Waste Forms for an Advanced Nuclear Fuel Cycle

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Outline

- Background
- Waste stream description
- Process and waste form description
  - vitrification to form glass
  - melting to form metal alloy
  - ceramic processing methods and materials
- Example cost evaluation of waste forms
- Concluding remarks

Background

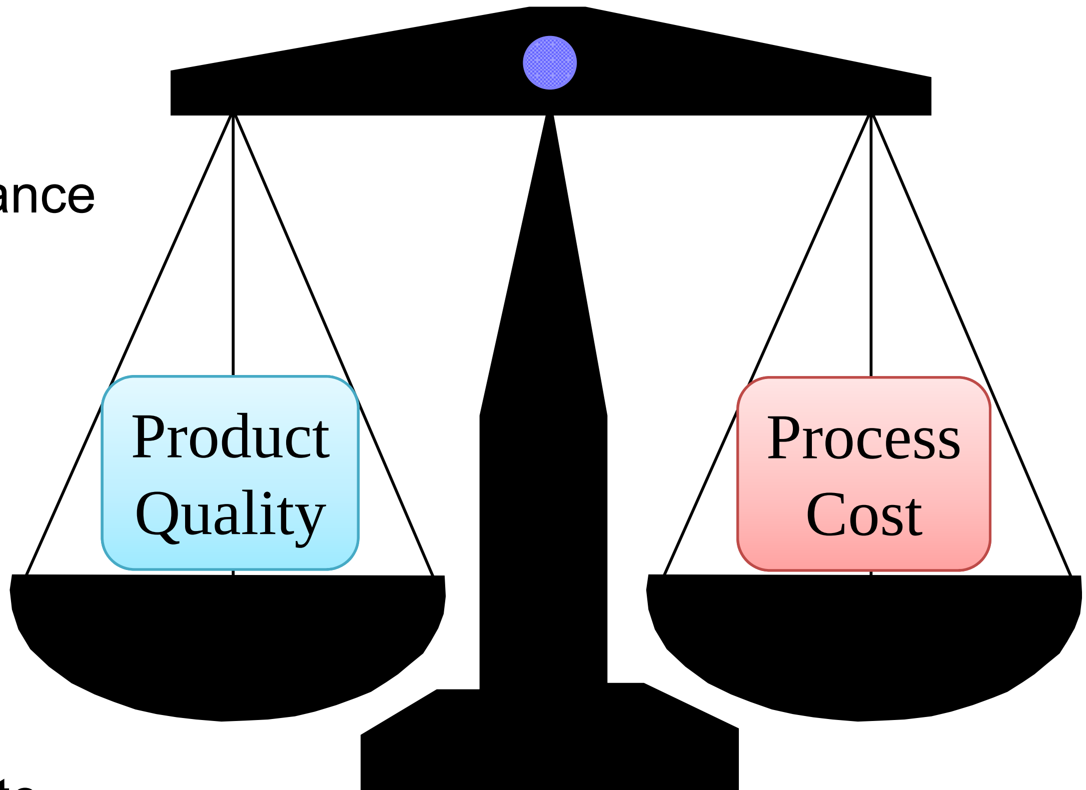
- Vitrification is the process of choice for separated highly radioactive wastes in virtually every reprocessing nation
  - vitrification is: 1) a proven process, 2) tolerant to wide range of waste compositions, 3) a fast continuous process, 4) generates no fine particulates, and 5) the EPA best demonstrated available technology
  - produces a waste form of good performance that is reasonably well understood
- An unprecedented level of waste management control can be achieved through advanced separations
  - separate streams by waste chemistry
  - each stream can be immobilized separately or combined with others
  - the waste forms can be selected to match the waste and disposal environment chemistries

Challenge

- How would we manage wastes from a U.S. nuclear fuel recycling process??

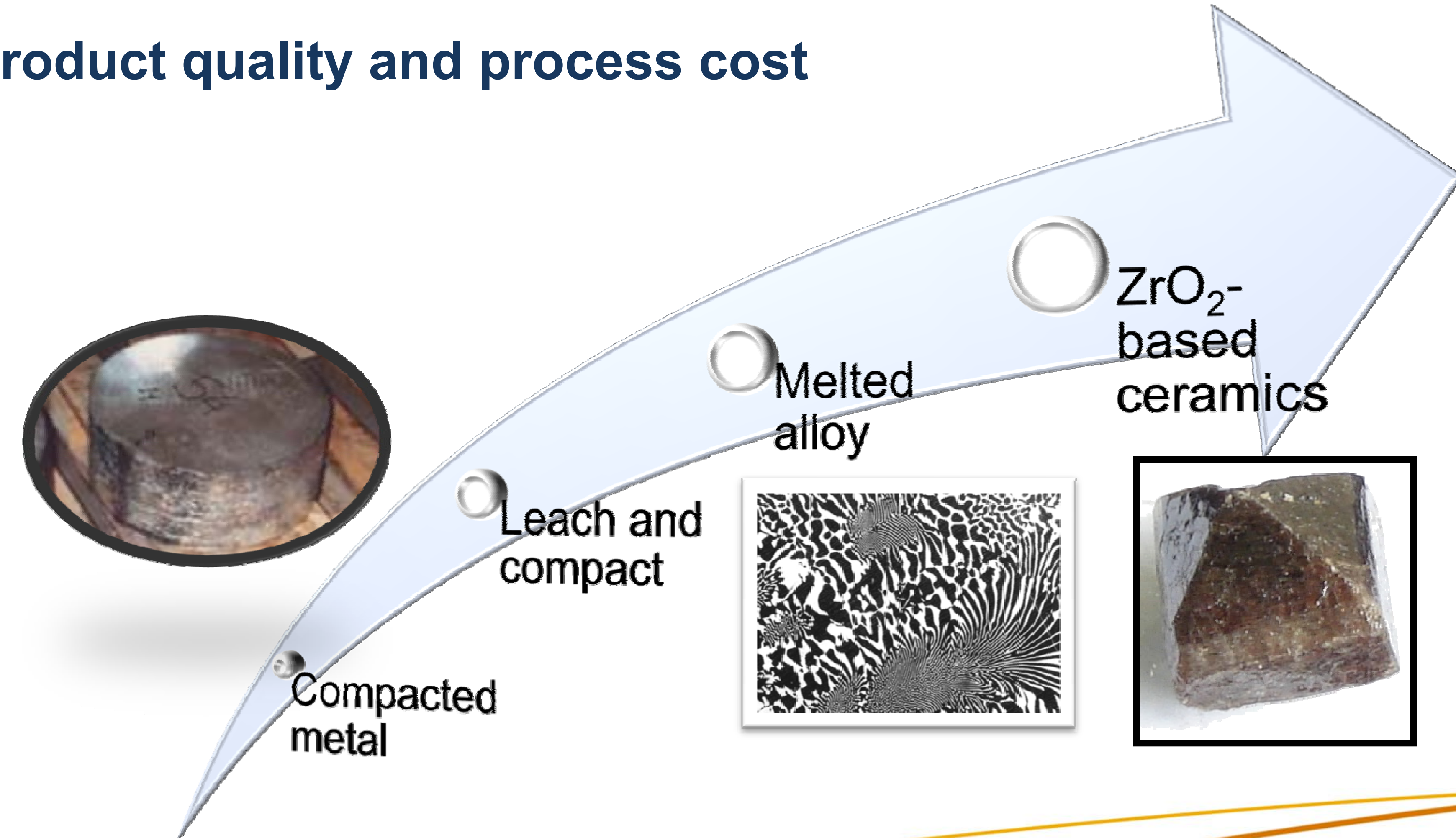
Waste Form Options

- Product quality
  - chemical durability
  - well understood performance
  - thermal stability
  - radiation stability
- Processing and cost
  - low waste form volume
  - small process footprint
  - continuous process
  - mature technology
  - minimum secondary waste
- Raises a critical question
  - how good is good enough?

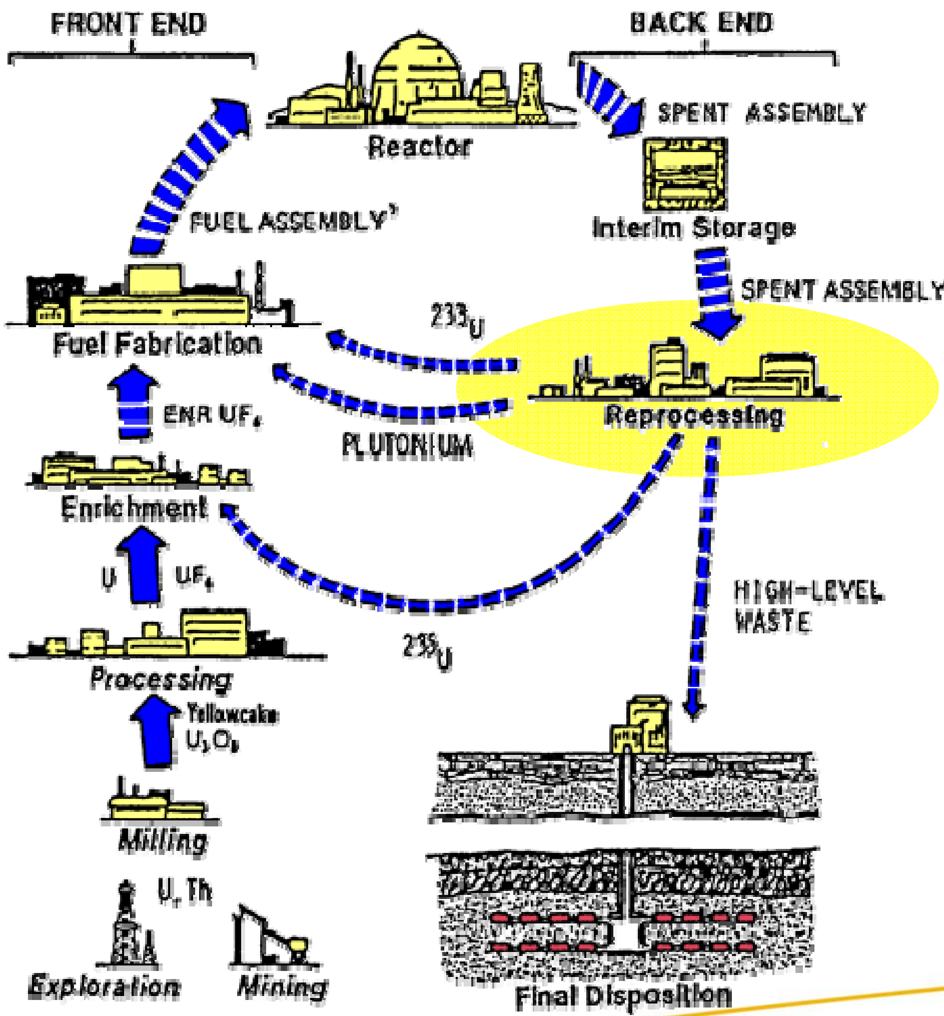


Example - Hulls and Hardware

Product quality and process cost



Wastes to be Treated



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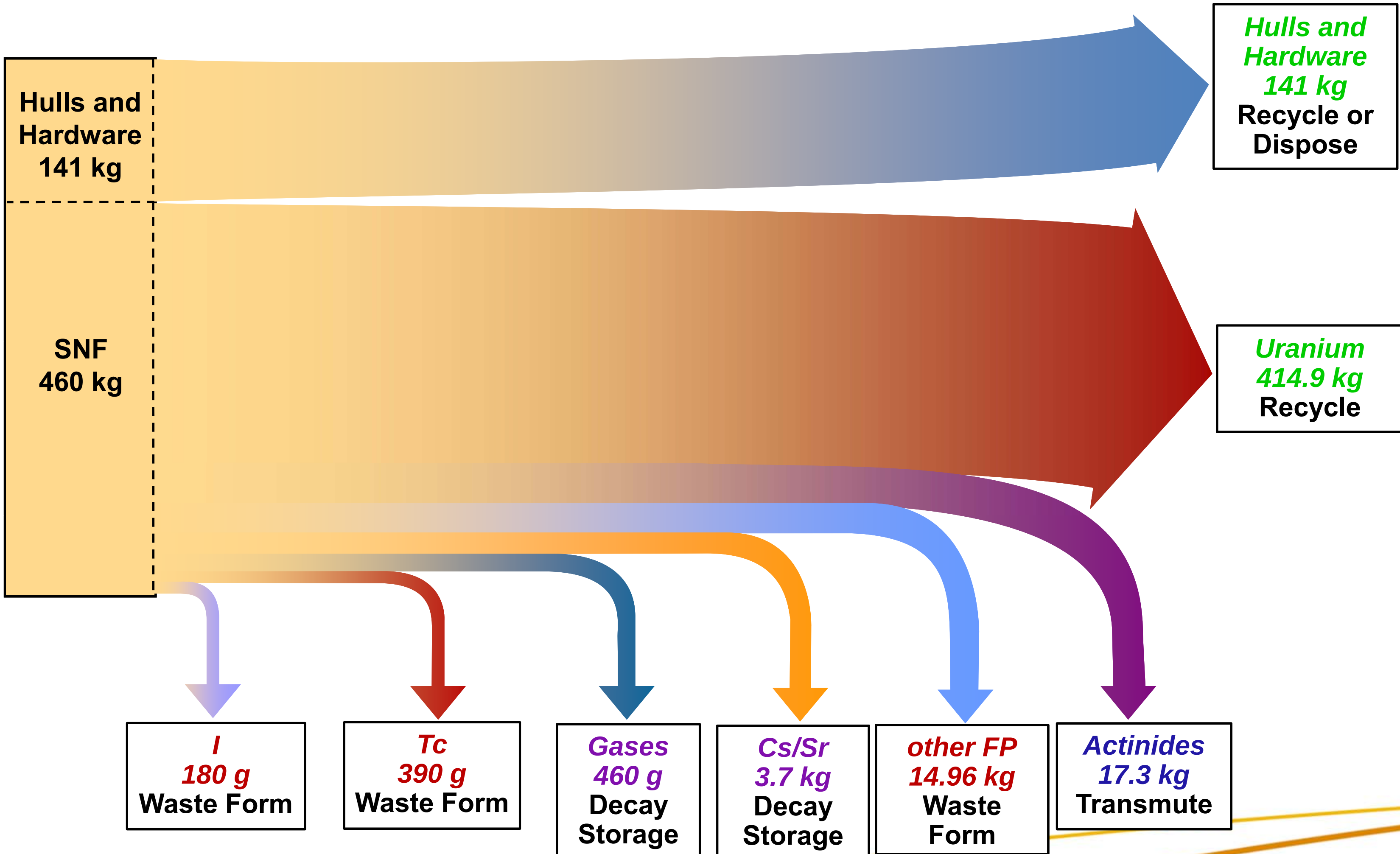
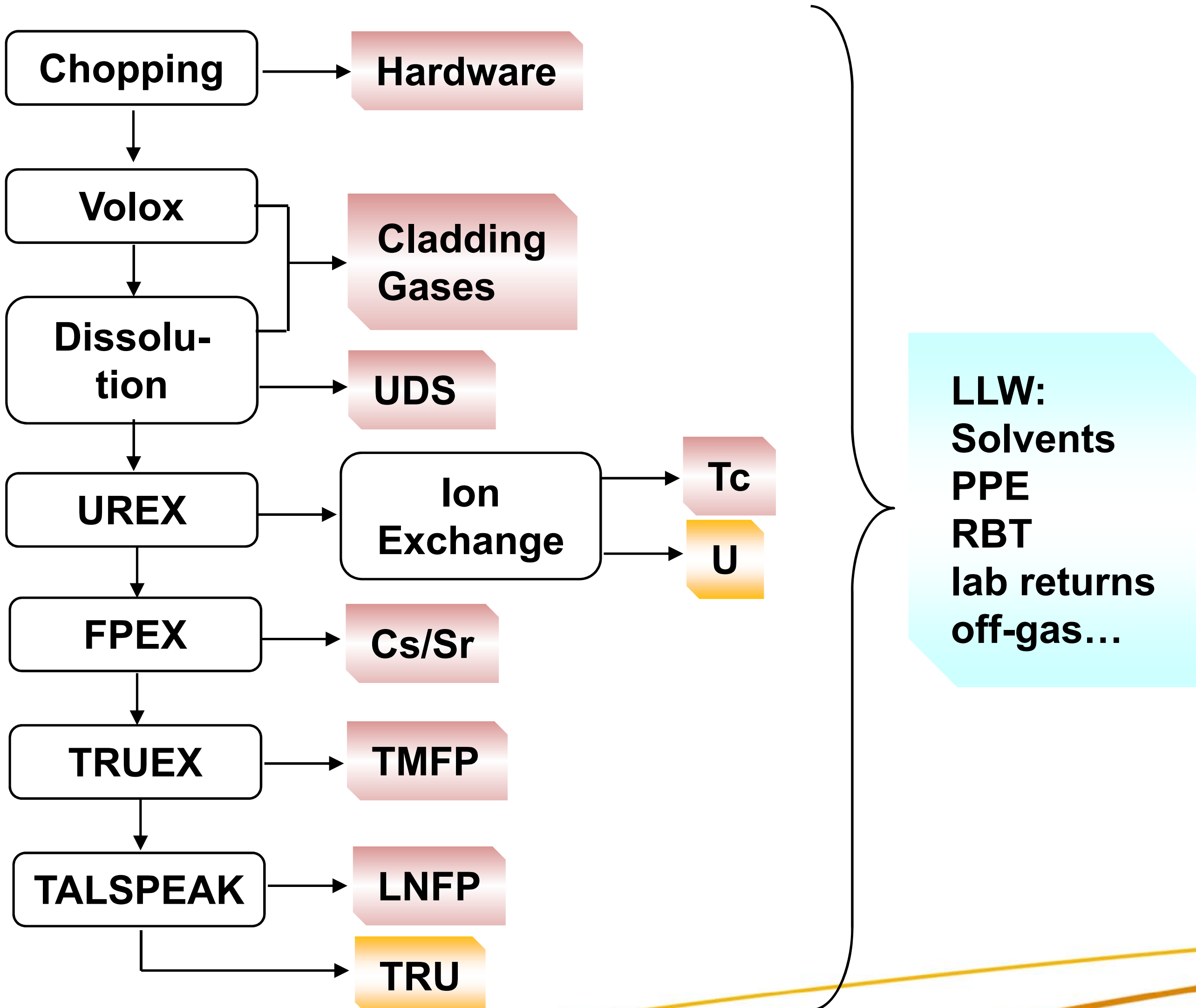
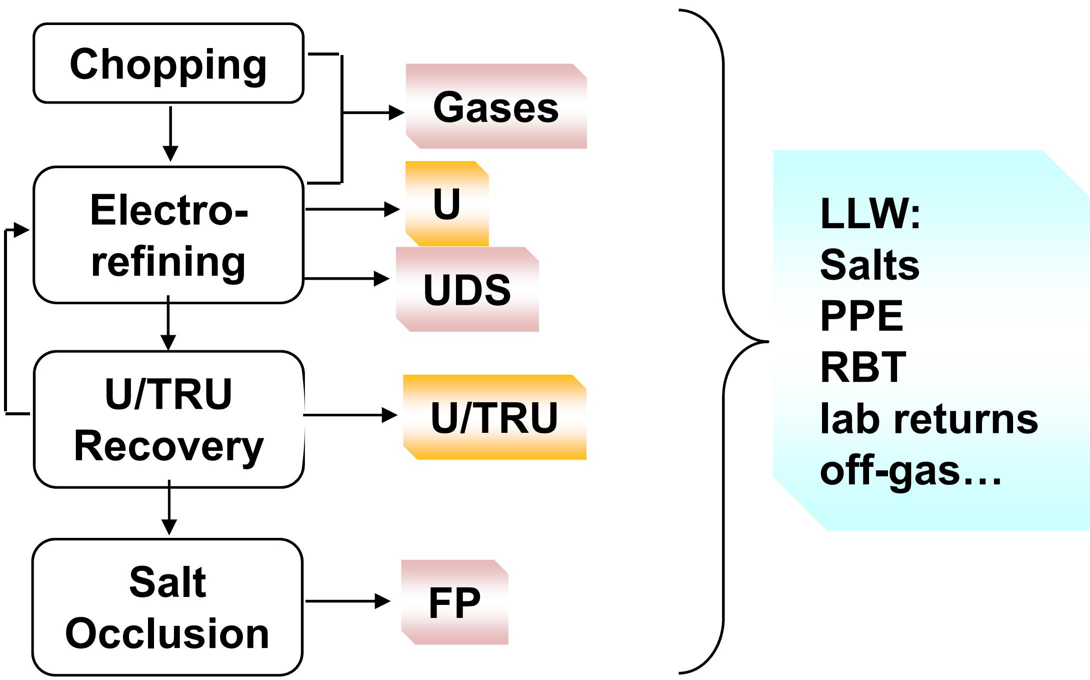
Fuel Components (Typical LWR Assembly)



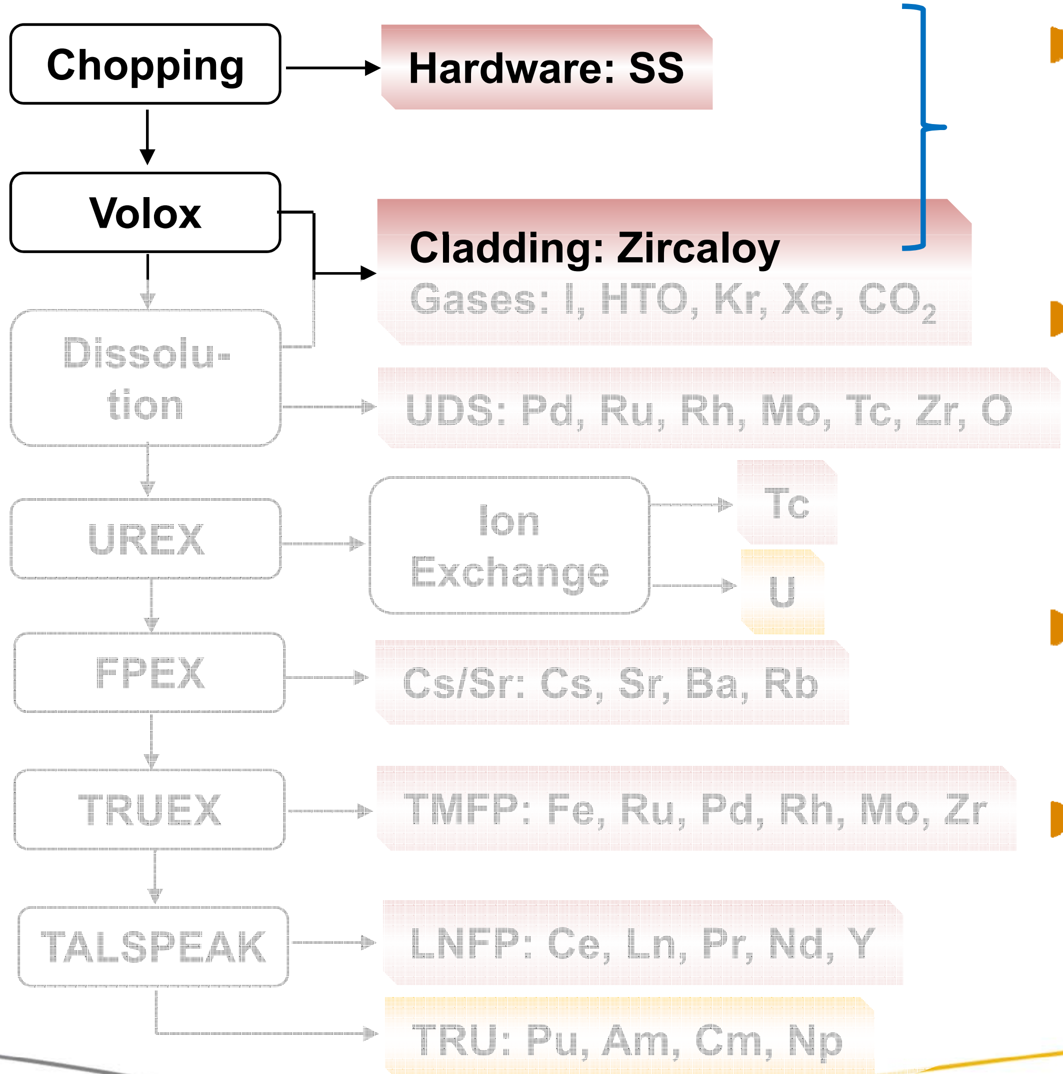
Potential Aqueous Process Waste Streams



Potential Electrochemical Process Waste Streams



Potential Aqueous Process Waste Streams



- Hulls and hardware are logically combined
  - they're metallic and contain activation products, ^{3}H, and embedded TRU
- Typical waste forms include:
  - compacted metal
  - Fe based alloy
  - Zr based alloy
  - zirconia rich ceramic (e.g., zircon)
- Combined wastes
  - UDS, TMFP, and/or Tc if in alloy
  - LNFP, TRU, Tc, UDS if in ceramic
- Strong potential for recycle

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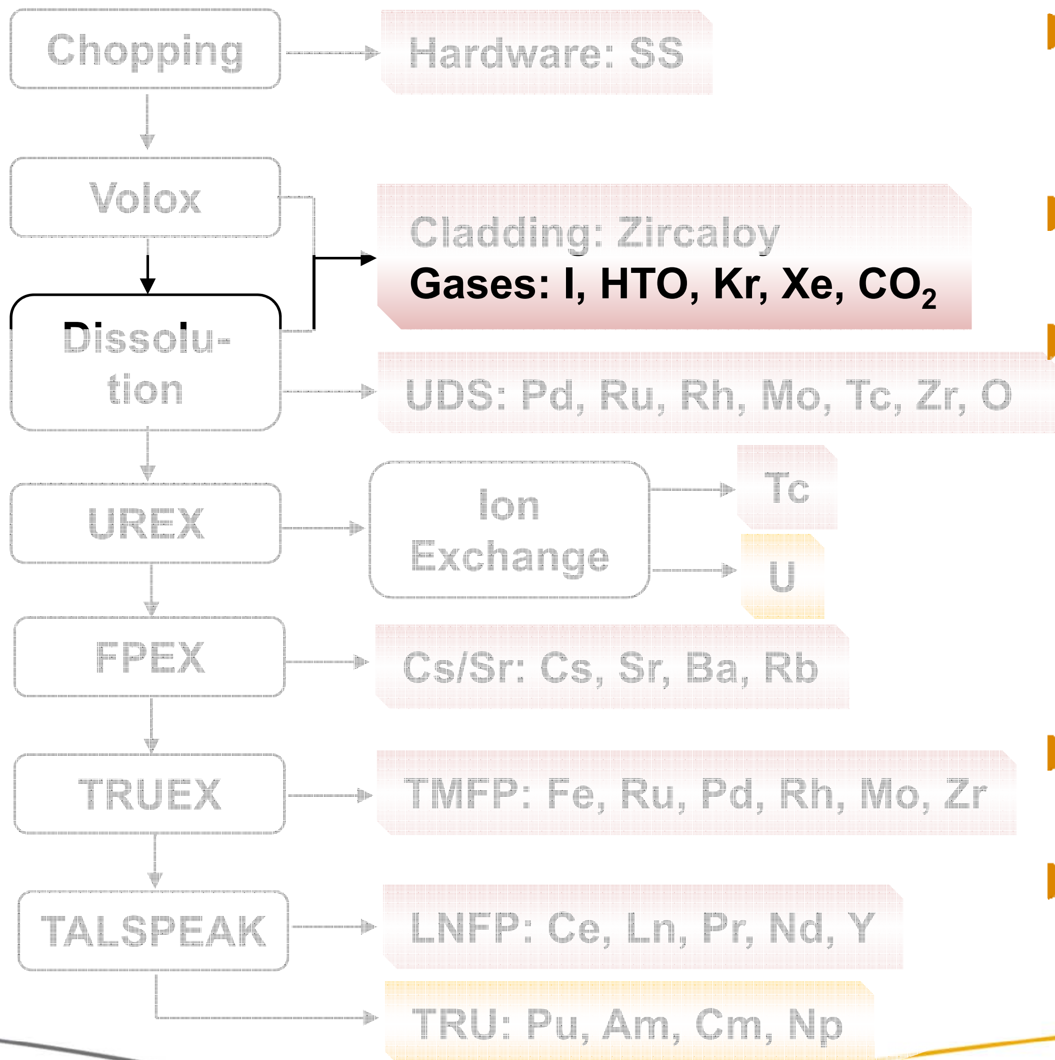
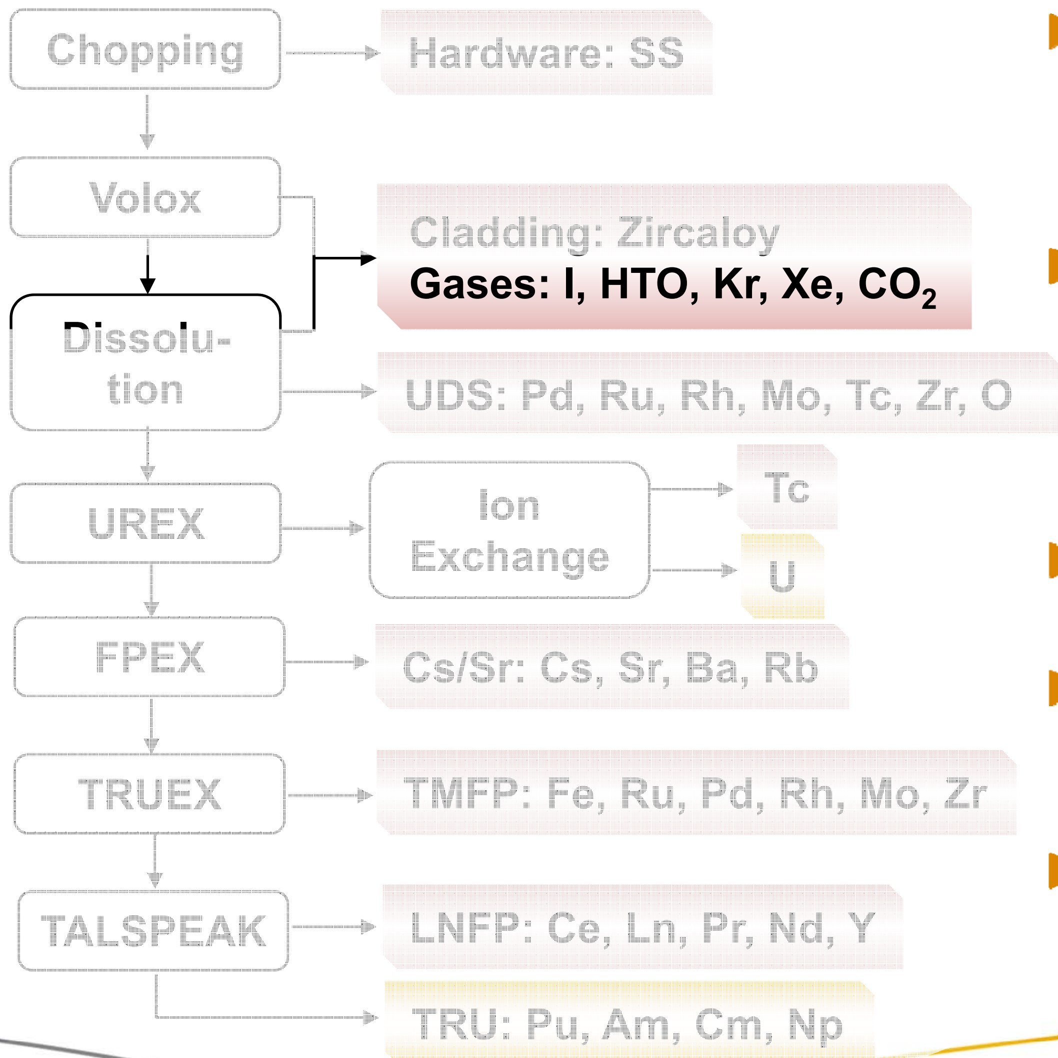
Potential Aqueous Process Waste Streams



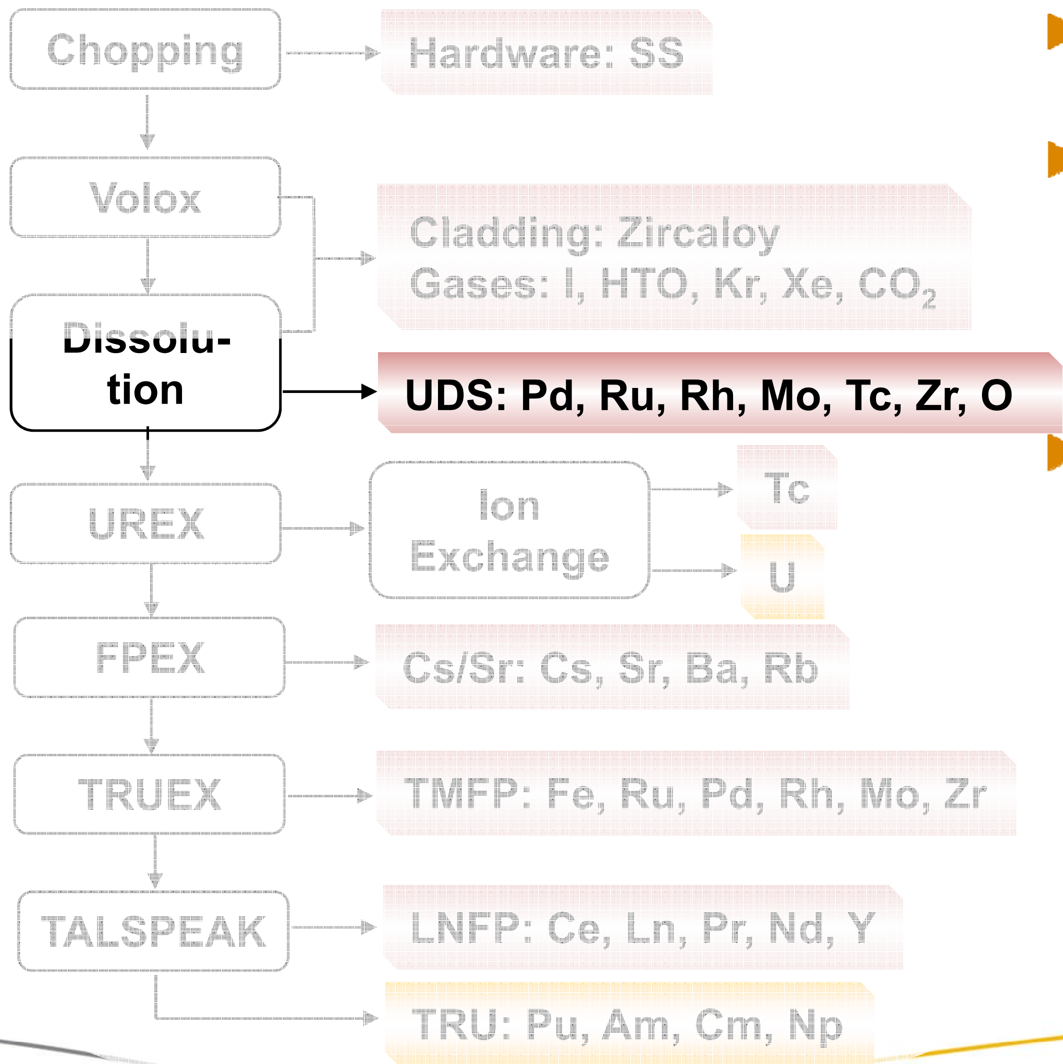
- ^{129}I is a long-lived mobile isotope requiring long-term immobilization
- Iodine capture is expected to be on silver mordenite (AgZ)
- Potential waste forms
  - encapsulated AgZ (low melting glass, silico-geopolymer, cement)
  - apatite-like minerals
  - low-melting glass
  - Bi-containing ceramics
  - others…
- Generally easy to capture, but, difficult to immobilize
- Combinations include salt waste from electrochemical processing

Potential Aqueous Process Waste Streams



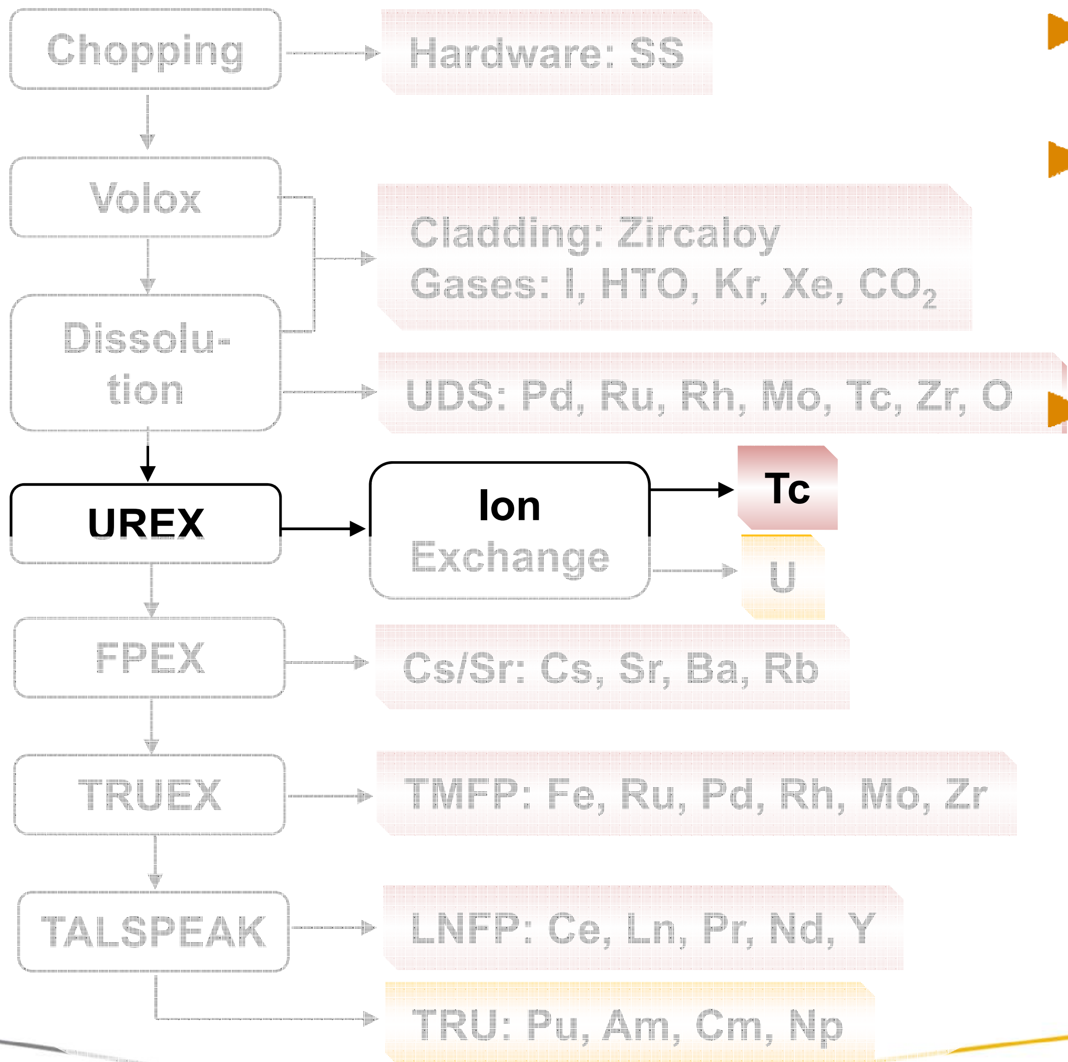
- Kr/Xe contains ^{85}Kr
  - require capture and storage if ≤30y cooled fuel is processed
  - Xe may be released if Kr removed
- Typical waste forms include:
  - compressed gas cylinders
  - ion implanted metal (e.g., Cu)
  - zeolites
- $^{14}CO_2$ may be released if separation specificity is high
- HTO must be captured and disposed of as LLW after decay storage
- Likely to combine HTO and $^{14}CO_2$ in cement waste form

Potential Aqueous Process Waste Streams



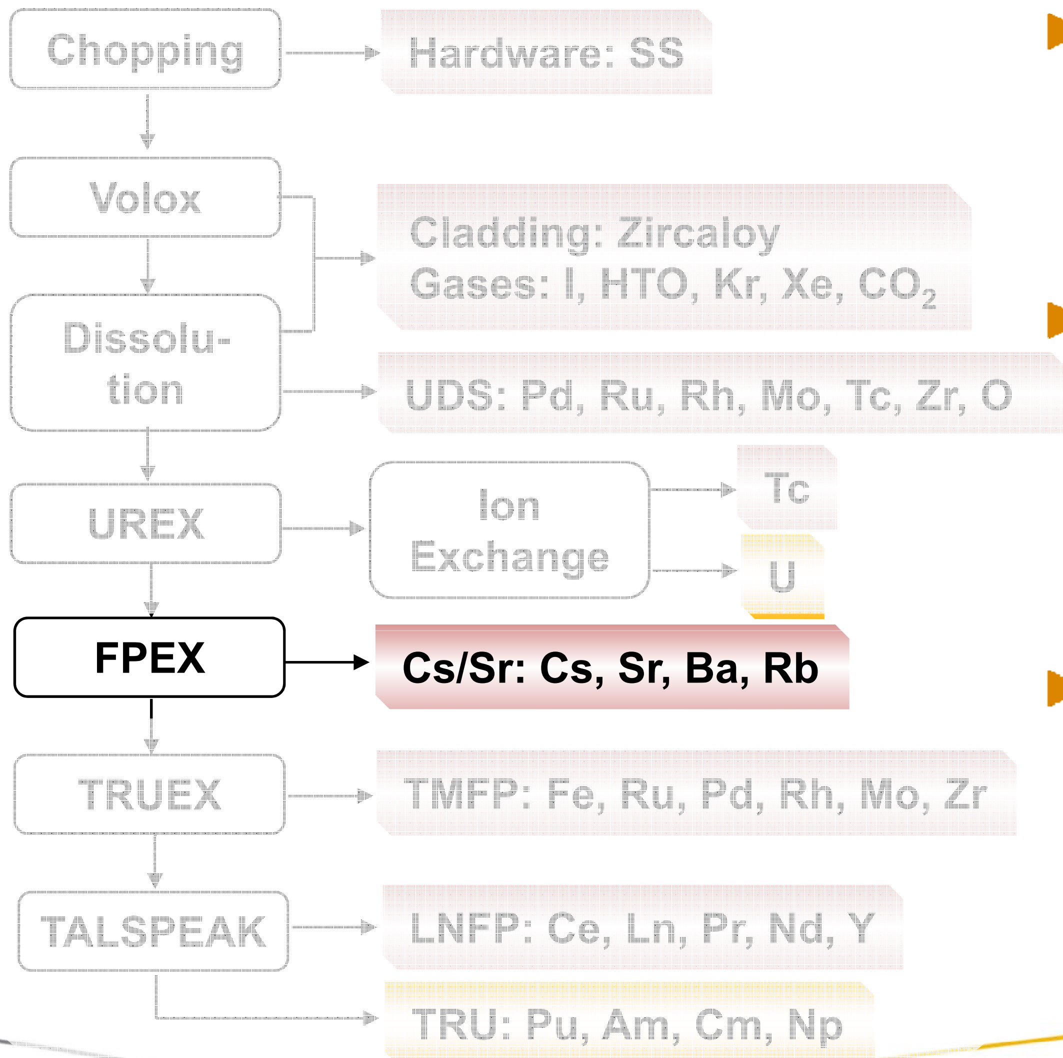
- UDS contains ^{99}Tc which is long -lived and mobile if oxidized
- Potential waste forms include
  - Fe or Zr rich alloy
  - glass with noble metal inclusions
  - pyrochlore - $[Tc,Zr,Mo,Ru,Rh,Pd]_2[LN,AN]_2O_7$
- UDS could logically be combined with:
  - Tc as both contain same primary radioisotope to immobilize
  - TMFP as both contain a large amount of noble metals, Zr, and Mo
  - or both

Potential Aqueous Process Waste Streams



- ^{99}Tc is long-lived and mobile if oxidized
- Potential waste forms include
  - Fe- or Zr-based alloy
  - glass
  - pyrochlore - $Tc_2[LN,AN]_2O_7$
- Tc could logically be combined with:
  - UDS as both contain same primary radioisotope to immobilize
  - or UDS + TMFP as both contain a large amount of noble metals, Zr, and Mo and need to immobilize Tc

Potential Aqueous Process Waste Streams



- Cs/Sr- high heat and dose from ^{134}Cs, ^{137}Cs, and ^{90}Sr decay
  - $Cs^{+} \rightarrow Ba^{2+}$ and $Sr^{2+} \rightarrow Zr^{4+}$
  - after ~300y only ^{135}Cs remains radioactive
- Potential waste forms include
  - titanates and niobates
  - aluminosilicates
  - glass (silicate or phosphate)
  - chlorides and fluorides (e.g.,WESF)
  - carbonates
- Cs/Sr likely combined with:
  - TMFP and/or LNFP in glass

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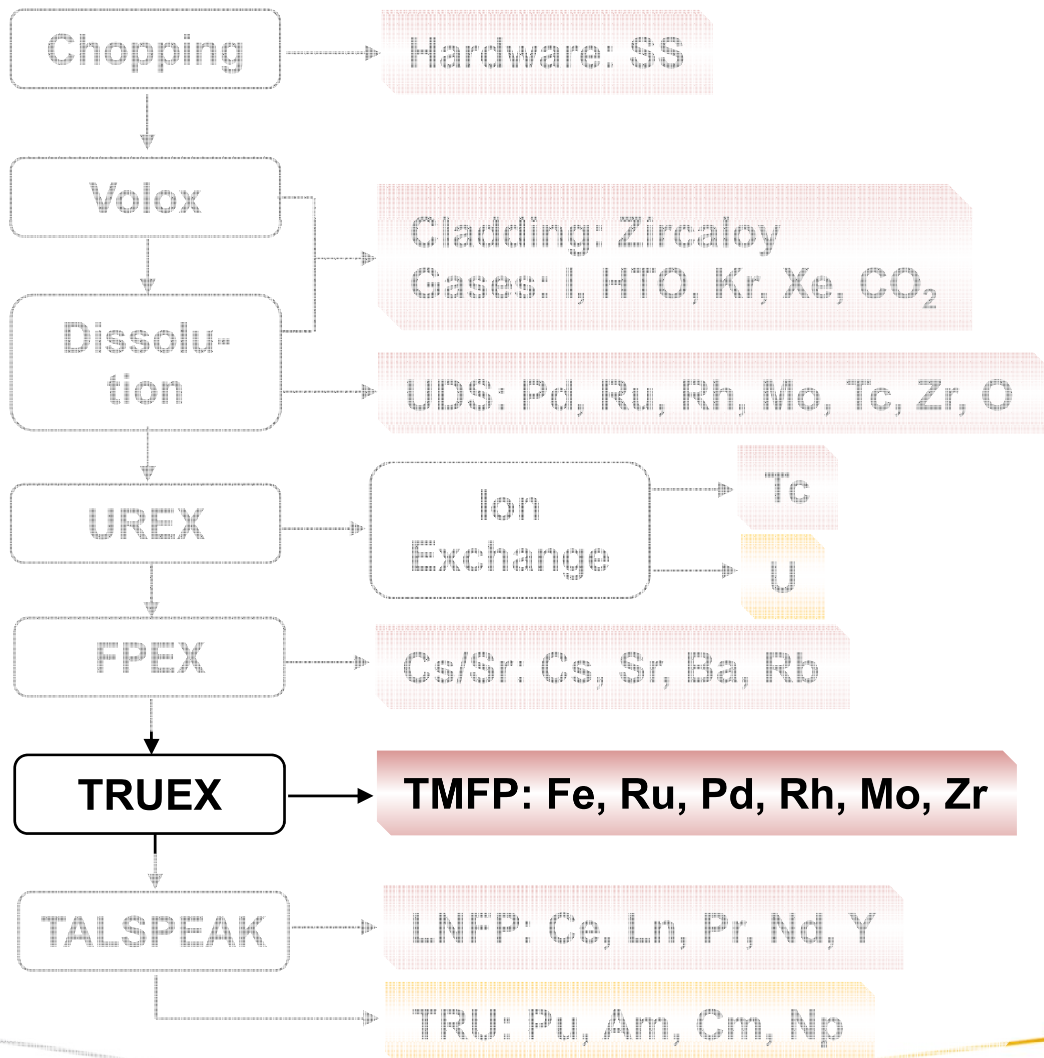
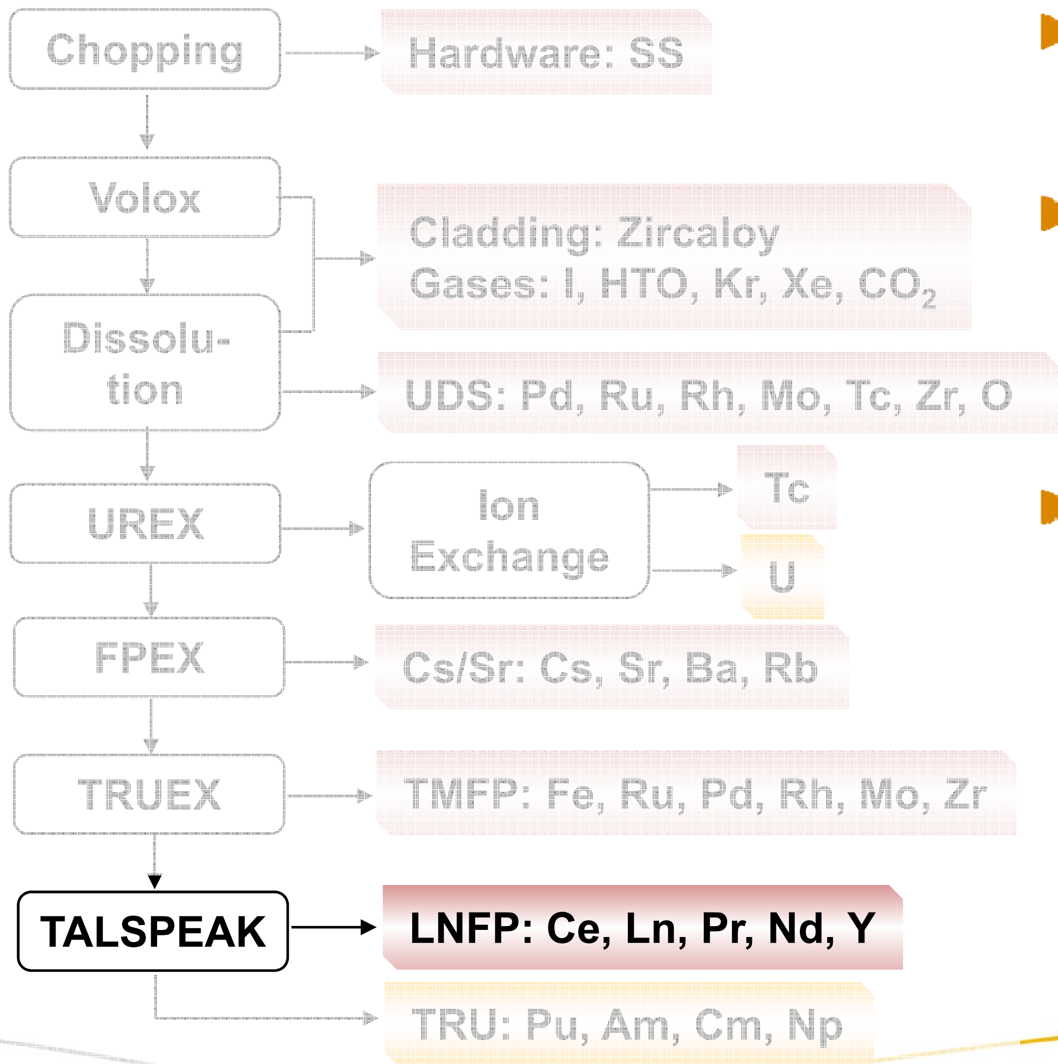
Potential Aqueous Process Waste Streams



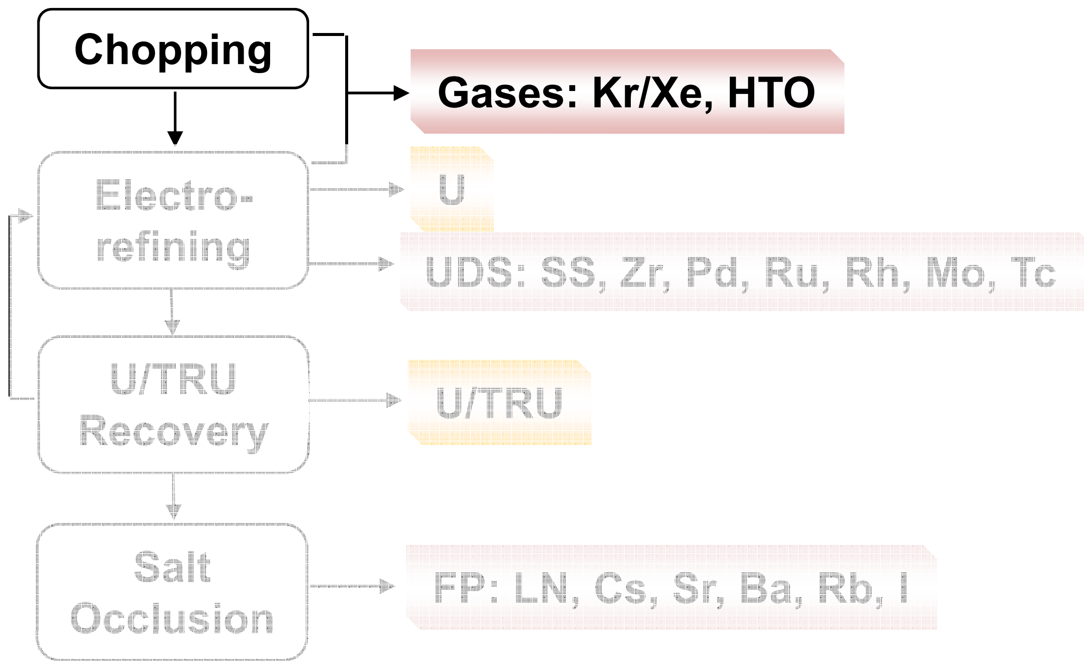
- TMFP (TRUEX raffinate) contains transition metals and many minor isotopes
- Potential waste forms include
  - Fe- or Zr-based alloy
  - glass with noble metal inclusions
  - pyrochlore - $[Tc,Zr,Mo,Ru,Rh,Pd]_2[LN,AN]_2O_7$
- TMFP could logically be combined with:
  - UDS as both contain a large amount of noble metals, Zr, and Mo
  - UDS + Tc if in metal waste form
  - LNFP and Cs/Sr if in glass waste form

Potential Aqueous Process Waste Streams



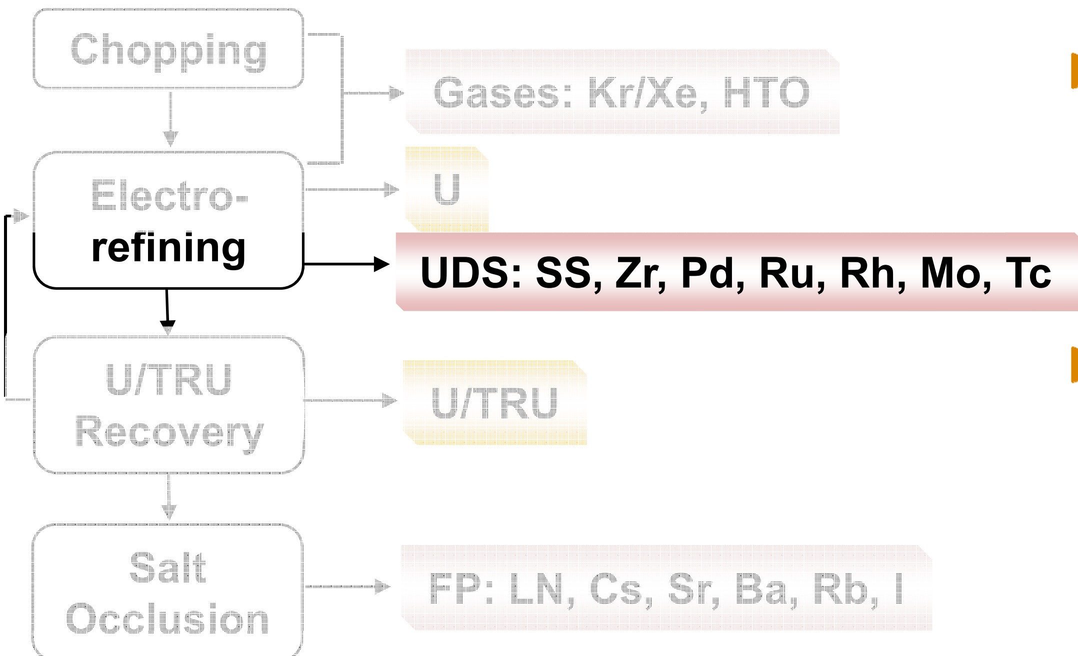
- LNFP (TALSPEAK product) contains lanthanides and trace transuranics
- Potential waste forms include
  - glass
  - pyrochlore - $[Tc,Zr,Mo,Ru,Rh,Pd]_2[LN,AN]_2O_7$
  - phosphate ceramics (e.g. monozite)
- TMFP could logically be combined with:
  - UDS, Tc, and/or TMFP if in pyrochlore
  - TMFP and/or Cs/Sr if in glass

Potential Electrochemical Process Waste Streams



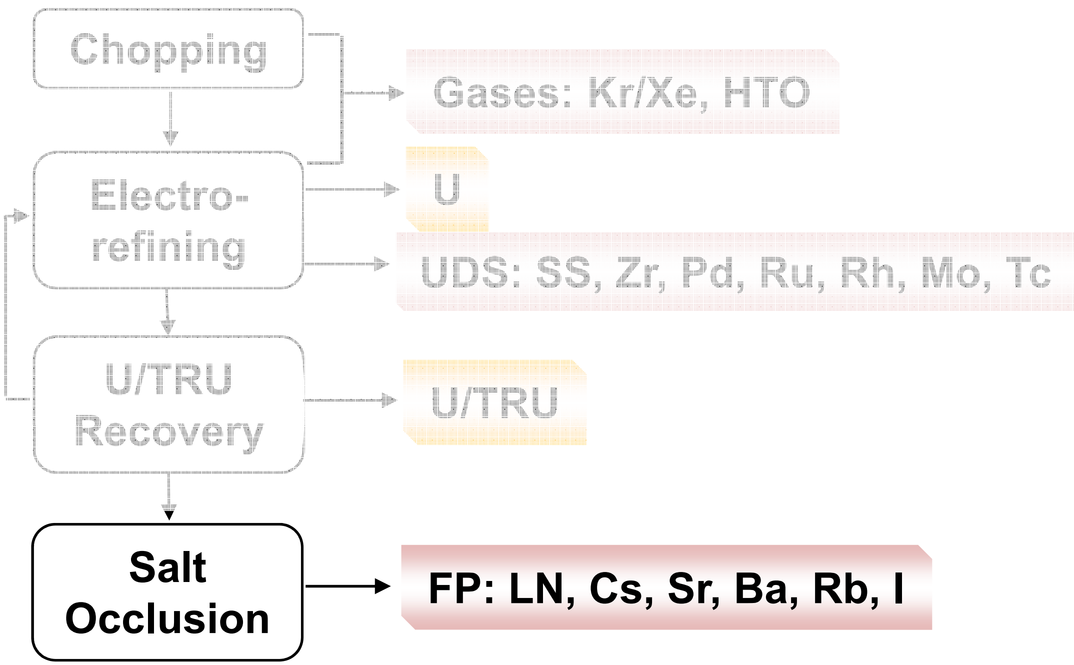
- Gasses
  - same treatment as aqueous
  - ^{3}H will partition to multiple streams

Potential Electrochemical Process Waste Streams



- UDS contains all metals more noble than uranium in chloride
  - recovered in a metallic form
- Potential waste forms include
  - Fe- or Zr-based alloy
  - glass with noble metal inclusions
  - pyrochlore - $[Tc,Zr,Mo,Ru,Rh,Pd]_2[LN,AN]_2O_7$
- UDS could logically be combined with
  - UDS, TMFP, Tc from aqueous if co-located facilities

Potential Electrochemical Process Waste Streams



- FP stream contains many fission products
  - in an alkali-chloride salt
  - likely to have TRU contamination
- Potential waste forms include
  - glass bonded sodalite
  - high chloride low-melting glass
  - other with additional separations
- FP could logically be combined with
  - iodine from aqueous processing
  - other waste from aqueous processing if additional separations

Potential Nuclear Waste Forms

- Glass
- Metal
- Ceramic

Potential Glass Streams

- Cs/Sr (high heat/dose)
- TMFP (noble metal concentrations)
- LNFP (high loading with only LNFP)
- Cs/Sr+LNFP
- Cs/Sr+LNFP+TMFP
- Cs/Sr+LNFP+TMFP+UDS +Tc (traditional HLW glass)

Glass Waste Forms

Structure

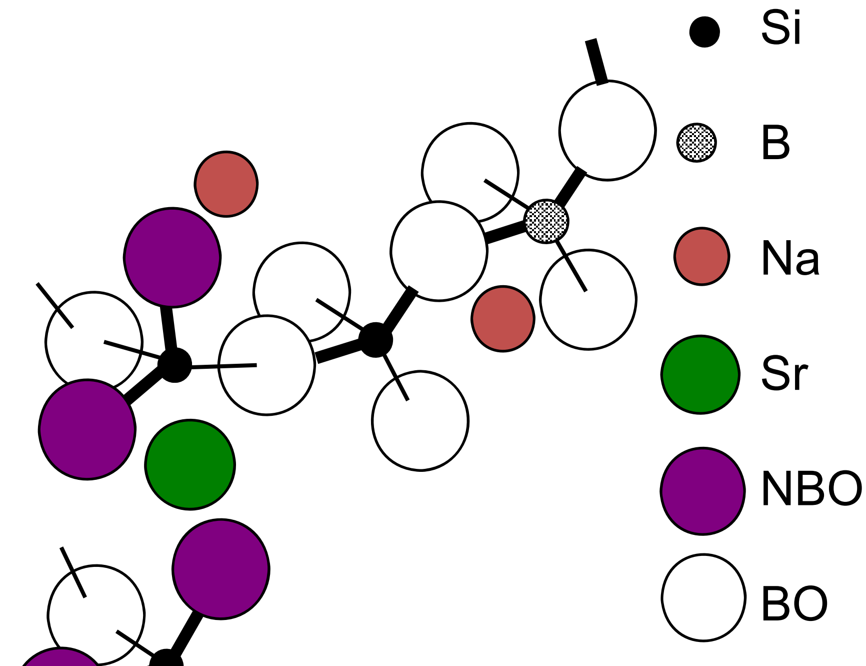
- Amorphous structure: more flexible than crystalline network
- SiO_4^{-4} tetrahedra form the continuous network
- B and Al are modified by waste elements to tetrahedral form
- Waste elements are integral part of glass structure… not simply contained or surrounded

Characteristics

- High flexibility to waste composition
- High speed continuous process

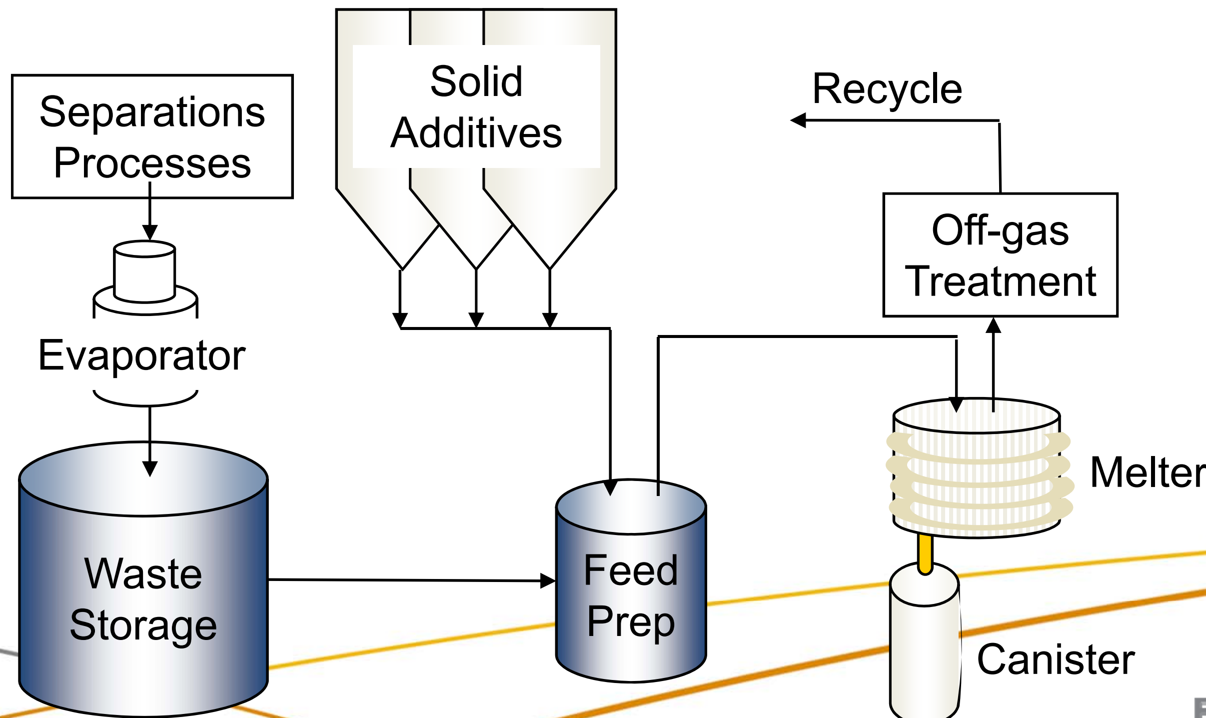
Primary options

- Alkali-borosilicate (ABS)
- Lanthanide-borosilicate (LaBS)
- Iron phosphate (FeP)

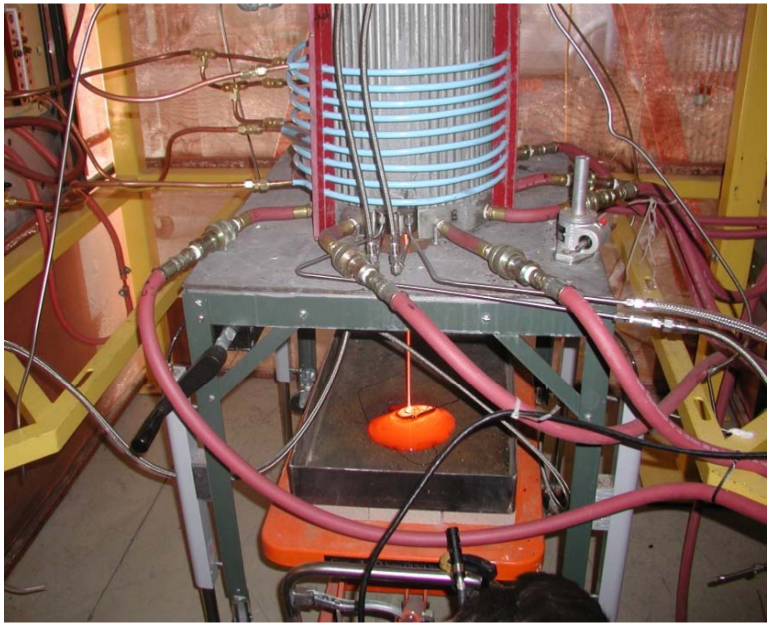


Vitrification Process

- Evaporate waste stream to heat, dose, solubility, etc. limits
- Blend with additives (e.g. aluminosilicates, reductant, …)
- Feed to melter (CCIM, HWIM, …)
- Cast into containers/allow to cool
- Seal, decontaminate or overpack, and store



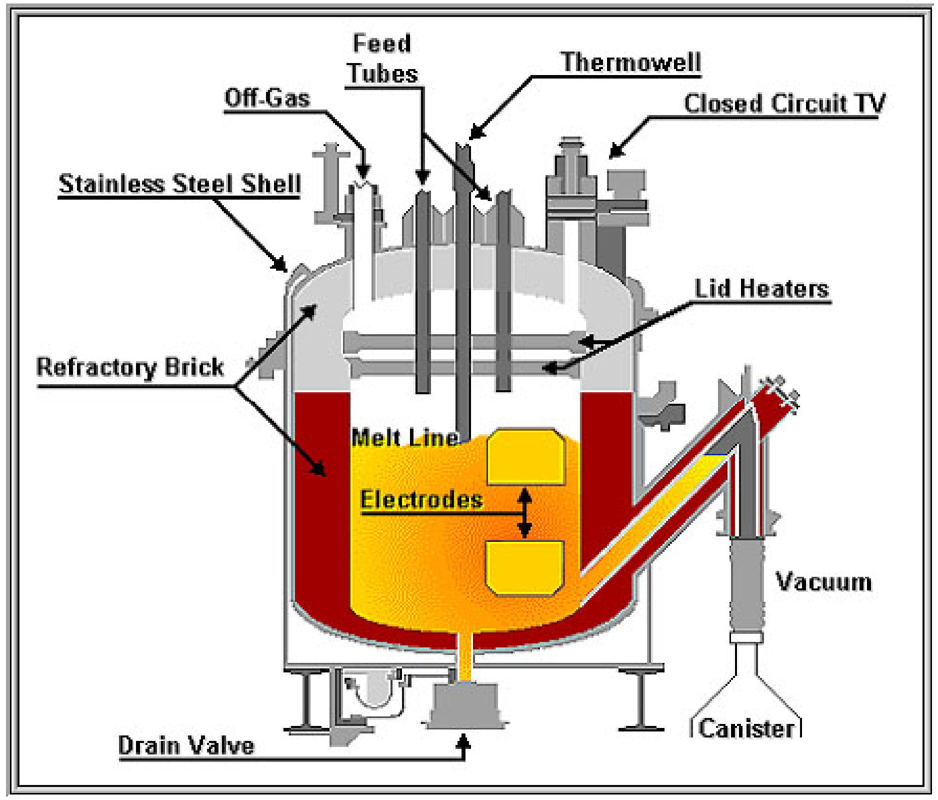
Melters



- A relatively small waste stream will be generated from a 800 MTIHM/y plant
  - from 30 MTG/y to a total of 130 MTG/y if all potential wastes are vitrified
  - translates to melter sizes of 300 mm to 800 mm diameter at 45 kg/(m^2·h)
- The CCIM and HWIM are thought to best meet this mission due to:
  - small in-cell size, high specific melting rate, tolerance to solid inclusions, ability to fully empty
- HWIM is better able to melt glasses with low alkali content (e.g., LNFP) since the induction couples to the crucible rather than the glass
- CCIM is better able to melt glasses with noble metals (e.g., TMFP or UDS) since the glass "scull" protects the melter

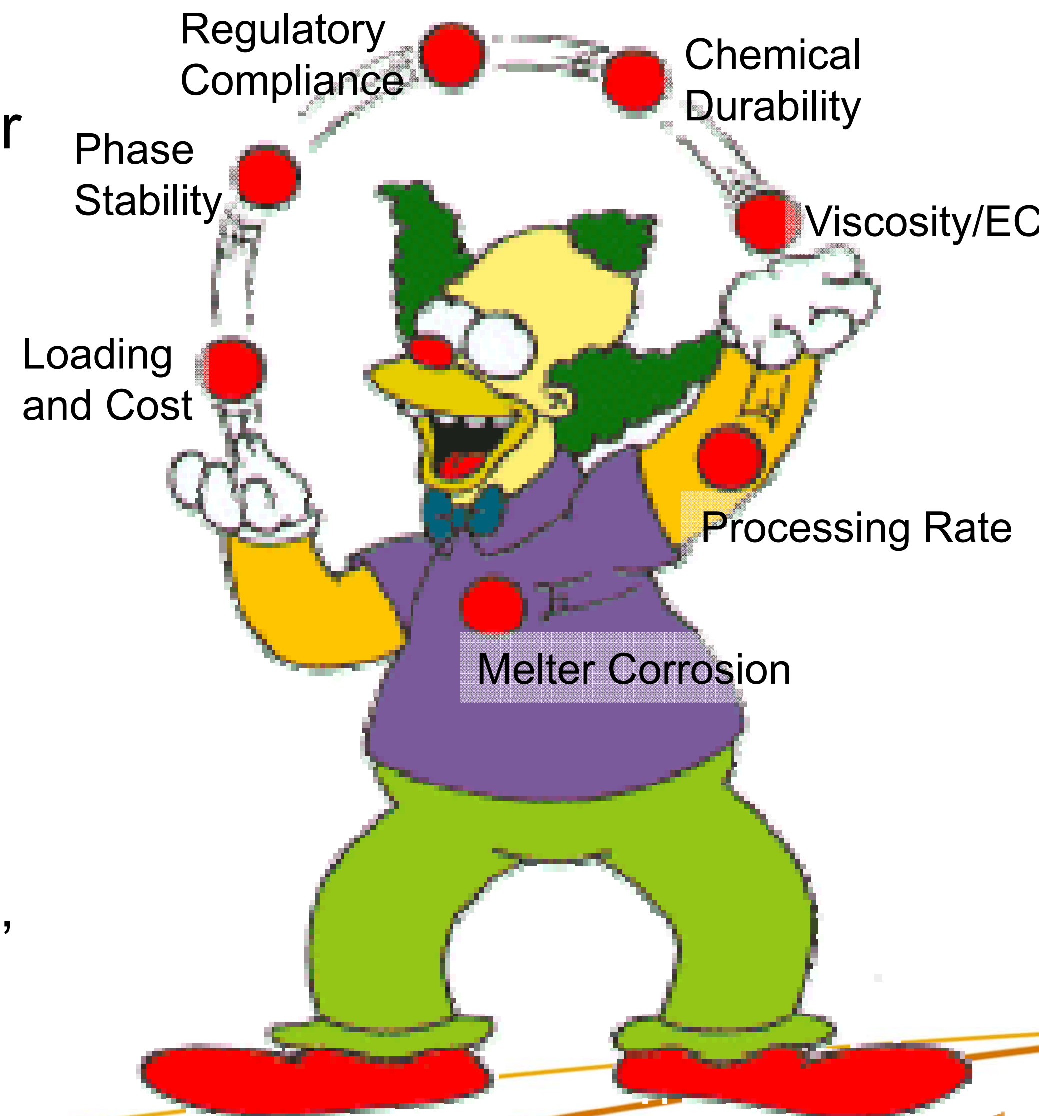
Melters – Joule-Heated, Ceramic-Lined

- Developed in the U.S. for vitrification of defense HLW
- Advantages of JHCM
  - large size capability (heat is deposited to volume rather than surface)
  - well demonstrated at WVDP, DWPF
  - relatively high design life
- Disadvantages
  - large size
  - temperature limits
- In short, well suited to tank waste, but, not for small scale new recycling plant



Glass Formulation

- Glass must meet a number of constraints:
  - product quality → chemical durability, thermal conductivity, radiation resistance, regulatory constraints, transition temperature, phase stability etc.
  - processability → melting temperature, crystal formation, inclusions, conductivity
  - economics → waste loading, processing rate, process TOE



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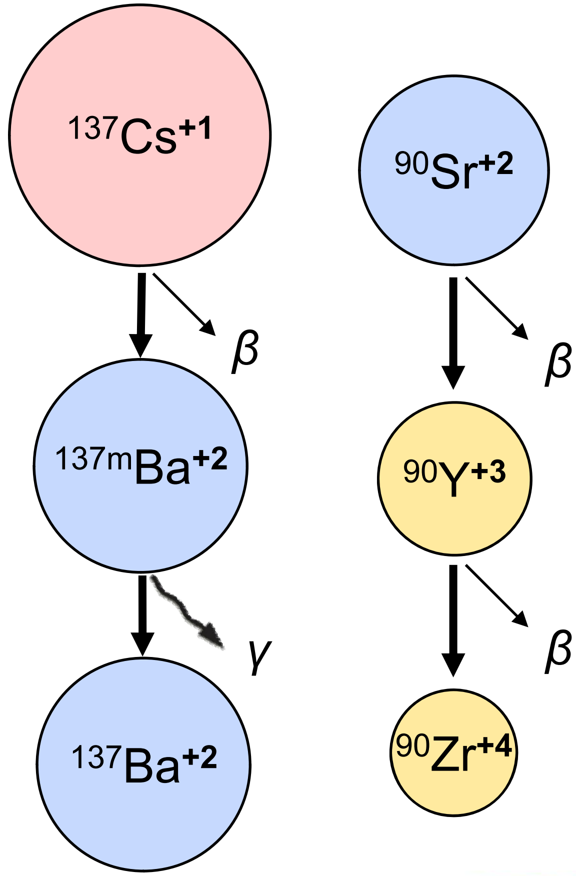
Key Formulation Considerations for Advanced Closed Fuel Cycle

- Temperature
  - glass with high Cs/Sr, requires high T_g to ensure that the glass stays as a solid with self-heating
  - this tends to require high melting temp. (limiting melter technology)
  - high thermal conductivity is a plus
- Radiation and decay tolerance
  - radiation generates high β-γ dose
  - decay changes chemistry
    $Cs^{+} \rightarrow Ba^{2+}$ and $Sr^{2+} \rightarrow Zr^{4+}$
  - high mobility and multivalent oxides
- Volatility (primarily Cs and halides)



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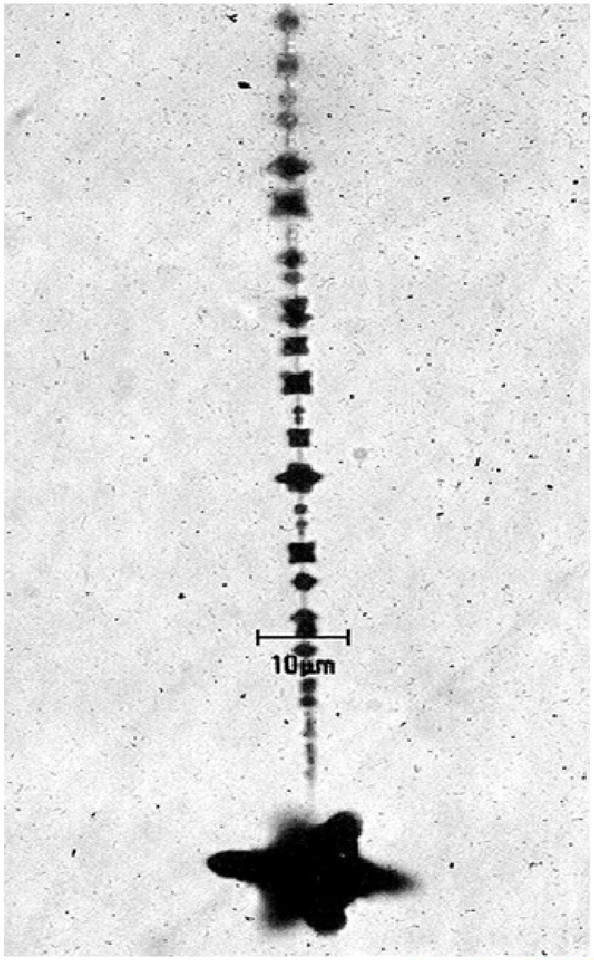
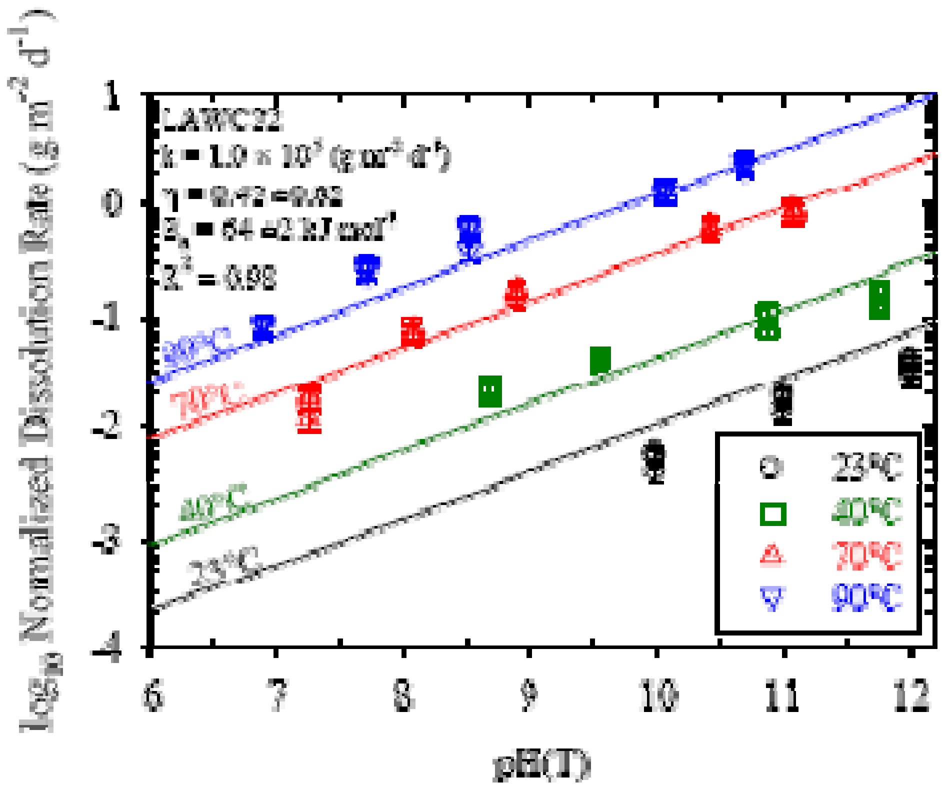
Key Formulation Considerations for Advanced Closed Fuel Cycle

- Solids
  - noble metals (Pd, Rh, Ru) are insoluble in most oxide glasses
  - need a melter technology that will tolerate solid inclusions
  - waste loadings may be set to maintain NM concentration below melter tolerance limit
- Waste solubility
  - many waste components are sparsely soluble in glasses
    - Mo, Cr, S in ABS, higher in FEP
    - AN, LN in FEP, higher in ABS, higher still in LaBS
- Chemical durability



Glass Corrosion Rate

- Waste is incorporated in the glass – that is bound on a molecular scale within the solid
- During reaction with water, the release of most waste components from glass is determined primarily by the rate of glass corrosion
- Need to couple experiments and modeling to estimate release



$$r = k_0 \cdot pH^{\eta} \cdot \exp\left[\frac{-E_a}{RT}\right]\left(1-\left[\frac{Q}{K_g}\right]\right)^{\sigma}$$

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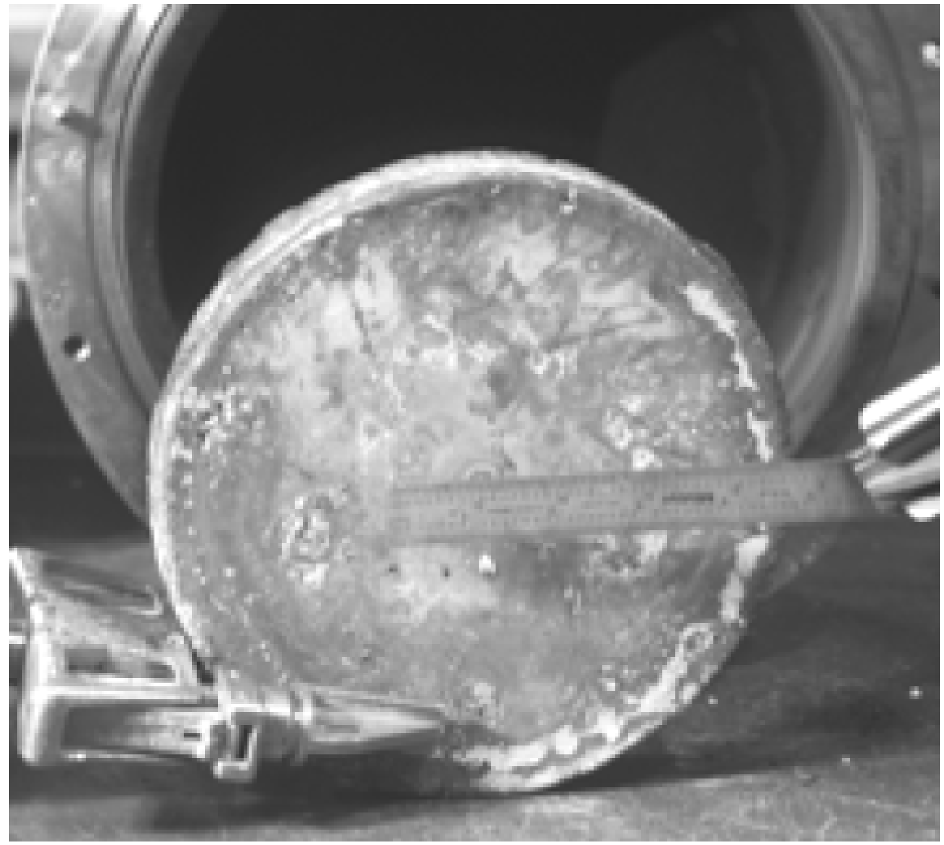
Pros and Cons of Glass as a Waste Form

Pros	Cons
Mature technology	Thermal process (difficult to permit)
Flexible to composition and process variations	Low tolerance to some components (Noble metals, S, Mo, etc.)
Well understood properties including chemical durability	Lower durability than many ceramic phases (e.g., zirconates and titanates)
Continuous process with no respirable fines	Low temperature limit to withstand radiolytic heat ($400 < T_g < 750°C$)
High tolerance to radiation and transmutation	Volatility of Cs requires recycle
Qualified for repository disposal	
High waste loading (low disposal volume)	
Typically single phase waste form	

Potential Metal Waste Streams

- UDS (contains glass-insoluble noble metals & Mo and Tc)
- Tc (aq) (best immobilized in reduced form)
- TMFP (aq) (contains glass-insoluble noble metals & Mo)
- UDS+Tc (aq)
- UDS+Tc+TMFP (aq)



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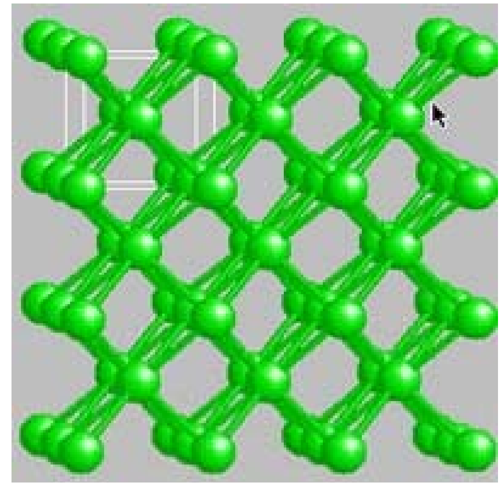
Metal Waste Forms



Structure

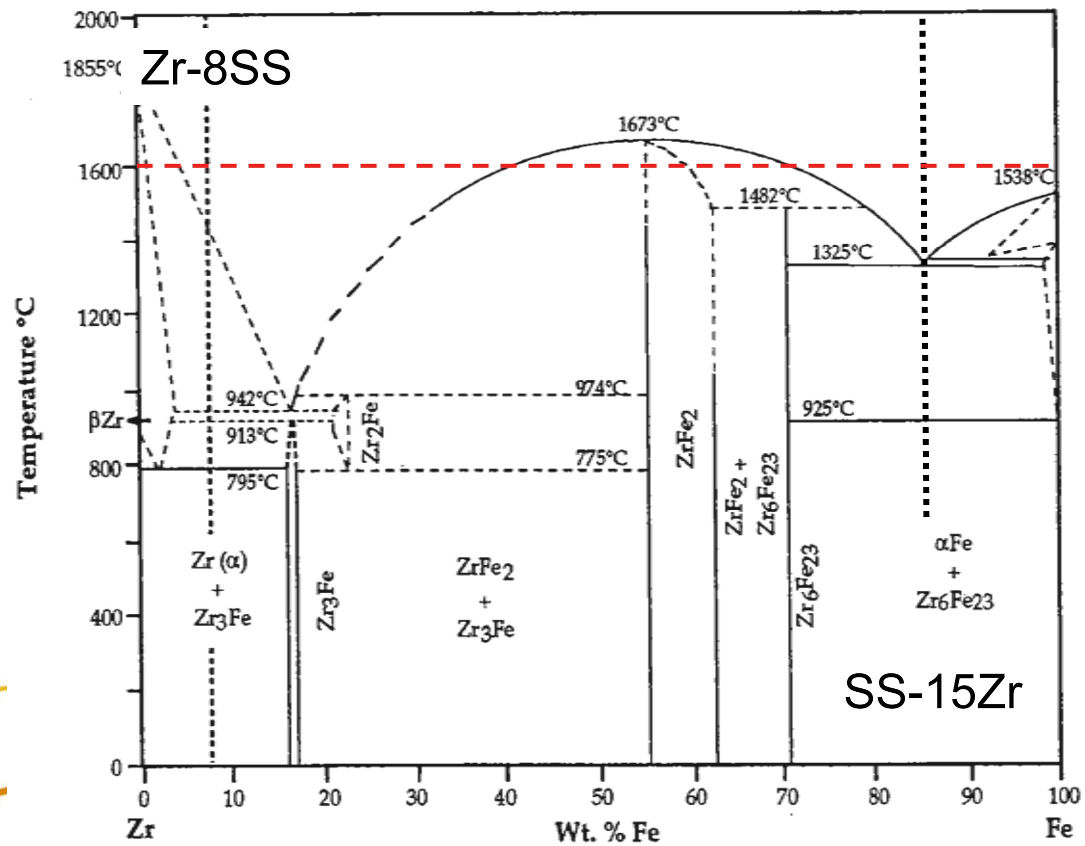
- Crystalline metals
- fcc, bcc, hcp, etc.

Characteristics

- Reduced waste form
- High density
- High thermal conductivity

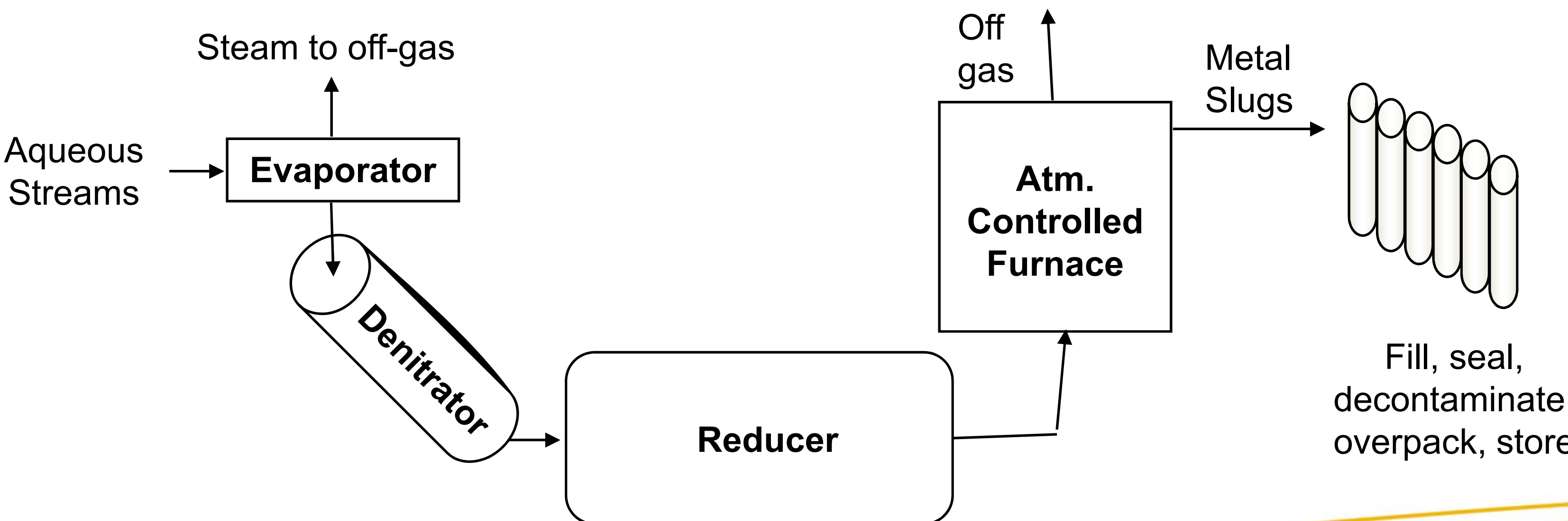
Primary options

- Zr
- Fe
- intermetallics



Metal Process

- Evaporation, calcination, and reduction to form metal
- Combine in crucible with coke and other metal streams
- Melt in crucible, move to canister in “slugs”
- Seal, decontaminate or overpack, and store



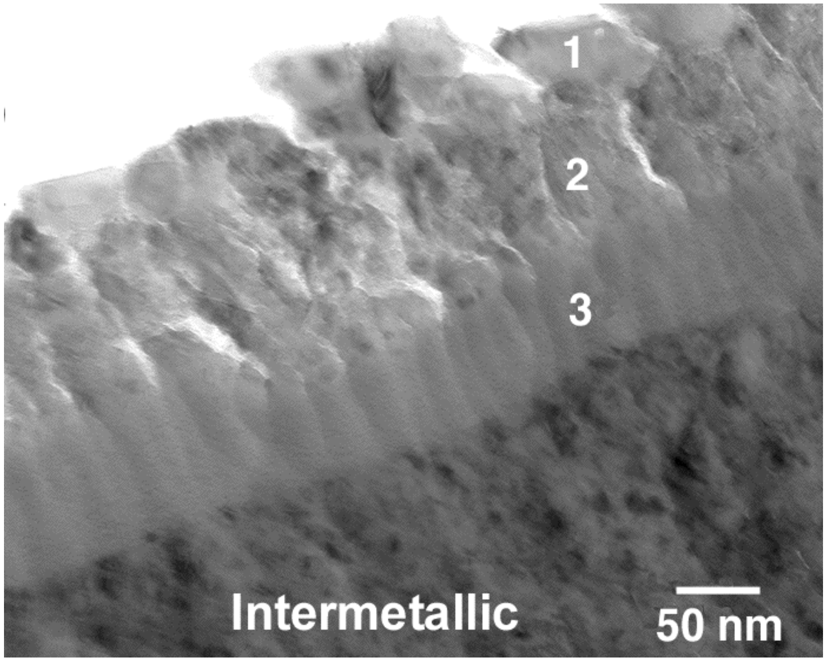
Metal Formulation

- Similar process to glass formulation
- Processing (temperature, microstructure development, slag formation, etc.)
- Product (phases formed, radionuclide partitioning, slag properties, corrosion rate)

	Maximum Solute Concentration, atomic%									
	Fe	Cr	Mn	Ni	Mo	Pd	Rh	Ru	Tc	Zr
γ-Fe	—	11.9	100	100	1.7	100	3	23	30	0.7
α-Fe	—	100	3	5.5	24	6.5	19	4	0	0.05
Fe_2M	—				33.3					66-73
FeM	—	45-50	0-100	0-100	43-57	0-100	0-100		15-66	
ZrM_2	66-73	64-69	60-80		60-67				No	—
ZrM								50	No	—
Zr_2M	32			33		66	33		No	—

Metal Corrosion Rate

- Metal corrodes by an oxidative process
  - electrochemical measurements are used to measure corrosion behavior
  - incongruent corrosion has been found for waste alloys (both Fe- and Zr-based alloys)
  - determining the phase preference of radionuclide is important
  - passivation layers may form and slow reaction
  - hydrogen embrittlement, SSC, and pitting are also key processes



$$\boxed{r = \frac{a_{max} \times \ln\left(1 + \frac{b \times a_{max}}{a_{max}} T_e\right)}{T_e}}$$

$$\ln a_{max} = 7.98 + (2.39 \times 10^{-4} \times [Cl^-]) - 1.23 \times pH$$

$$\ln (b \times a_{max}) = -0.10 + (0.015 + 5.82 \times 10^{-6} \times [Cl^-]) \times T - 0.698 \times pH$$

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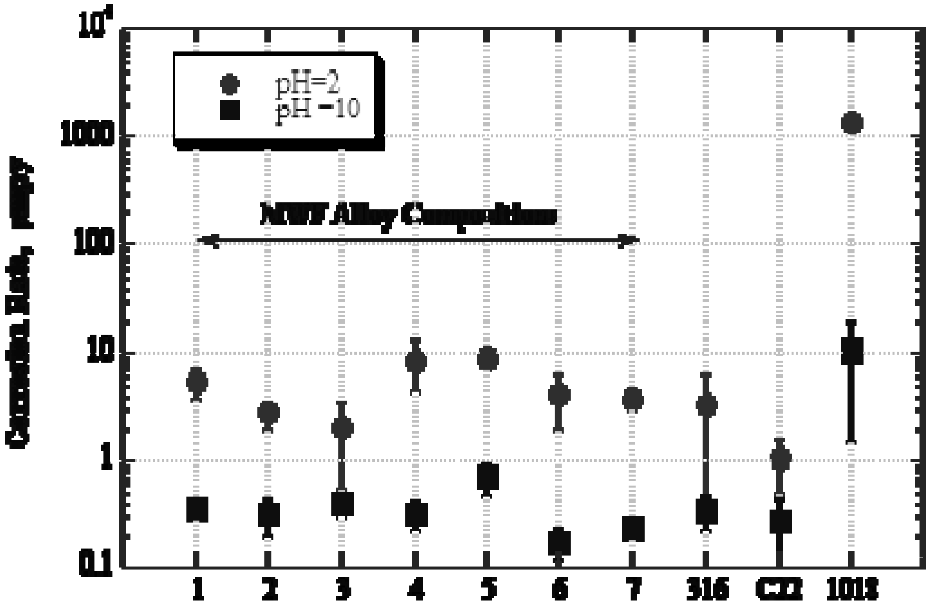
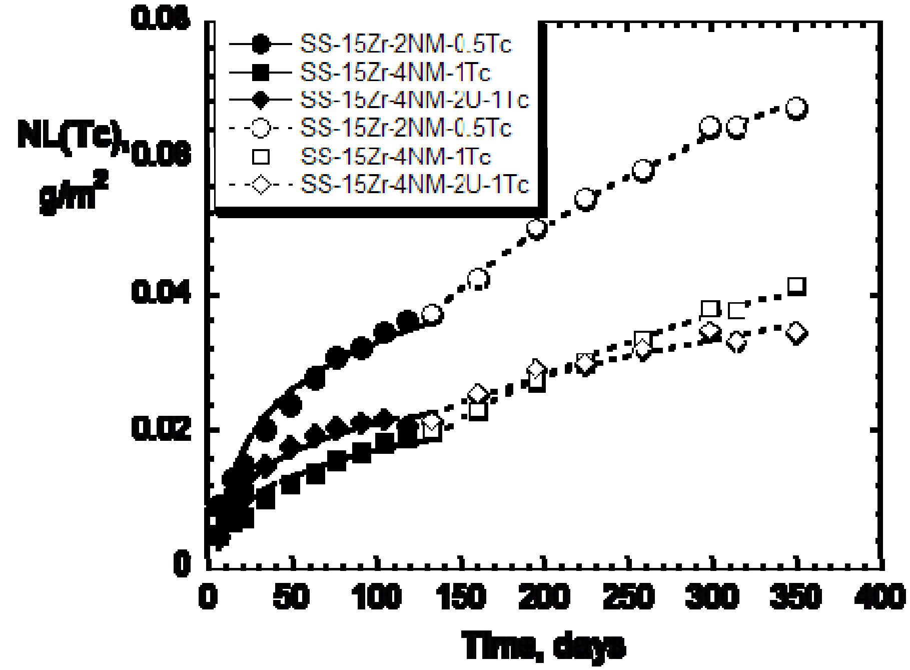
Metal Waste Form Corrosion





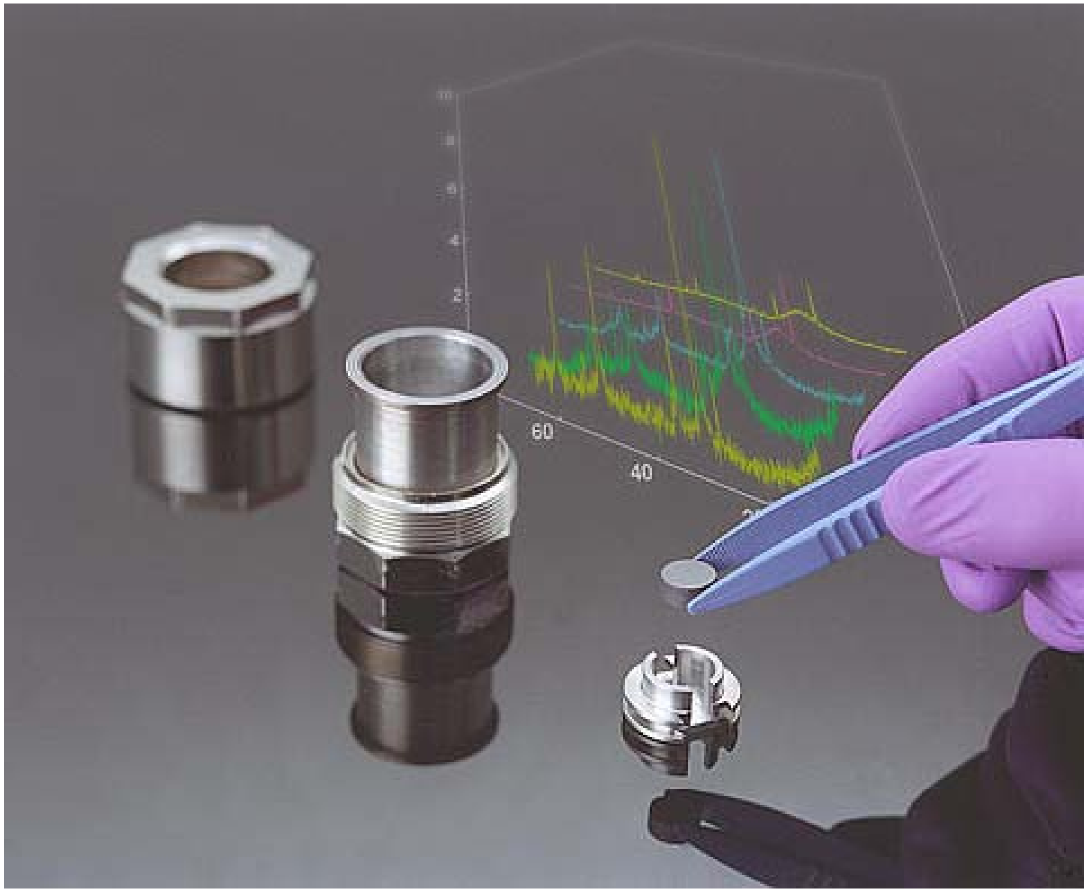
1: SS-15Zr-5U-2Tc
2: SS-15Zr-1Nb-1Pd-1Rh-1Ru-1Tc
3: SS-20Zr-1Nb-1Pd-1Rh-1Ru -1Tc
4: SS-5Zr-1Nb-1Pd-1Rh-1Ru -1Tc
5: SS-15Zr-0.6Ru-0.1Pd-11U-0.3Tc
6: SS-15Zr
7: SS-15Zr-1Nb-1Pd-1Rh-1Ru

Pros and Cons of Metal as a Waste Form

Pros	Cons
Somewhat flexible to composition and process variations	Thermal process (difficult to permit)
High tolerance to radiation and transmutation	Requires reduction process when applied to Tc and TMFP
High waste loading (low disposal volume)	Lower durability than many ceramic phases (e.g., zirconates and titanates)
Maintains reducing environment, limiting Tc releases	Durability and processability not well understood
High thermal conductivity, allowing possibility of high storage temperatures	Multiphase waste form
	Handling of metallic slugs required
	Non-continuous (batch) process

Potential Ceramic Streams

- Iodine
- Cs/Sr
- TMFP
- LNFP
- LNFP+TMFP
- LNFP+TMFP +UDS+Tc



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Ceramic Waste Forms

Structure

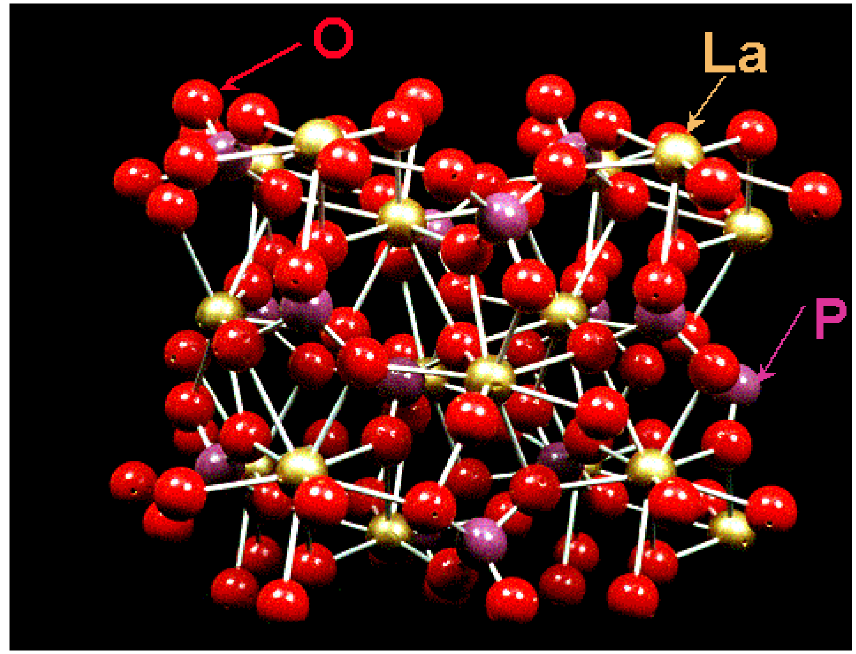
- Thermodynamically stable crystalline oxides
- Regular network with long-range order

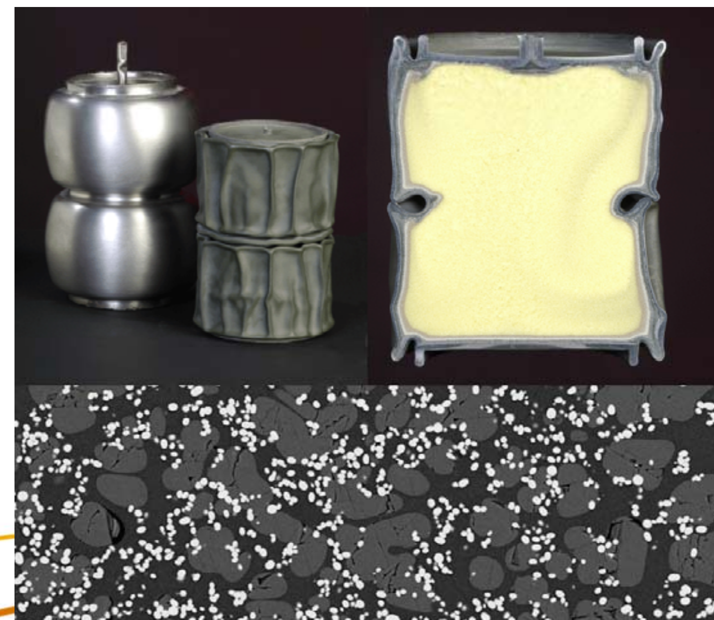
Characteristics

- Very high durability
- High thermal stability

Primary options

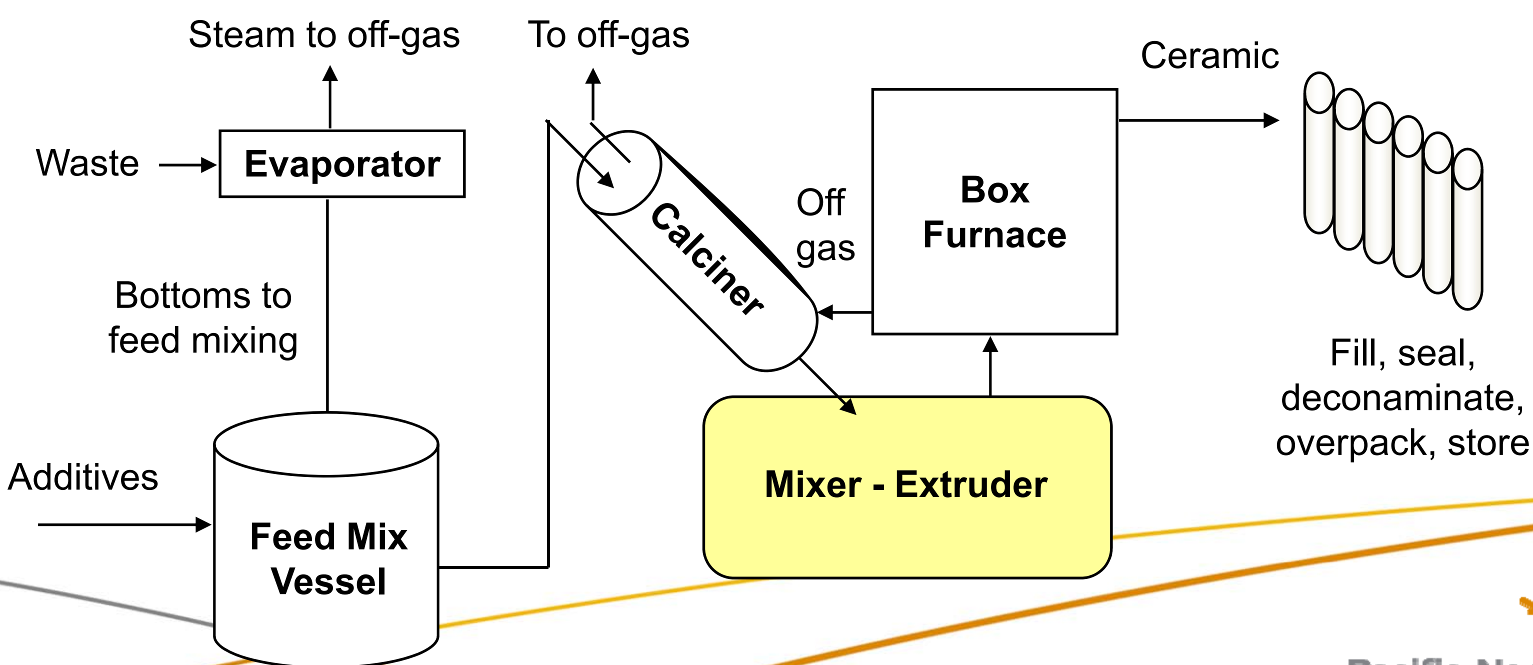
- Alumino-silicates
- Titanates
- Zirconates
- Phosphates





Example Ceramic Process

- Evaporate waste stream to heat, dose, solubility, etc. limits
- Blend with additives
- Calcine mixture to remove water and organics or nitrates
- Form green ceramic (press, extrude, etc.)
- Ramp heat in box furnace (dry, react, sinter, and slow cool)
- Load into canisters, seal, decontaminate or overpack, and store

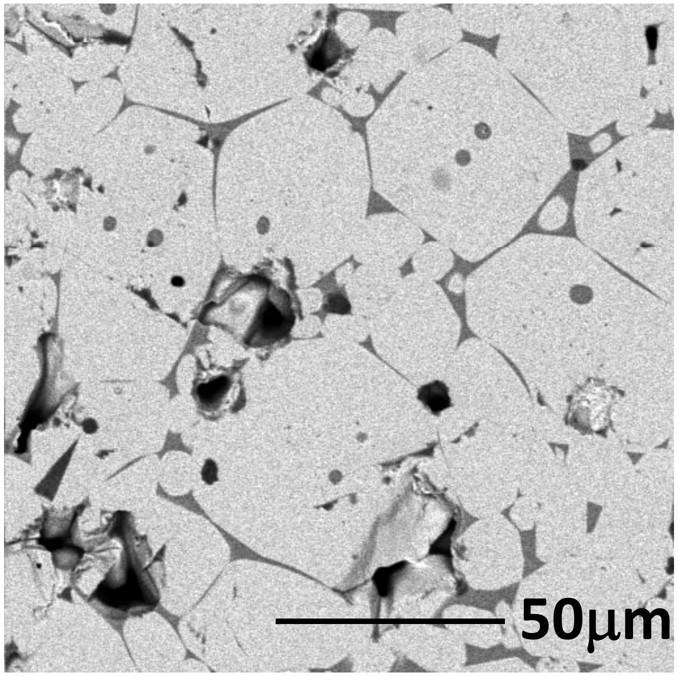


Ceramic Process Alternatives

- There is not a single ceramic process, but, many process steps that can be combined for an optimal total process
- Head end
  - absorption/adsorption
  - precipitation
  - calcine
  - sol-gel
- Forming
  - filter press
  - cold press
  - extrusion
  - casting
- Heating
  - furnace (tunnel or box)
  - hot isostatic press (HIP)
  - hot uniaxial press (HUP)

Ceramic Formulation

- Just as their isn't a single ceramic process, there isn't a single *ceramic*
- Typically, a target phase or phases are selected and additives are optimized to adjust processability and product quality
  - pyrochlore: $[Ru,Pd,Zr,Tc,Rh]_2[LN,AN]_2O_7$
  - zircon: $[Zr,AN,Th]SiO_4$
  - zirconolite: $[Ca,Ba,Sr][Zr,AN]Ti_2O_7$
  - monozite: $[LN,AN]PO_4$
  - pollucite: $[Cs,Rb][Al,Fe]Si_2O_6$
  - celcian: $[Ba,Sr][Al,Fe]_2Si_2O_8$



- Processing (phase formation, process temperature, densification rate, ripening or grain growth, shrinkage, etc.)
- Product quality (phase formation, grain boundary composition, microstructure, radiation damage, chemical durability)

Pros and Cons of Ceramic as a Waste Form

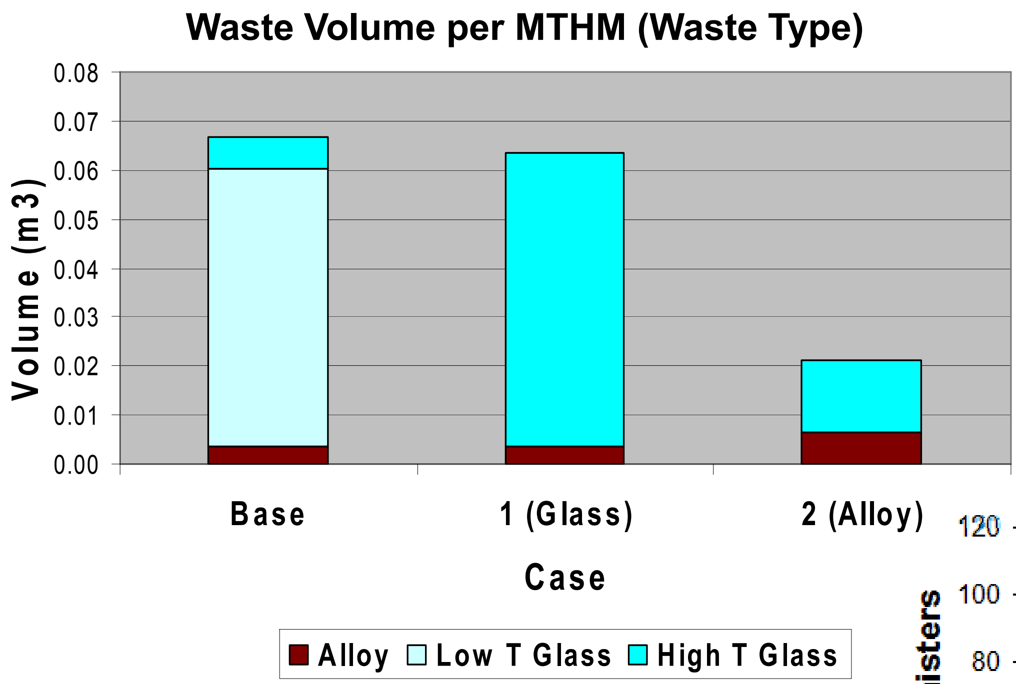
Pros	Cons
Very durable waste forms	Thermal process (difficult to permit)
Thermodynamically stable in disposal environment	Expensive relative to glass
High thermal stability	Potentially generates respirable fines
	Multiphase waste form
	Handling of ceramic parts required
	Non-continuous (batch) process

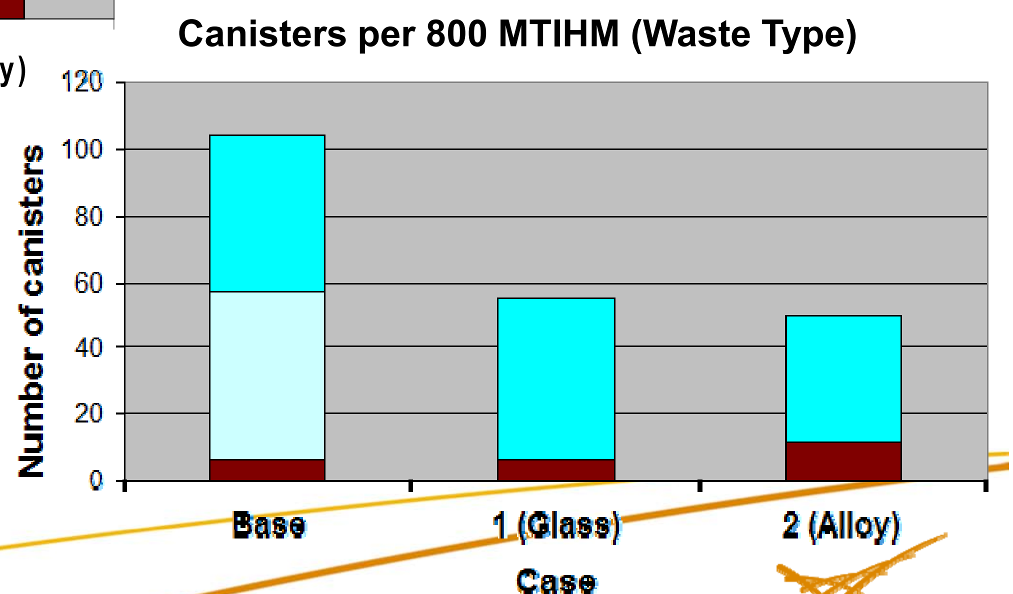
Example Cost Analysis

- Three options considered for immobilizing five wastes
  - base-case uses five waste forms for the five primary aqueous waste streams
  - options 1 and 2 reduce to two waste forms
  - option 1 removes the need for Cs/Sr separation and has roughly the same waste volume as the base
  - option 2 requires TMFP reduction and has the lowest waste form volume

<table>
<tr><th>Case</th><th>UDS</th><th>Tc</th><th>TMFP</th><th>LNFP</th><th>Cs/Sr</th></tr>
<tr><td>Base</td><td>Fe-alloy</td><td>Zr-alloy</td><td>ABS glass</td><td>LaBS glass</td><td>ABS glass</td></tr>
<tr><td>Opt 1</td><td colspan="2">Fe-alloy</td><td colspan="3">ABS glass</td></tr>
<tr><td>Opt 2</td><td colspan="3">Fe-Alloy</td><td colspan="2">LaBS glass</td></tr>
</table>

Option Comparison: HLW Volume and Canisters





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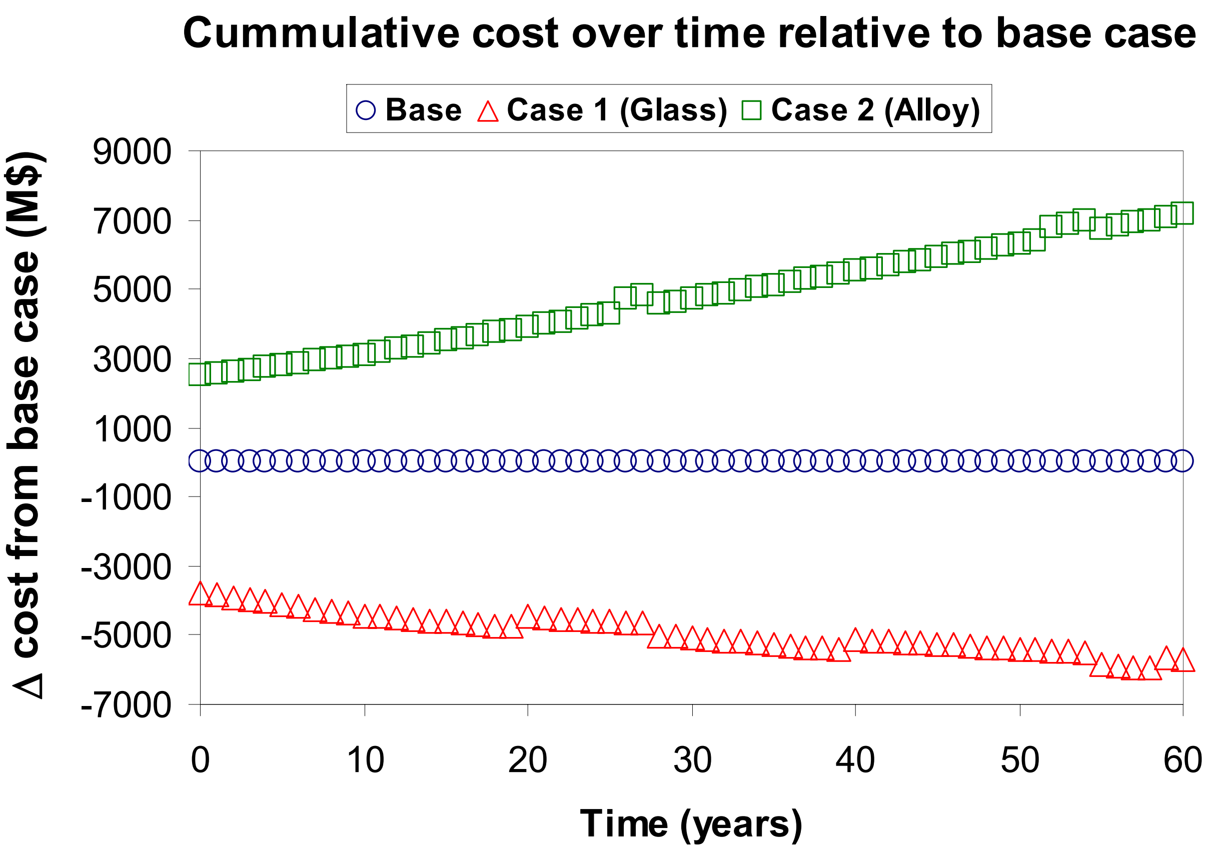
Trade Study Results

- Combining TMFP, Cs/Sr, and LNFP into a single glass waste form is the most cost effective option
  - many sensitivities evaluated
  - lines vary, but, order doesn't change
  - capital and operating costs of FPEX and TMFP reduction out weigh waste volume costs
- Only costs evaluated not other benefits



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Concluding Remarks

- There will be (most likely) an opportunity to rethink the waste management strategy for a U.S. closed fuel cycle
  - advanced separations flowsheets will allow for an unprecedented level of control over waste management
  - waste forms can be tailored to match waste chemistry and disposal environment
- Several options are available for each potential waste stream
  - need further development on each option (material and process)
  - selection depends on cost benefit analysis… cost is easy to estimate
- This presentation was aimed at giving a flavor for some of the waste stream and waste form option
  - many other separations flowsheets were not discussed
  - there are many other waste form options not discussed in detail (e.g., cement, glass-ceramic, composites, etc.)

Acknowledgements

- I greatly appreciate you attention and interest
- Many thanks go the U.S. Department of Energy (DOE) Offices of Environmental Management and Nuclear Energy for their generous support of waste form and waste process development at PNNL and elsewhere
- I'm honored to have been invited by the organizers to present this material
- Some figures where borrowed from: ANL, ANSTO, Areva, Geosafe, INL, Rockwell, The Simpsons, SRNL, WTP, and www.webminerals.com -- some with permission
- Pacific Northwest National Laboratory is operated by Battelle for the DOE under Contract DE-ACO5-76RL01830

Backup Slides

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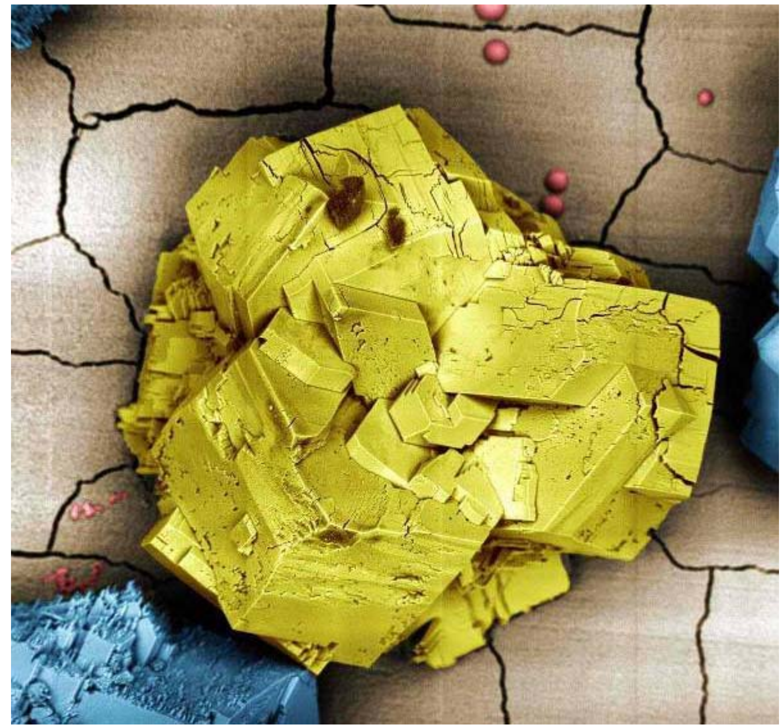
AFCI Waste Form Campaign Mission

- Develop and demonstrate durable waste forms and processes to enable safe and cost-effective waste management as an integral part of a closed nuclear fuel cycle by establishing a fundamental understanding of behavior through closely-coupled theory, experiment, and modeling.



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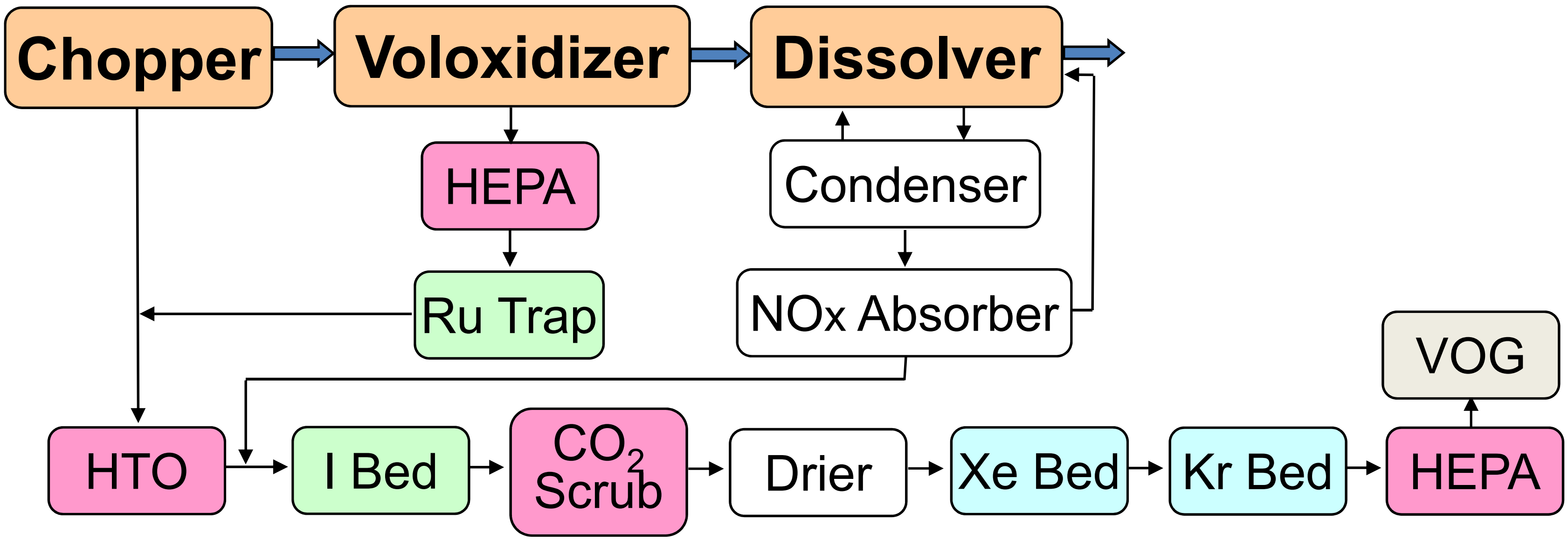
Potential Aqueous Primary Off-gas Streams



HTO
Catalytic oxidizer
MS (3A) capture
Release from MS
Condense
Mix in cement
In-can grouting
Dispose - SLB

I Bed
Ag-Z capture
Remove bed
Encapsulate
Package
Dispose - Rep

CO_2 Scrub
NaOH scrub
Remove
Mix in cement
In-can grouting
Dispose - SLB

Xe Bed
Cryogenic Ag-Z
Release from Ag-Z
Combine w/ Kr or Release

Kr Bed
Cryogenic HZ
Release from HZ
Combine w/ Xe
Pressurize
Bottle
Decay storage

HEPA
Remove
Leach
Dispose - SLB

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Alkali and Alkaline Earths

- FPEX strip solution contains Cs, Sr, Ba, and Rb
- Heat and dose from ^{134}Cs, ^{137}Cs, and ^{90}Sr decay
  - fraction of isotopes depends on fuel age
  - $Cs^{+} \rightarrow Ba^{2+}$ and $Sr^{2+} \rightarrow Zr^{4+}$
  - after ~300y only ^{135}Cs remains radioactive
- Potential waste forms

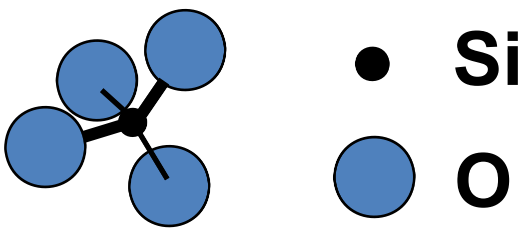
Form	Titanate/ Niobate	Alumino-silicate	Glass	Fluoride/ Chloride	Carbonate
durability	very high	high	high	low	low
temp resistance	very high	high	med	low	low
dose resistance	very high	unknown	high	high	unknown
decay tolerance	unknown	unknown	high	unknown	unknown
cost	very high	high	med	low	low

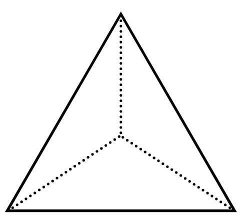
What is Glass?

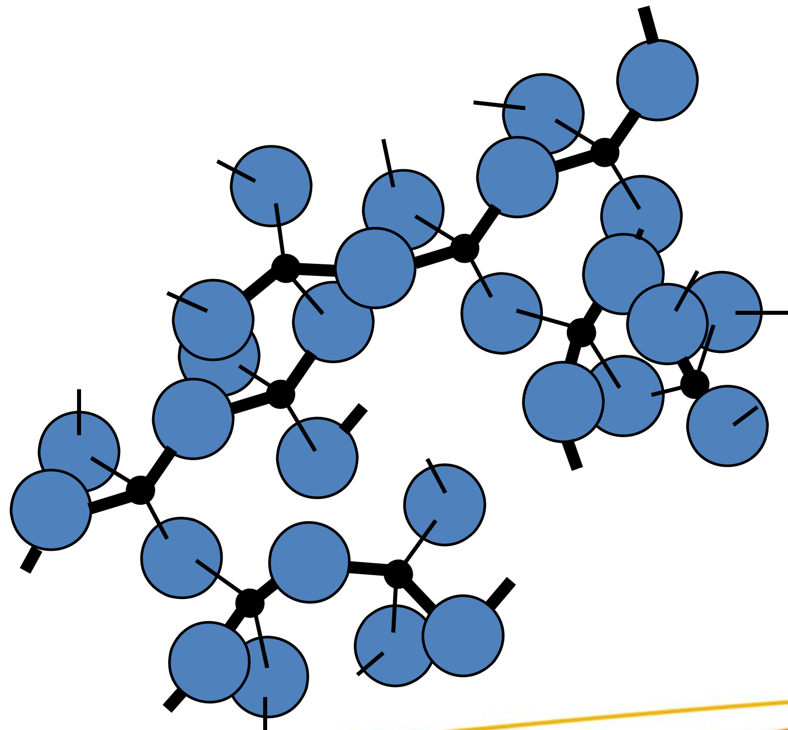
- Comparison between amorphous and crystalline materials
  - amorphous materials are built up from a random arrangement of a structural/chemical motif, whereas crystalline analogues are built up from an ordered arrangement of motif
  - as in crystalline materials, where order exists at short- and long-range, amorphous materials also exhibit a considerable degree of short-range order (i.e., the variation in the structural/chemical motif can be relatively small)

What is Glass?

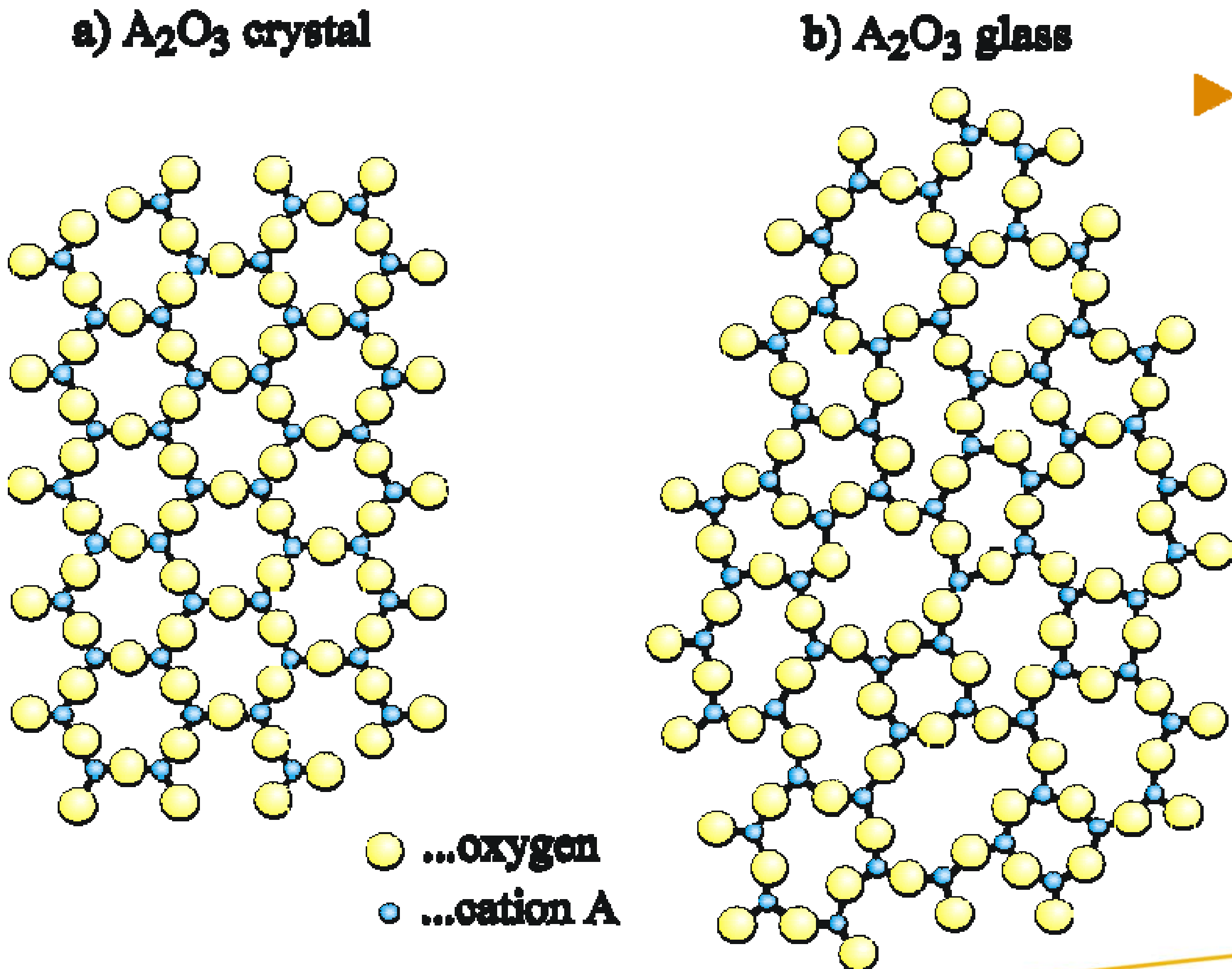
- Structural and chemical aspects
  - For silica, SiO_2, the structural and chemical building block or motif is the tetrahedrally coordinated orthosilicate monomer, SiO_4^{-4}
  - Amorphous silica is built up from a random arrangement of orthosilicate motif







What is Glass?



- Comparison between amorphous and crystalline materials
  - crystalline materials can be conceptually constructed by performing symmetry operations on a motif

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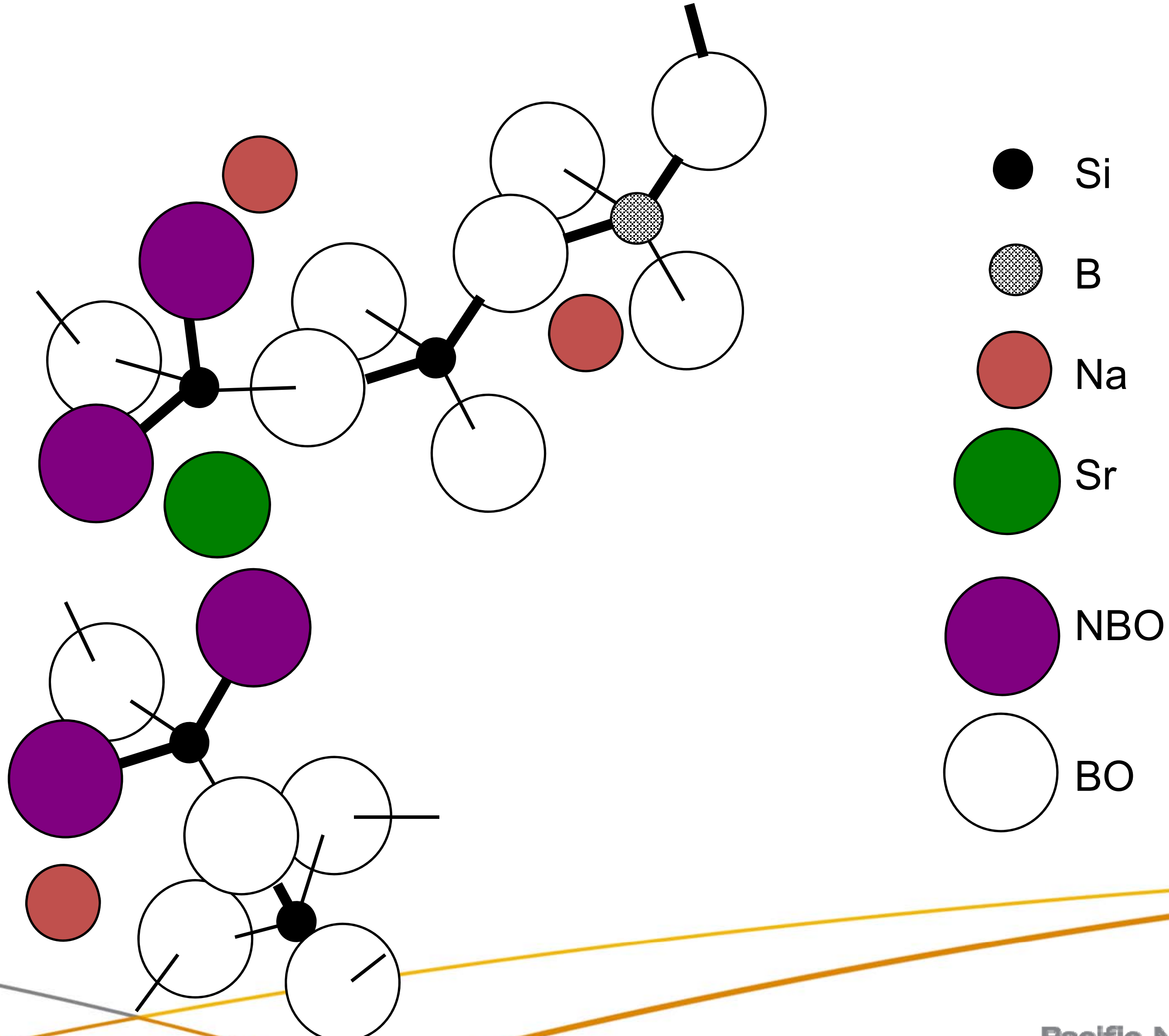
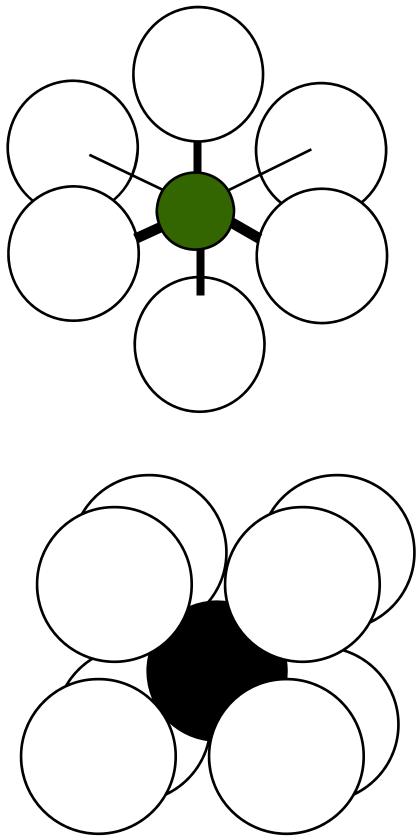
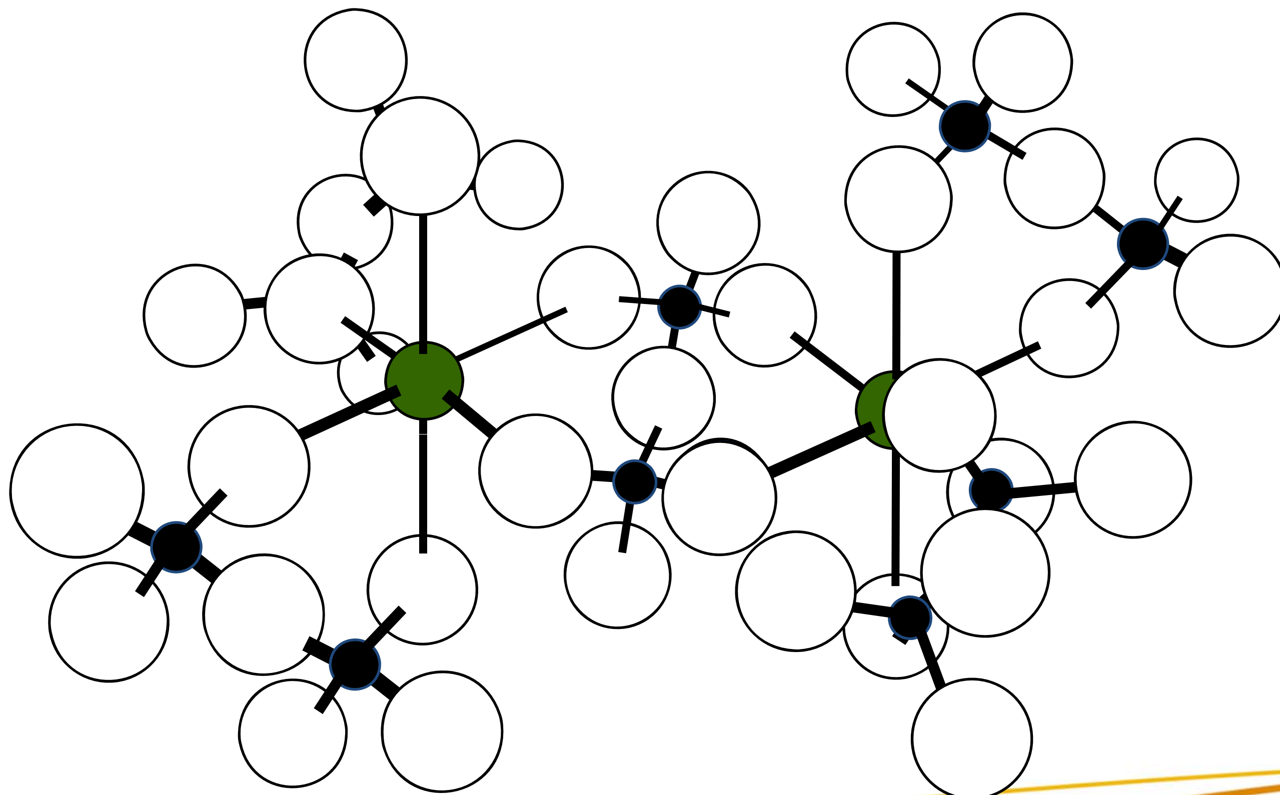
Borosilicate Waste Glasses



Six and Eight Fold Coordinated Cations





Compacted Metals

- Hulls
  - <1% fuel after voloxidation and filter
  - leaching removes all measurable residual fuel meat
  - likely to be >100nCi TRU/g due to activation of U impurities in Zircaloy → GTCC
- Hull/hardware compaction
  - proven technology employed worldwide
  - potential for encapsulation in metal matrix

Zircaloy and SS Scrap from Cogema Plant, LaHauge
from: Management of Cladding Hulls and Fuel Hardware,Tech report 258, IAEA, Vienna, 1985

Technology Readiness Assessment (TRA)

- Evaluating a technology (waste process/form) for readiness to implement/produce
- Concept from NASA, adopted by DOD, now being adopted by DOE
- Generates technology readiness level (TRL, 1-9) and technology maturation plan (TMP)

Stage	**Technology Development** →				**Demo** →		**Comm** →	**Operations**	
Scale	**Concept** →	**Lab** →		**Bench** →	**Pilot** →	**Full** →			
System	**Beaker** →	**Pieces** →		**Prototypes** →		**Plant** →			
Env.	**Laboratory** →		**Simulated** →		**Relevant** →			**Operational**	
Material	**Thought** →	**Simulants** →		**Simulant/wastes** →		**Simulants** →	**Wastes** →		
Decisions	**CD0** →			**CD1**		**CD2/3**		**CD4**	
TRL	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**

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