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NEAMS Safeguards and Separations IPSC FY-10 Overview

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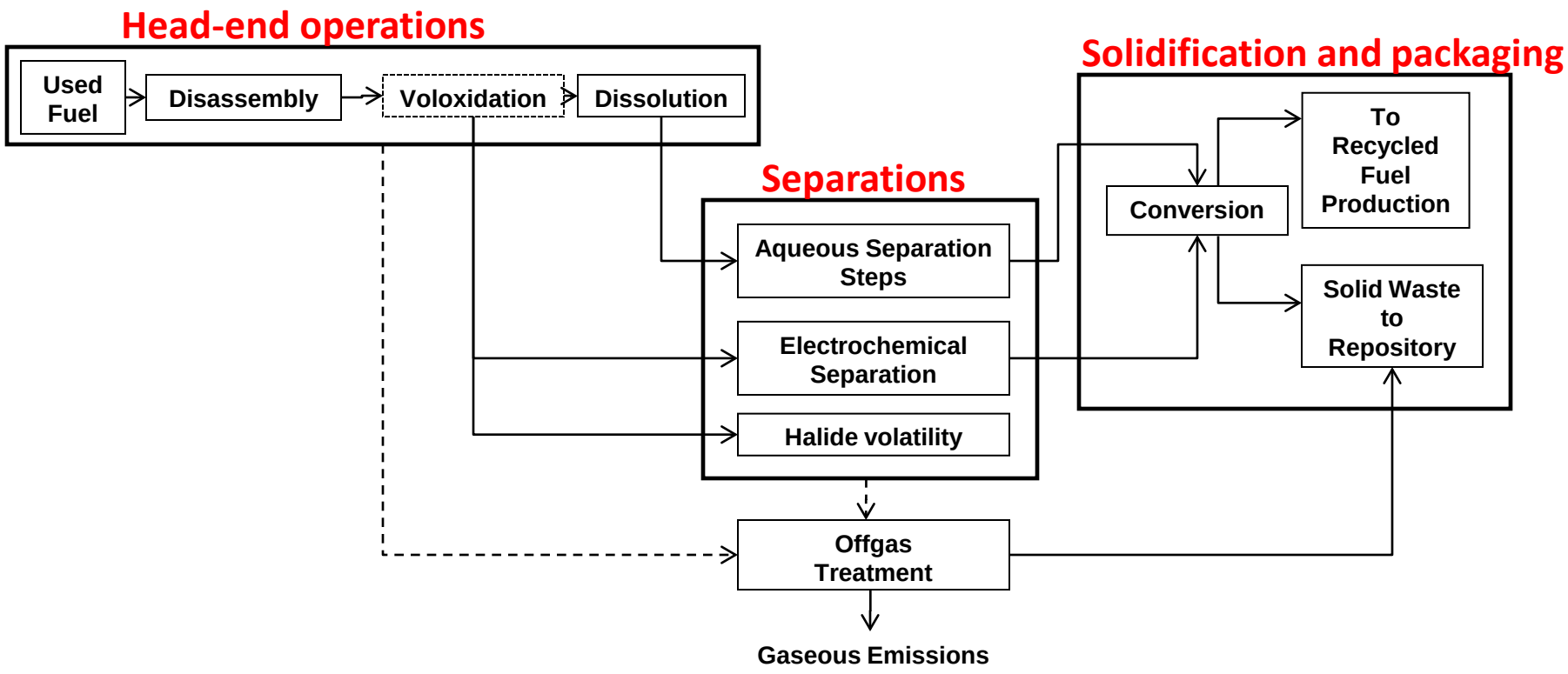
Washington DC
October 18-20, 2010

Scope


Head-end operations
Used Fuel
Disassembly
Voloxidation
Dissolution
Separations
Aqueous Separation Steps
Electrochemical Separation
Halide volatility
Solidification and packaging
Conversion
To Recycled Fuel Production
Solid Waste to Repository
Offgas Treatment
Gaseous Emissions

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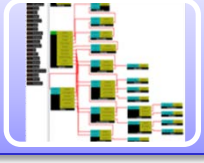
SafeSeps IPSC Elements

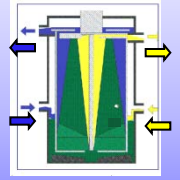
Plant-Level Toolkit

Set of component codes (toolkit) and a scheme for building a flexible plant simulator able to couple all processes, instrumentation, and analyses.



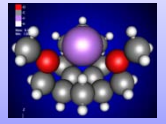
Unit Operations Components

Underlying codes for plant toolkit at the unit operations tier level. Prioritized development of components in the major sub-systems of the plant.



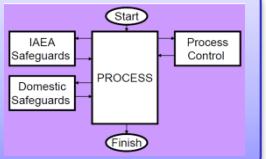
Subscale Modeling

Subscale simulation codes for calibration of unit ops components. Length and time scales varying from atomic to continuum.



Safeguards Performance Modeling

Support Safeguards by Design and NRTA approaches
Process monitoring models
Diversion path analysis
Material attractiveness analysis



Walk-through models

Virtual facility models to enable design and analysis
Incorporate process performance data and monitoring data in visualization tool

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SafeSeps IPSC: Near-term and Long-term Goals

Plant-Level Toolkit	Integrated end-to-end code with preliminary models	High-fidelity end-to-end code
Unit Operations Components	Preliminary models (mostly empirical)	Advanced models (informed by PIRT)
Subscale Modeling	Agent design software improved by quantum chemistry	Predictive capability for unit operations and tools for process design; Support campaign needs
Safeguards Performance Modeling	Simple models for instrument performance	Advanced performance models
Walk-through models	Preliminary end-to-end visualization capability	Virtual facility models that incorporate process performance data and monitoring data

Prioritizing M&S Needs

System-level requirements for a reprocessing plant: safeguards; separations

↓

M&S Requirements for safeguards; separations

→

Phenomena Identification and Ranking Table (PIRT) analysis

↓

Prioritized list of phenomena for M&S focus

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PIRT analysis

- **PIRT analysis was done in FY10 to identify and rank phenomena for importance and knowledge gap**
- **SME participation from Sep/WF and MPACT Campaigns**

Subsystems Considered

Fuel receipt	Clarification	Cross-cutting
Disassembly/ shearing	Feed Prep	Conversion
Voloxidation	Accountability tank	Off-Gas Treatment
Dissolution	Separation	Secure storage
Hulls washing	Pyrochemistry	Others

Figures of Merit For Importance

A. Proliferation

B. Performance

C. Safety and health

D. Economics

Knowledge Gap

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Importance = High and Knowledge Gap = High (1/3)

Subsystem	Component	Phenomenon
Fuel receipt	Fuel assemblies	Isotopic inventory
		Instrument models
Dissassembly/shearing	Disassembly/shearing	Gas release
		Fissile product content (pyro case)
Voloxidation	Off-gas filtration system	Deposition of semi-volatiles
Aqueous separation	Solvent Extraction	Solution thermodynamics – molecular level
		Mass/momentum/heat transfer predictive
		Radiolytic and hydrolytic stability
	Ion exchange	thermodynamics – molecular level
		Mass/momentum/heat transfer predictive
		Radiolytic and hydrolytic stability
	Crystallization/ Precipitation	thermodynamics – molecular level
		Mass/momentum/heat transfer predictive

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Importance = High and Knowledge Gap = High (2/3)

Subsystem	Component	Phenomenon
Non-aqueous	Pyrochemistry	Performance model
		Process monitoring
Halide volatility	Process monitoring	Reactive gas (flow rates, composition-inlet and outlet)
	Containment	Physical structure
	Chemical reactor	Thermodynamics
		Kinetics
		Pu and Np volatility
	Gas processing system	G/S separation
		Deposition of volatiles
		Recycle/disposal of reactive gas
Conversion	Nitrate-to-oxide	Nucleation, growth and aggregation
	Calcination	Nucleation, growth and aggregation

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Importance = High and Knowledge Gap = High (3/3)

Subsystem	Component	Phenomenon
Off-Gas treatment	Adsorption (applies to C14, I)	Reaction diffusion models – semi-empirical
	Absorption (applies to Krypton and Tritium)	Reaction diffusion models (semi-empirical)
MC&A	Integrated MC&A	Integration of process monitoring
		Security systems
	Hot cell	3-d physical representation

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Importance = High and Knowledge Gap = Medium (1/3)

Subsystem	Component	Phenomenon
Operations for other fuel types	VHTR	Graphite fuel disassembly
		Graphite fuel disassembly
	Oxide fuel head end for pyro	Oxide reduction
Voloxidation	Process monitoring	Mass flow
	Kiln	Heat transport model
		Fraction of G/S (empirical)
		Coupled heat/mass transfer model
Dissolution	Dissolver	G/S/L chemistry model-engineering
	Hulls washing	G/S/L chemistry model-macro
Feed prep	Prep vessel	Chemistry (Incl. precipitation, redox)
Accountability tank	Instrumentation	Process monitoring
	Crystallization/ Precipitation	thermodynamics – molecular level

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Importance = High and Knowledge Gap = Medium (2/3)

Subsystem	Component	Phenomenon
Aqueous separation	Solvent Extraction	Solution thermodynamics – semi-empirical
		Equipment performance model
	Ion exchange	thermodynamics – semi-empirical
		Equipment performance model
		Gas generation
	Crystallization/ Precipitation	thermodynamics – semi-empirical
		Continuous/batch operation
		Equipment performance model
		Transient behavior
Non-aqueous	Pyrochemistry	Electrohydrodynamics
		Electron transfer/ reactions
		Thermodynamics
Halide volatility	Process monitoring	Mass flow
	Chemical reactor	Heat transport model
		Fraction of G/S (empirical)
		Coupled heat/mass transfer model

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Importance = High and Knowledge Gap = Medium (3/3)

Subsystem	Component	Phenomenon
Cross-cutting	Cross-cutting	Corrosion and material compatibility
		Process monitoring and control
		Pipe flow model
		Tank model
Conversion	Nitrate-to-oxide	Thermodynamics, kinetics
		Product characterization, material accountability
	Calcination	Thermodynamics, kinetics
		Product characterization, material accountability
Off-Gas treatment	Adsorption (applies to C14, I)	Macroscale model of decontamination factor
	Absorption (applies to Krypton and Tritium)	Macroscale model of decontamination factor

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Challenge Problems - Summary

Challenge Problem	Elements
Separations Process Models – Enable a significant advance in modeling of separations processes: Multiphysics modeling of unit operations in separations processes, informed by experimental data.	*Aqueous:* centrifugal contactor - enable prediction of stage efficiency for given conditions
	E-chem: electrorefiner, including thermodynamics, kinetics; current efficiency; contamination effects
	Dry process: G/S system characteristic of voloxidation, halide volatility
Dynamic plant modeling capability – demonstrate capability for flexible integration of plant level process and monitoring codes	The capability will be demonstrated first for a PUREX plant, because of the relative abundance of existing data.
Advanced Monitoring Models – Develop performance models for advanced monitoring technologies – including those under development in MPACT	Performance of HKED, Lead Slowing Down Spectroscopy, Ultra-High Resolution Spectrometry/ Microcalorimetry. Other system-level approaches such as neutron balance.
Agent Design to Process Design - Extend capability of molecular modeling to separations systems	Solvent extraction – identify candidate agents, predict distribution coefficients, interfacial transport
Off Gas Modeling – Develop molecular models of phenomena of importance in gas-treatment systems	Fundamental mechanisms of capture and mass transport in silver-based and novel sorbents

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Safeguards & Separations Challenge Problem 1:
Separations Process Models

STATUS QUO

US Modeling and Simulation tools for reprocessing plants do not involve multi-scale models for dominant phenomena

- **Models do not exist for many key elements and processes**
- **Existing models lack needed fidelity**

NEW INSIGHTS

Effort directly addresses R&D Objectives 3 and 4

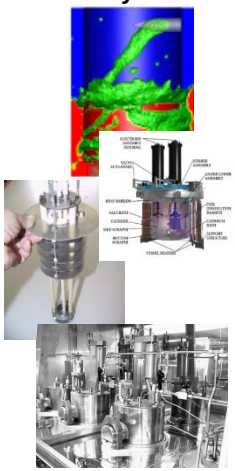
Advanced modeling and simulation will enable a significant improvement in the capability to predict performance of key separations processes.

Models will also support safeguards by design and other non-proliferation objectives.

Enable a significant advance in modeling of separations processes: Multiphysics modeling of unit operations in separations processes, informed by experimental data.

Predict performance of separations processes

- *Aqueous:* Enable prediction of stage efficiency of a solvent extraction contactor for given conditions
- *Electrochemical :* Enable prediction of electrorefiner performance by modeling thermodynamics, kinetics; electrohydrodynamics including interfacial effects; contamination effects
- *Alternative dry processes:* gas/solid systems characteristic of voloxidation, halide volatility processing.



Engaged FCR&D Stakeholders:

- Separations and Waste Forms Technical Area
- Materials Protection, Accounting, and Control Technology (MPACT) Technical Area

Important Milestones:

- Develop high fidelity contactor performance model (FY14)
- Integrated electrochemical cell model (FY11)
- High fidelity electrochemical cell model incorporating key phenomenological models (FY15)
- Halide volatility performance model (FY15)

DATA NEEDS

Data needed for validation:

Data from relevant AFCI/FCR&D experimentation

Current/Planned FCR&D Experiments

- Contactor CFD validation using electrical resistance tomography and other methods
- Electrorefiner performance data
- Voloxidation experiments
- Dissolution experimentsd

END-OF-PHASE GOAL

High-fidelity models for performance of key separations processes; validated by experimental data obtained in coordination with FCR&D Campaigns:

- **Aqueous**
- **Electrochemical**
- **Dry Processes**

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Safeguards & Separations Challenge Problem 2:
Demonstrate Dynamic Plant Modeling Capability

STATUS QUO

US Modeling and Simulation tools are not suitable for optimizing a separations plant with integrated safeguards

- **Operational data exist but existing models lack needed fidelity**
- **Models are not fully integrated**

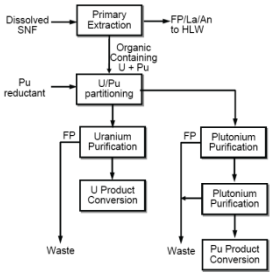
NEW INSIGHTS

Effort Directly Addresses R&D Objectives 3 & 4

Demonstration of a full plant for PUREX plant performance. Extension to additional flowsheets will facilitate evaluation of a variety of advanced fuel cycle options and drive down uncertainties in estimating the material inventory at any location in the plant.

Develop an integrated plant model that allows dynamic simulations of the performance of a separations plant and integrated safeguards. Demonstrate capability with a PUREX plant flowsheet.

- PUREX is chosen as the pilot plant for modeling because of the relative abundance of data.
- The model will initially incorporate relatively simple models for each process – including legacy models where available.
- Code will incorporate the capability to bridge to subscale models to provide required fidelity in chemical and physical processes.



Engaged FCR&D Stakeholders:

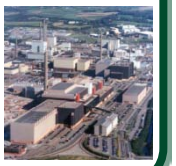
- Separations and Waste Forms Technical Area
- Materials Protection, Accounting, and Control Technology (MPACT) Technical Area

Important Milestones:

- Perform PIRT analysis to rank relevant phenomena and to guide detailed modeling. (FY10)
- Demonstrate prototype on PUREX flowsheet (FY12)
- Demonstrate integration of detailed multiscale models for prioritized unit operations and safeguards technologies (FY15)
- Demonstrate connection to fundamental predictive models for properties and other process parameters required (FY17)
- Demonstrate plant visualization capability (FY17)
- Demonstrate an end-to-end simulator for one scenario: processing of UOX spent fuel (FY18)

DATA NEEDS

PUREX plant operational data available from DOE sites, industry, or international partners.



END-OF-PHASE GOAL

Demonstration of the dynamic plant model for a PUREX plant.
Set stage for extending capability to advanced flowsheets

Sustainable Fuel Cycles

Reduced Proliferation Risk

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Safeguards & Separations Challenge Problem 3: Safeguards Models

STATUS QUO

Reprocessing remains the hardest element of the fuel cycle to safeguard.

Integrated plant models do not currently include detailed safeguards performance models; current practice is largely to utilize statistical analysis based on measurement data.

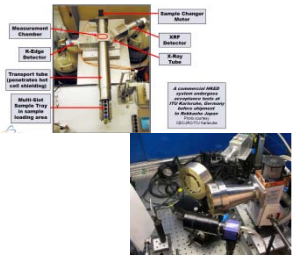
NEW INSIGHTS

Effort Directly Addresses R&D Objectives 4

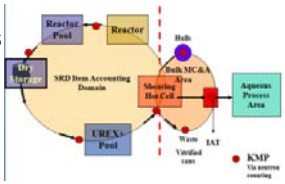
Utilize advanced modeling to drive down uncertainties in estimating the material inventory at any location in the plant. Develop the capability to address a broad range of input conditions for used fuel. Develop the capability to optimize instrumentation deployment to reduce safeguards costs.

Use advanced modeling methods to improve materials accountability capability in reprocessing scenarios.

- Develop models for performance of key process monitoring methods, including destructive assay (DA) and non-destructive assay (NDA) techniques. This includes techniques under development in the MPACT campaign, such as LSDS and HKED.



- Investigate integrated characterization methods such as neutron balance – measurement of the neutron emission rate from the curium in the spent fuel that can be used to quantify the associated fuel mass and the plutonium content



Engaged FCR&D Stakeholders:

- Materials Protection, Accounting, and Control Technology (MPACT) Technical Area

Important Milestones:

- Perform PIRT analysis to rank relevant phenomena and to guide detailed modeling. (FY10)
- Complete HKED modeling (FY11)
- Neutron balance studies (FY12)
- LSDS/other methods (FY13-FY15)

DATA NEEDS

Data on measurement uncertainties for different species in mixed solutions.

PUREX plant operational data available from DOE sites, industry, or international partners

END-OF-PHASE GOAL

Significant improvement in materials accountability during reprocessing.

Reduced Proliferation Risk

16

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Safeguards & Separations Challenge Problem 4:
Agent Design to Process Design

STATUS QUO

Highly selective and robust separating agents and processes are needed for effective and economically viable aqueous and alternative flowsheets in closed and modified open fuel cycles

NEW INSIGHTS

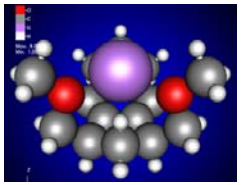
Emerging theory and computational tools can accelerate the development of separating agents and processes

- **Molecular models**
- **Multiphysics models of reaction and transport**

Develop tools to predict performance of separating agents and processes

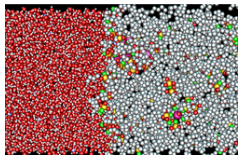
Molecular-level tools **to identify candidate separating agents**

- **Parallelized molecular mechanics codes**
- **Quantum mechanics for improved scoring**



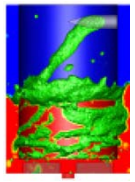
Molecular dynamics models **to predict performance of agents and processes**

- **Distribution coefficients**
- **Interfacial transport kinetics**



Continuum models **to predict performance of separation processes**

- **Transport in multiphase solvent extraction systems**



Engaged FCR&D Stakeholders:

- Minor Actinide Separation Sigma Team of Separations and Waste Forms Technical Area

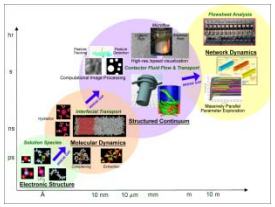
Important Milestones:

- Molecular mechanics agent design on massively parallel computing FY-10
- Agent design improved by quantum chemistry FY-12
- Improved design by computational thermochemistry FY-14
- Molecular interfacial transport of agents by design FY-16
- Microstructured mixture behavior of agents FY-18
- Plant-level module for extraction kinetics FY-19

DATA NEEDS

- **Results from Minor Actinide Separation Sigma Team experiments**
- **Fundamental data on physical properties, solution thermodynamics, etc.**
- **Developments from BES/ASCR studies**

END-OF-PHASE GOAL



Tools enabling predictive molecular design of separations processes

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Safeguards & Separations Challenge Problem 5: Off-gas Treatment

STATUS QUO

- No operating recycling plant in the world can meet all current or projected US regulatory requirements
- *Challenge:* Fuel recycling systems will need to capture volatile fission products from dilute streams with exceedingly high separation factors

NEW INSIGHTS

Process modeling can accelerate development of environmentally compliant recycle systems

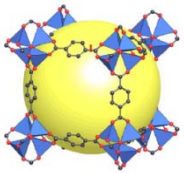
Models of key unit operations can generate dynamic source terms for material inventories and performance of off-gas treatment components.

Models will enable prediction of gaseous emissions of fuel recycling plants under a variety of alternative configurations and operating conditions.

Approach

Develop models of key unit operations to predict performance of off-gas treatment components and volatile emissions from fuel recycling plants. This effort comprises:

- Complexity – multiple multiphase reactive chemical and physical systems, with relevant phenomena from molecular to macroscale.
- Coupled phenomena – operations involving multiple reaction and transport processes; dynamic interactions between units, instrumentation, and analysis tools.
- New technical ground – models for most processes do not exist or lack the needed level of fidelity for design and evaluation. Subscale modeling may accelerate development of new separating agents for off-gas treatment.



Engaged FCR&D Stakeholders:

- Off-gas Sigma Team of Separations and Waste Forms Technical Area
- Materials Protection, Accounting, and Control Technology (MPACT) Technical Area

Important Milestones:

- PIRT process to deliver functional requirements – FY10
- Demonstrate prediction of iodine capture using initial modules for voloxidizer, dissolver, and offgas treatment – FY12
- High-fidelity models of critical processes – FY17
- Incorporate molecular-level models of advanced sorbents – FY19

DATA NEEDS

- **Results from Off-Gas Sigma Team experiments**
- **Fundamental data on physical properties, thermodynamics, kinetics, etc.**
- **Subscale modeling from FMM**
- **Developments from BES studies**

END-OF-PHASE GOAL

Options for optimized and environmentally compliant recycle systems

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FY11 Tasks are addressing
- high-priority items identified by the PIRT
- challenge problems

Plant-Level Toolkit

- Interface with NEAMS ECT/CT for architecture development
- Work with individual v0.9 module developers
- Demonstrate basic functionality with coupled unit operation modules

Unit Operations Components

- AMUSE integration into v0.9 structure
- Electrorefiner v0.9
- Dissolution v0.9
- Voloxidation v0.9
- Off-gas v0.9
- Requirements for NEAMS Solvent Extraction code
- Initial scoping of G/S systems

Subscale Modeling

- Integrate QM tools in HD
- Structure generation algorithms
- Contactor CFD model – surfactant, multiple droplets; Begin theory work to upscale
- Electrorefiner (requirements development, and initial efforts)
- Adsorption/absorption modeling

Safeguards Performance Modeling

- SSPM integration into v0.9 structure
- Dose calculation module
- Component models (Tank, piping)
- Complete HKED model
- Begin LSDS model

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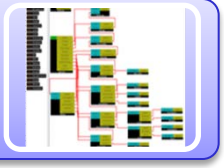
Presentations

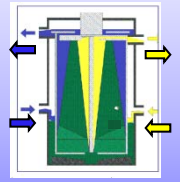
Plant-Level Toolkit

Architecture – Valmor de Almeida (ORNL)



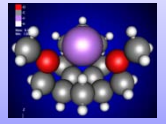
Unit Operations Components

Electrochemical processing model (ANL)
Dissolution (ANL)



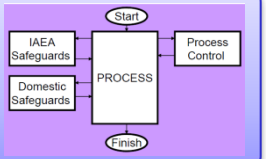
Subscale Modeling

Device-scale simulations – Kent Wardle (ANL)
Droplet dynamics: Marianne Francois (LANL)
Agent Design: Valmor De Almeida (ORNL)
Enrique Batista (LANL)



Safeguards Performance Modeling

Hybrid K-Edge Densitometry Modeling: Mike Collins (LANL)



Walk-through models