information on reaction properties and subsequent dissolution properties of product powder (e.g. undissolved solids)
- **Flexibility was built into the test plan to permit selection of alternative fuel materials, depending upon availability**

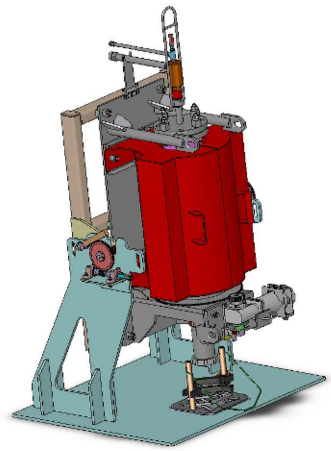
Batch	Fuel Source	Fuel Amt (kg)	Oxidant	Temperature (°C)
1	Surry-2	1–2	Air	500.
2	North Anna	2	Air	600.
3	North Anna	2	oxygen	550.

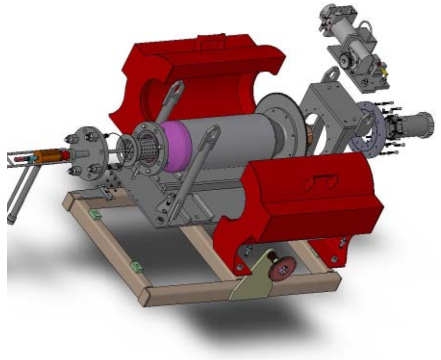
Small Scale Voloxidation

- **Most of the retort tube is enveloped by the furnace**
- **Rotating tube**
- **Variable operating environment**
- **Removable Hulls basket**
- **Powder can integrated into design**
- **Tilting platform can force material into hotter zones**

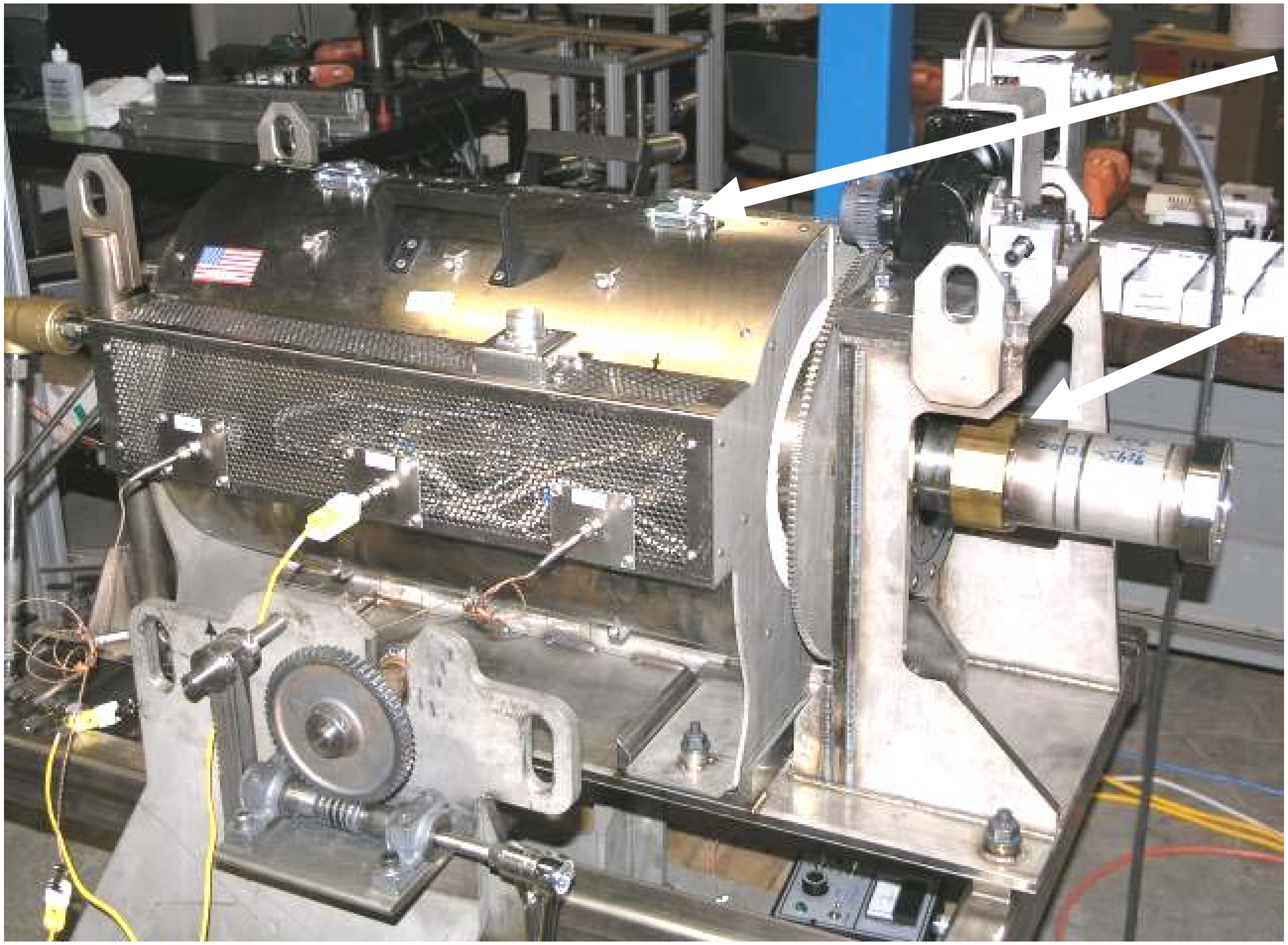








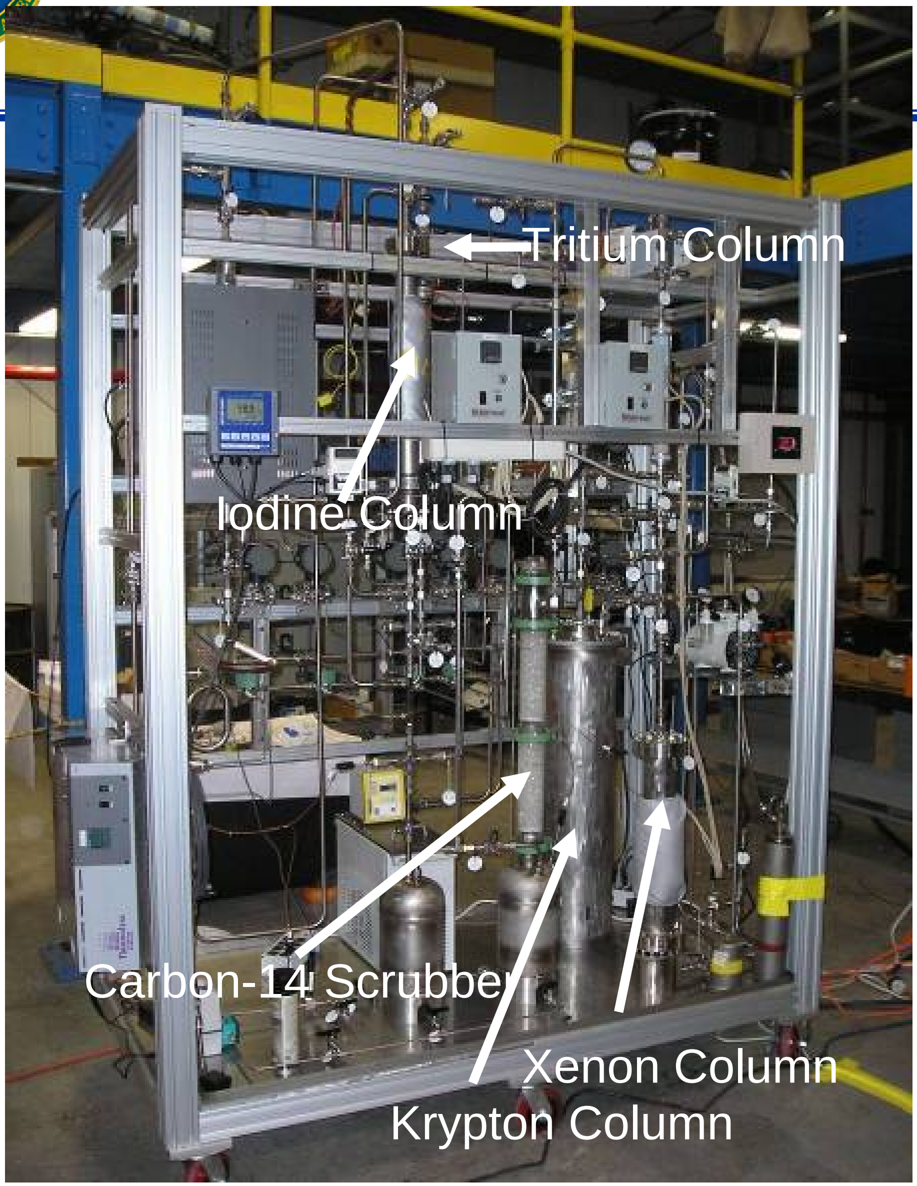
Voloxidation



Rotary Kiln w/ Heater in Place

Product Canister

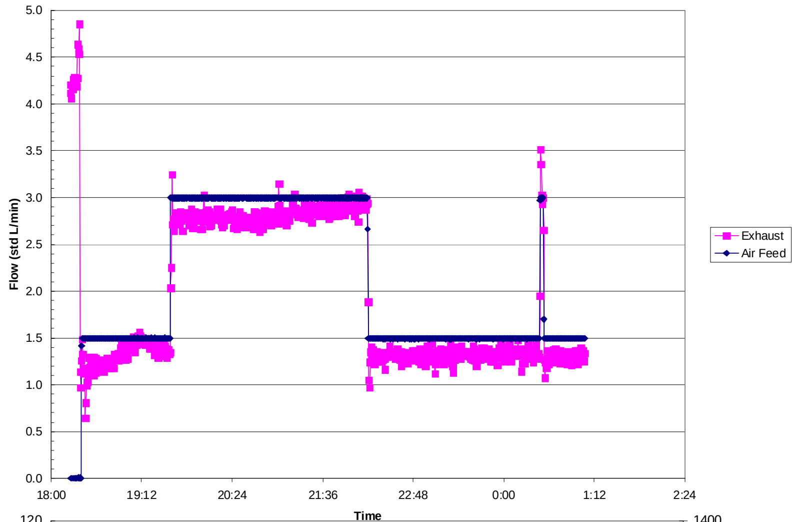
Voloxidizer Off-Gas Rack

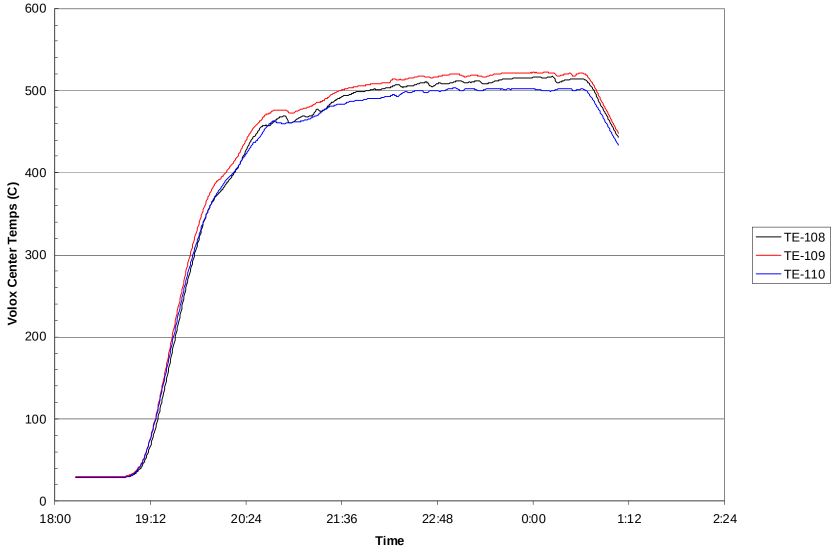


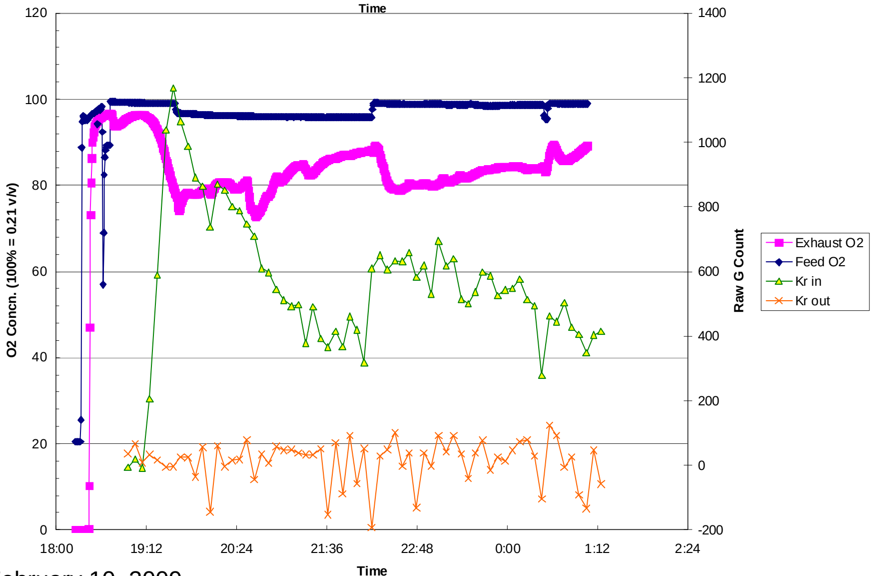
Voloxidation Run 2—Batch 1

- **First batch was comprised of Surry-2 fuel**
  - Initial enrichment 3.11%
  - Burnup: 36 GWd/MT heavy metal
  - Cooling time: 27 years (discharged from reactor in 1981)
  - Amount: 1704 g (316 g hulls, 1388 g fuel)
- **Experiment**
  - Fuel was oxidized in air at about 500°C
  - The run was prematurely terminated due to apparent cessation of oxygen consumption
    - Occurred after 4 hours of operation at temperature
    - Weighing of product powder and hulls showed that fuel was only 70% oxidized
    - There was potential for a cold zone near the closure flange, causing slow kinetics
  - Complete batch of fuel was re-run to finish the oxidation
    - Operation at temperature for 4 hours was done to ensure complete oxidation
      - 1542 g fuel powder recovered (indicates DU carry-over from shakedown run)
    - The voloxidizer was periodically tilted from the horizontal to cause fuel to migrate from the cold zone into the hot furnace zone
    - Both oxygen consumption and ^{85}Kr evolution was used to monitor progress and reaction endpoint

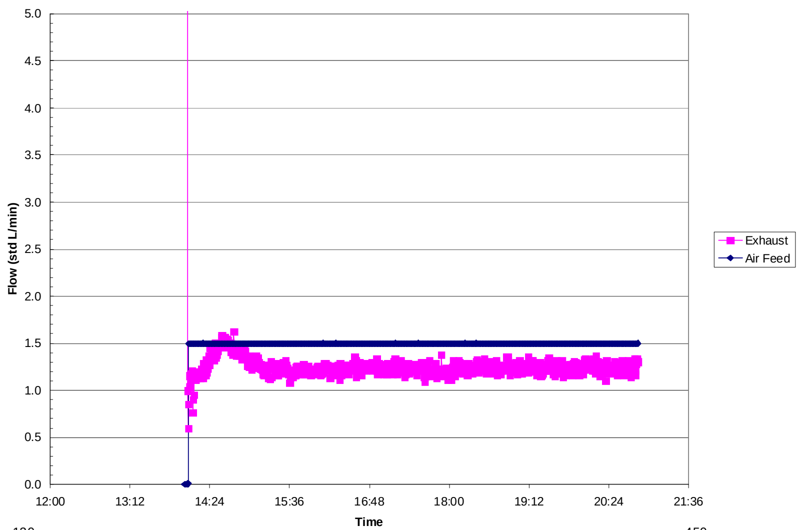
Run 2—Batch 1—Part A
O_2 Concentration, Air Flow, and Temperatures

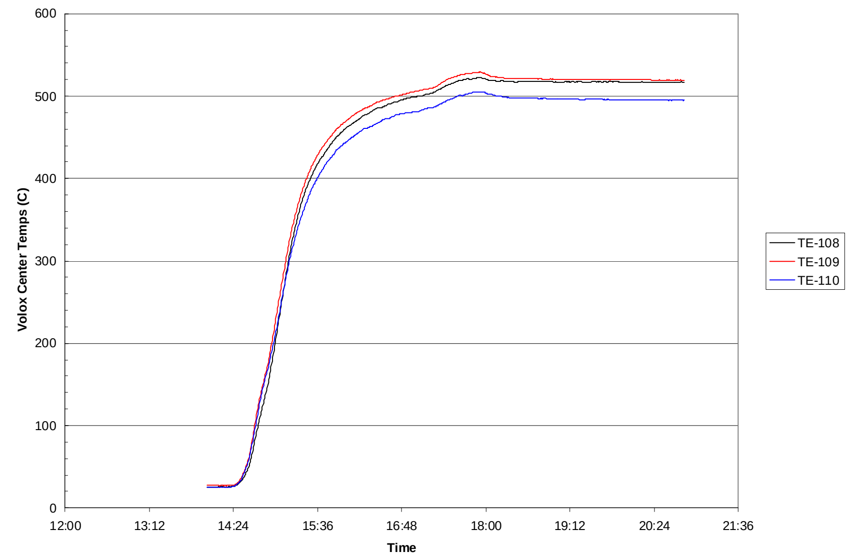


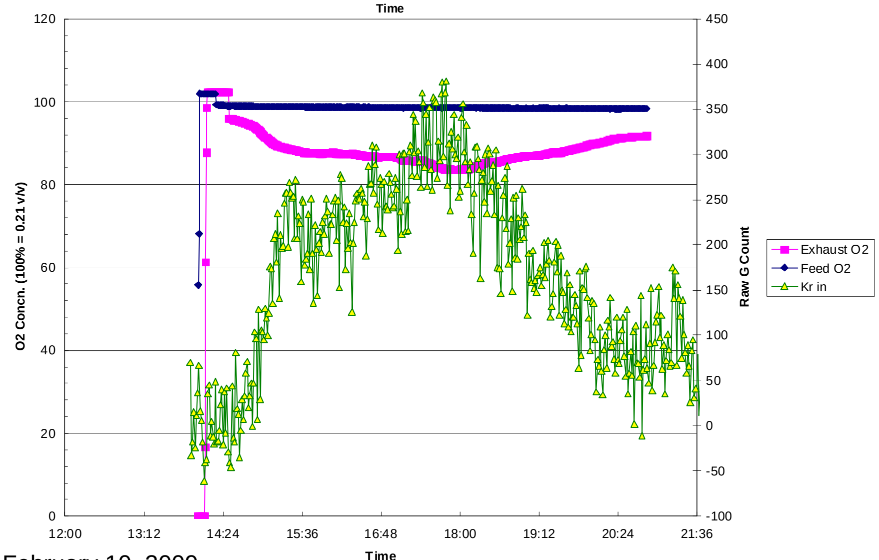




Run 2—Batch 1—Part B
O_2 Concentration, Air Flow, and Temperatures







Summary of INL Off-Gas Work

- **FY08: Initiated bench-scale iodine capture studies using nonradioactive constituents**

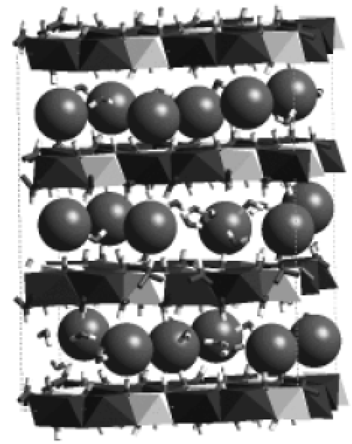
- **FY09: Continue iodine capture work:**
  - Use commercial sorbent IONEX Type Ag 900
  - Determine DFs under varying conditions (iodine concentration, co-constituents, temperature, residence times)
    - *Dissolver off-gas may contain iodine at ~10 ppm*
    - *Combined vessel off-gas may contain iodine at ~10 ppb*
  - Determine sorbent capacity

Summary of INL Off-Gas Work

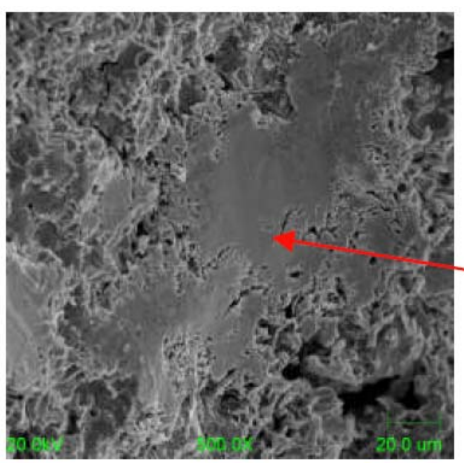
- **FY09: Install additional sorption columns for water vapor, CO_2 and Xe (ambient temperature operations)**
  - An integrated bench-scale treatment unit
  - Currently plan to run with nonradioactive constituents
  - Establish baseline capture of water vapor, CO_2 and Xe on commercial adsorbents
  - Test alternative sorbents
  - Design Kr capture system, which may be added in future

Encapsulants and Glasses as Iodine Waste Form

- **Bismuth-containing Encapsulants**
  - Two Trademark Applications for US Patents submitted;
    - *2007, 2008*



Bi-I-O *in situ* layered oxides



- **Low-Temperature Glasses for encapsulation of AgI, I_2 or AgI-MOR**
  - Glass transition temperatures lower than sublimation temperatures of AgI or I_2
  - Glass compositions that enhance iodine retention

Materials for Iodine Waste Streams - FY09 Plans at SNL

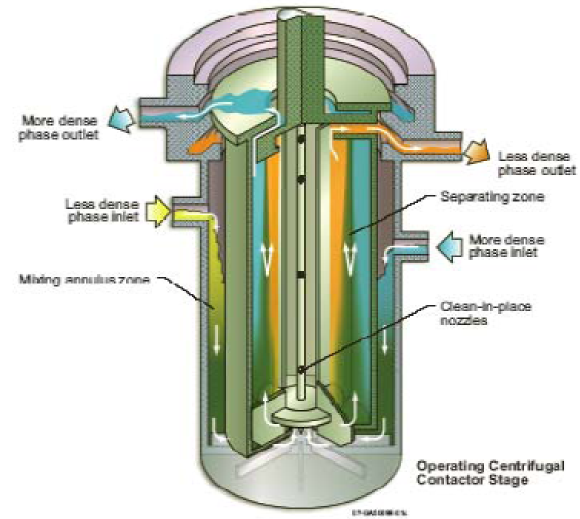
- **Sorption of I_2 gas into traps for encapsulants**
  - Low-temperature glasses
  - Alternative waste forms
- **Primary focus will be on "standard" iodine loaded AgZ materials from ORNL and INL surrogate studies and CETE Hot tests**
  - Retention of iodine
- **Alternate sorbents or recovery operations to produce better waste forms**

Off-gas Treatment Take-aways

- **Multiple air emission regulations may apply to spent fuel reprocessing facilities**
- **Gaseous fission product control efficiencies up to 99.99% (DF = 10,000) may be required**
- **Technologies for controlling gaseous fission products are less established; new technologies and waste forms are under evaluation**
- **Ongoing R&D is continuing to determine performance, operating conditions, and waste forms for gaseous fission product control technologies**

Role of Centrifugal Contactors in Aqueous Separations

- **Centrifugal contactors will likely play a significant role in advanced aqueous recycle of used nuclear fuel**
  - Utilized in portions of production scale facilities
  - Used for development of advanced separation processes
- **Have several advantages over pulse columns and mixer-settlers**
  - Less space required
  - High efficiency
  - Reach steady-state rapidly, allowing for rapid recovery from upsets and shutdown/restart
  - Reduced solvent contact times



- **But not without a few disadvantages**
  - Processes with slow kinetics
  - Processes with likelihood of large quantities of solids (clean-in-place capability has potential to mitigate)
  - Remote maintenance

Commercial Centrifugal Contactor Testing at the INL in 2008

- Testing of remotely operable/maintainable 5-cm contactors
- Design of remotely operable/maintainable "production" scale 12.5-cm contactors

- Design and construction of 30 stage 5-cm contactor pilot plant
- Temperature profile testing in the newly constructed contactor pilot plant

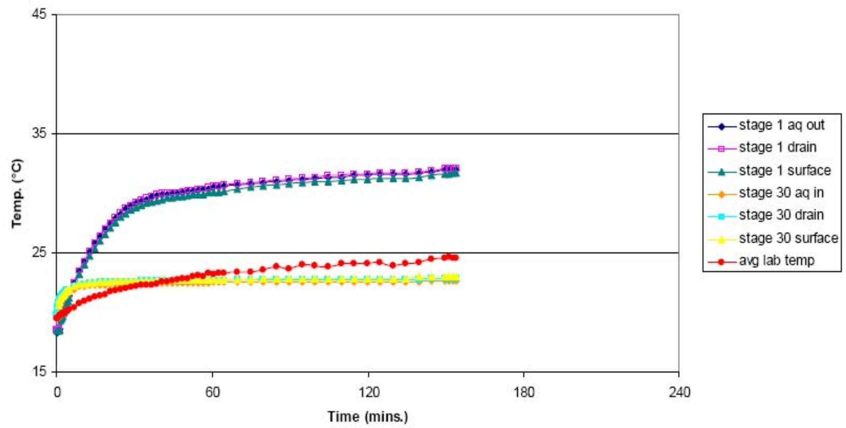
Development of Remote 5-cm Centrifugal Contactor

30 Stage 5-cm Centrifugal Contactor Pilot Plant

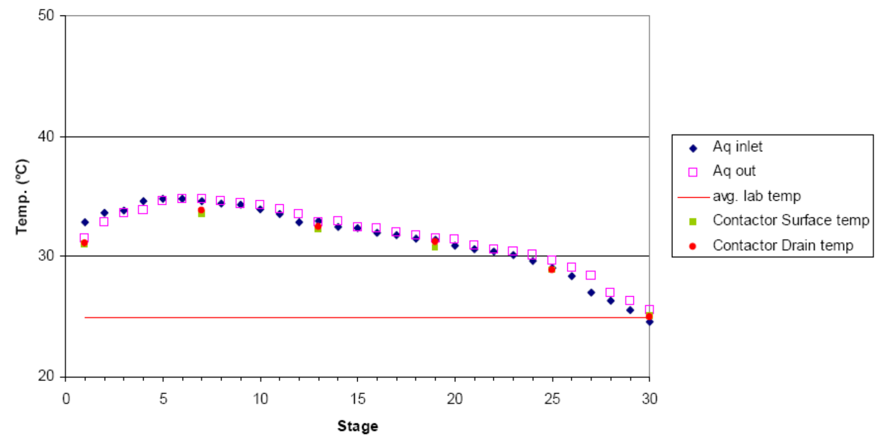
Test Objectives of Temperature Profile Testing in 30 Stage Pilot Plant

- **Aqueous separation flowsheets typically require some level of temperature control – often different for various sections**
- **Laboratory scale centrifugal contactor testing (2-cm) results in large temperature increase for the process solutions due to heat generated from the motors and low flowrates**
  - 2-cm centrifugal contactors with heat exchanger jackets were designed and utilized to alleviate this issue
- **Will jacketed heat exchangers be required for engineering and production scale centrifugal contactors?**
  - With larger flowrates it is expected that the temperature impact from motor heat will be reduced
  - Heating or cooling the process feed solutions may be enough to accomplish temperature control, preventing the need for a complex heat exchanger system for the centrifugal contactors
  - ANL has developed a computer model to predict process temperature based on system design. Limited experimental data is available - Data from testing will be used by ANL to validate/improve their model.

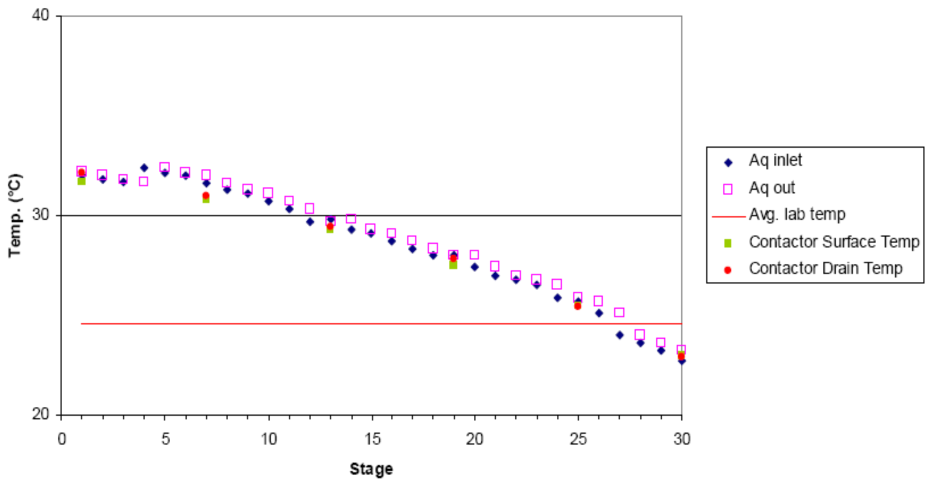
Temperature Profile Testing



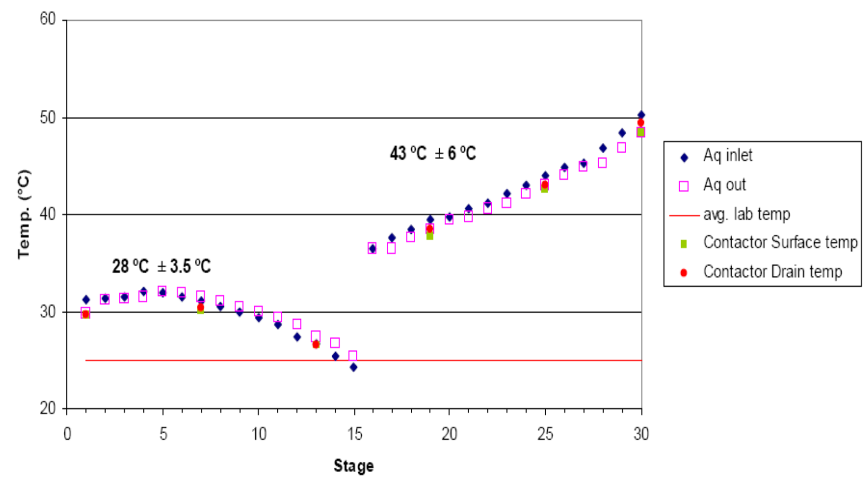
Aqueous Only – Approach to Steady State – Ambient Temp



Two Phase Flow – Temperature Profile – Ambient Temp



Aqueous Only – Temperature Profile – Ambient Temp



Two Phase Flow – Temperature Profile – Strip at 50C

Summary of Results of Temperature Profile Tests

- **Process solution temperature increased when feed solutions were at ambient temperature**
- **At feed temperatures of 50 ºC, heat losses were greater than the heat gain due to motor operation or heat of mixing which resulted in processes solution temperature decreasing below the feed temperature**
- **Control of the feed solution temperature has a significant impact on process solution temperature**
- **Process temperature control in flowsheets using CINC 5-cm centrifugal contactors could likely be accomplished for many flowsheets by controlling the temperature of the feed solutions**
- **Processes that require tighter temperature control may require jacketed contactor or insulation**

Testing Planned for FY 2009

- A prototype remotely maintainable 12.5-cm centrifugal contactor will be constructed and tested
- TRUEX mass transfer testing and temperature control testing will be performed using the 30 stage centrifugal contactor pilot plant
- A location and layout that will allow testing of the pilot plant with depleted uranium and/or low levels of radiotracers will be evaluated
- A pulse column will be set up for testing
  - Will be moved from the University of Idaho
  - Two inch glass pulse column that has approximately 20 feet of active height and upper/lower disengagement heads
  - Equipped with nozzle plates and an air pulser

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Questions

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