GNEP Waste Form Technology Development

Global Nuclear Energy Partnership

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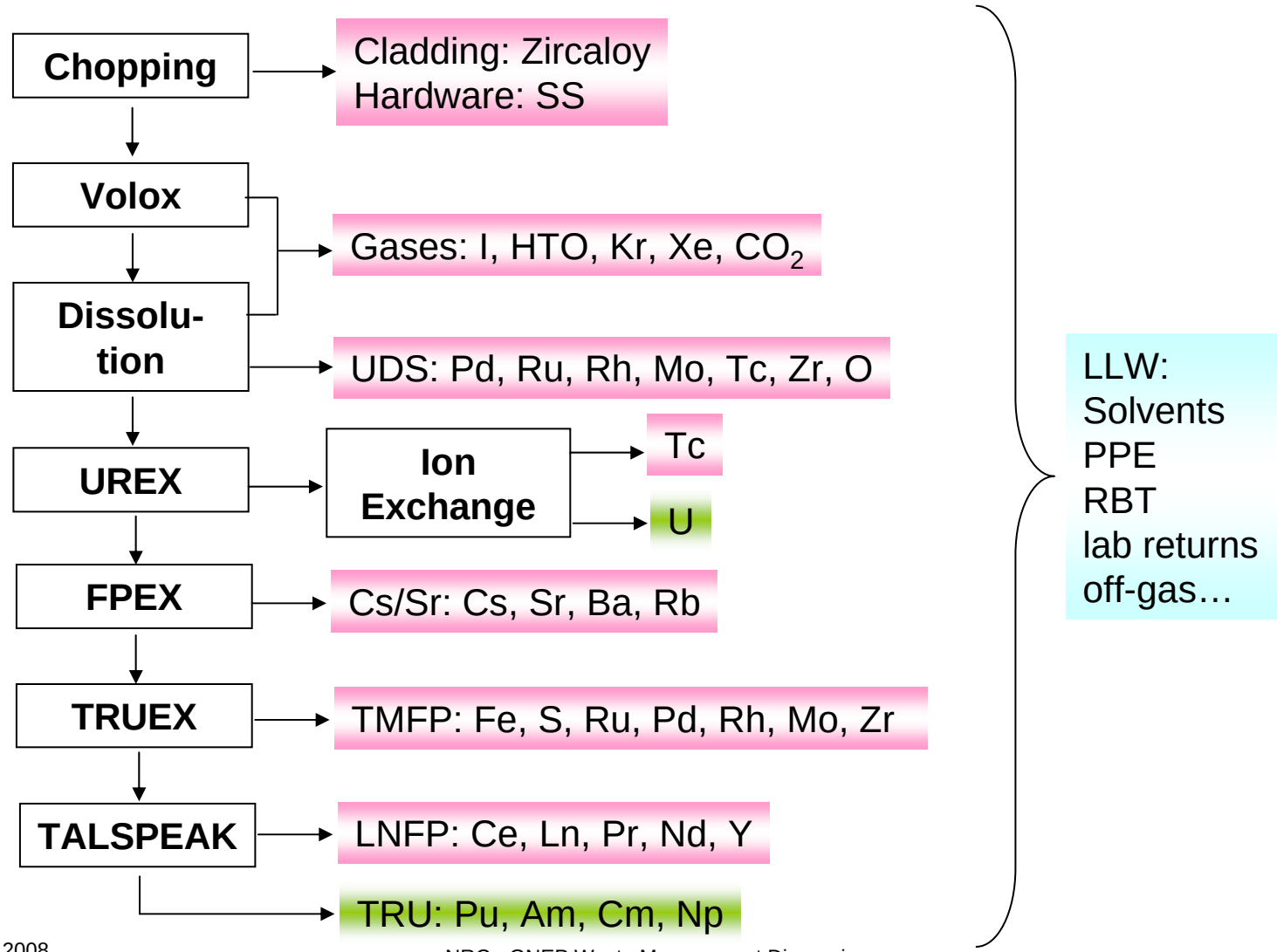
Outline

- Aqueous Waste Streams and Waste Forms
- Electrochemical Waste Streams and Waste Forms
- Technology Readiness Assessment
- Testing Approaches/Waste Form Qualification

Aqueous Process & Waste Streams

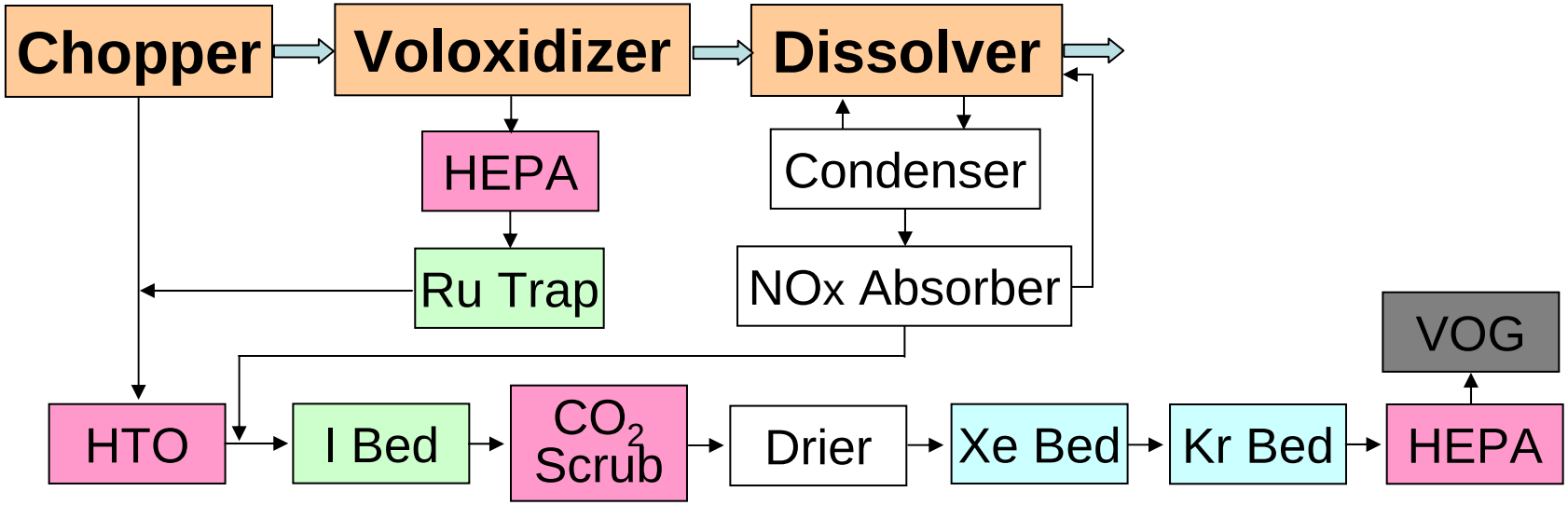


Compacted Metals

- **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

Aqueous Primary Off-gas Streams



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

Ag-Z capture
Remove bed
Encapsulate
Package
Dispose - Rep

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

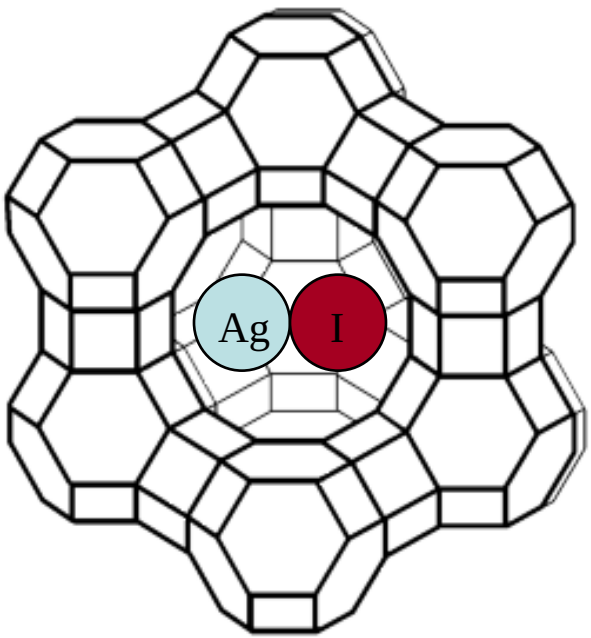
Cryogenic Ag-Z
Release
from Ag-Z
Combine w/ Kr
or Release

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

Remove
Leach
Dispose - SLB

Iodine on Ag-Z

- **Mordenite selected as baseline getter/waste form**
  - demonstrated at pilot scale in US
  - adopted for use in Hanford WTP
  - high DF and loading as getter
  - nitrate tolerant
  - base of a good waste form
- **Likely to be encapsulated**
  - to form monolith
  - improve leach resistance
  - geo-polymer at low temperature (<200°C)
  - low temperature glass (<500°C)



Silver zeolite for ^{129}I capture and immobilization

Aqueous Tc and UDS

Dissolver Clarifier

Filter UDS solids, blend with other metal streams

UREX → **Tc IX** → *$UO_2(NO_3)_2$* → *U_3O_8 or UO_3*

TRU/FP

Reduce to metal

steam reform precipitated $[NBu_4]TcO_4$ w/ C ~ 800°C

Blend

Potential additives: Zr (hulls), Fe (SS), UDS, reduced TMFP; mix in crucible (e.g. coated C)

Melt

furnace under Ar, N_2 or vacuum $\leq$1600°C

Package

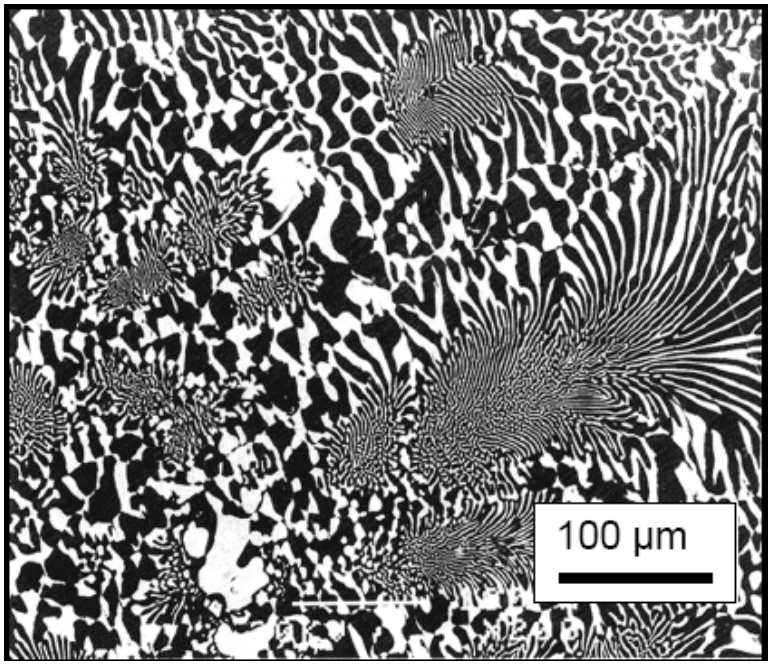
remove crucible from furnace, load into canister, decon canister, seal

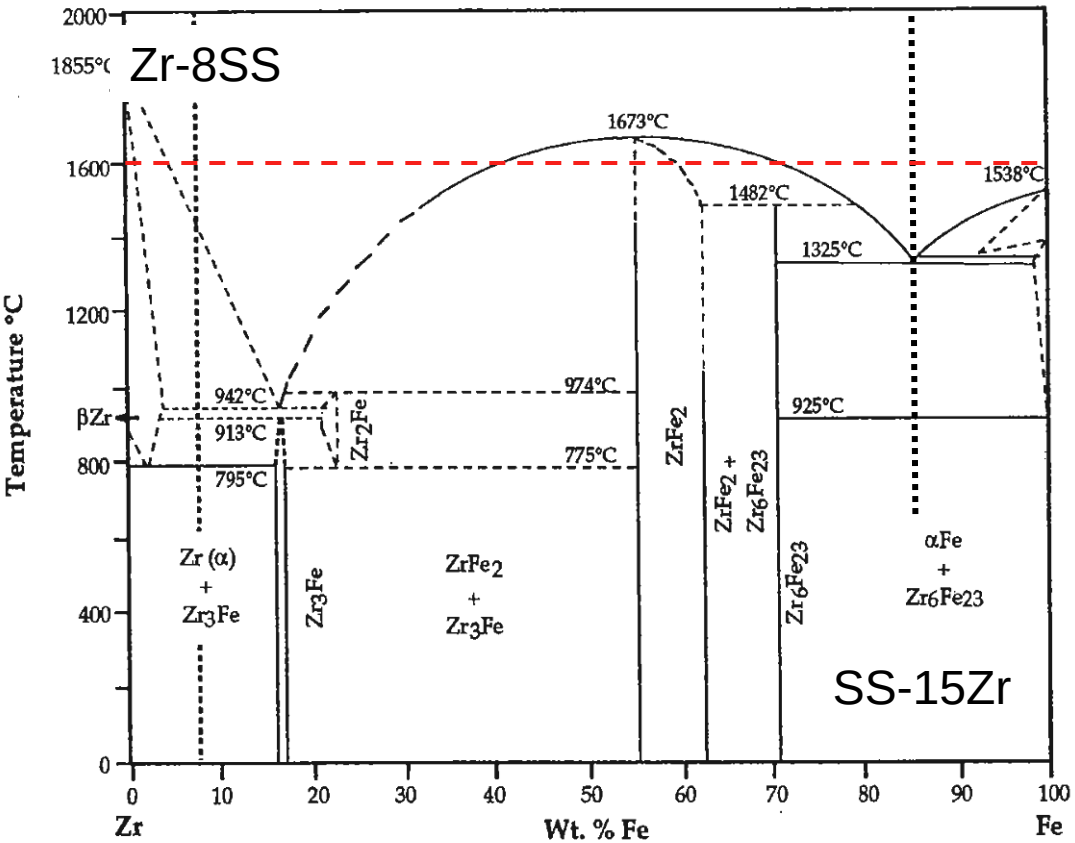
Dispose

repository

Metal Waste Form

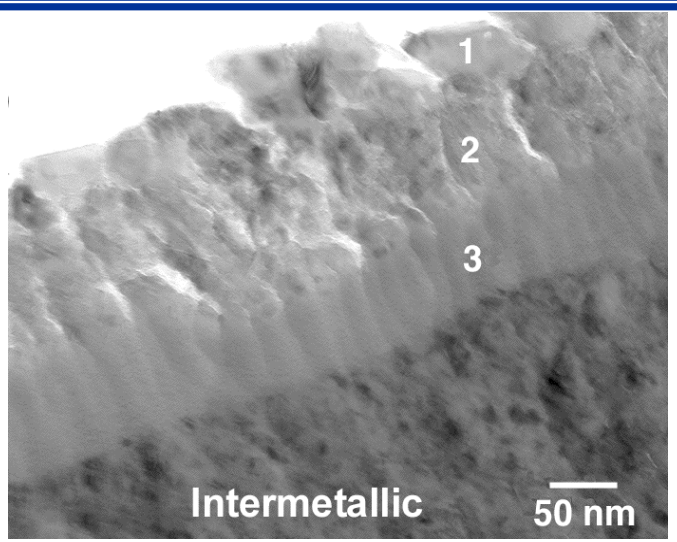
- **Based on Zr-Fe alloys**
  - solubility of Tc, PGM, Mo, and other impurities in durable phases
  - corrosion rate similar to alloy 22 and SS316

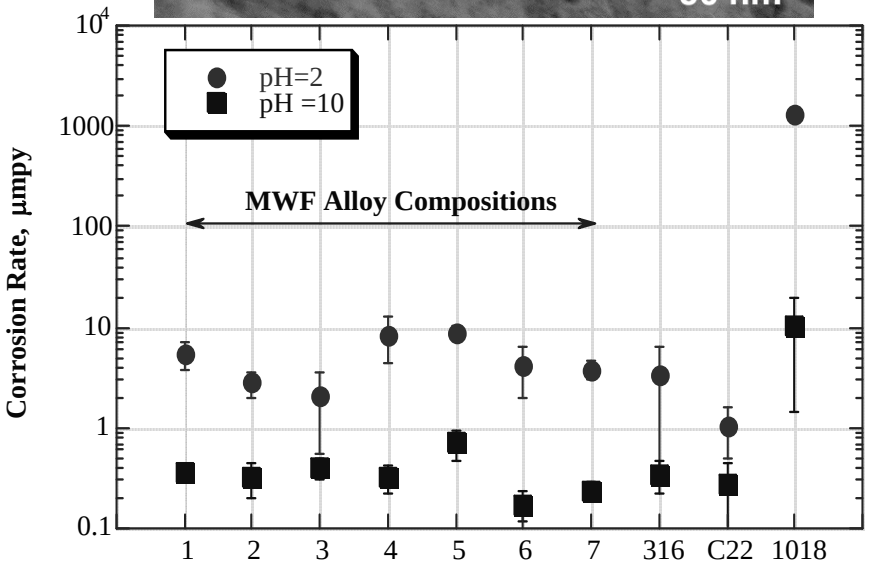


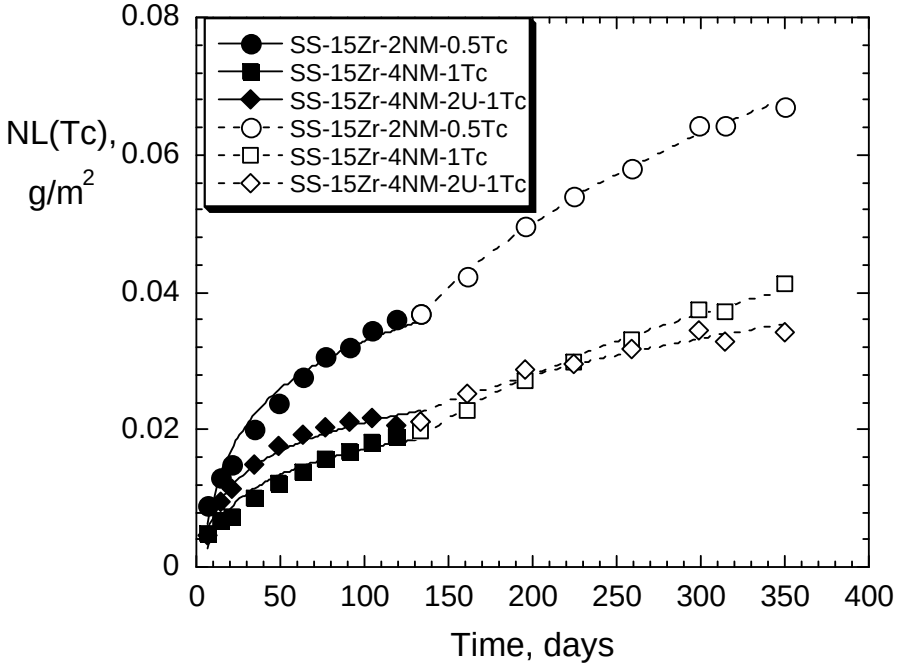


Testing to Evaluate the Suitability of Waste Forms Developed for Electrometallurgically-treated Spent Sodium-bonded Nuclear Fuel for Disposal in the Yucca Mountain Repository, ANL-05/43, Argonne National Laboratory, 2005

Metal Waste Form Corrosion

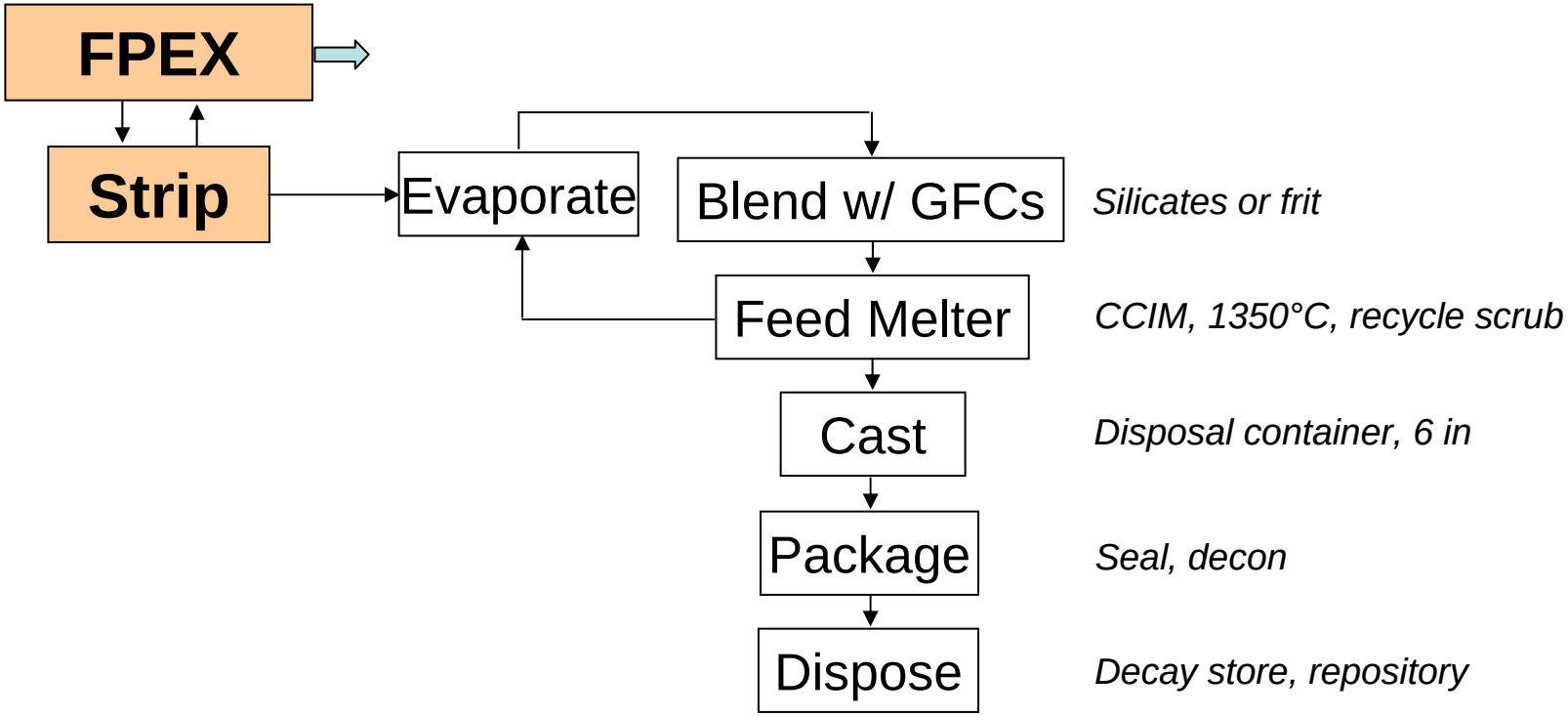






1: SS-15Zr-5U-2Tc
2: SS-15Zr-1Nb-1Pd-1Rh-1Ru-1Tc
3: SS-20Zr-1Nb-1Pd-1Rh-1Ru -1Tc
4: SS-5Zr-1Nb-1Pd-1Rh-1Ru -1Tc
5: SS-15Zr-0.6Ru-0.1Pd-11U-0.3Tc
6: SS-15Zr
7: SS-15Zr-1Nb-1Pd-1Rh-1Ru

Aqueous Cs/Sr



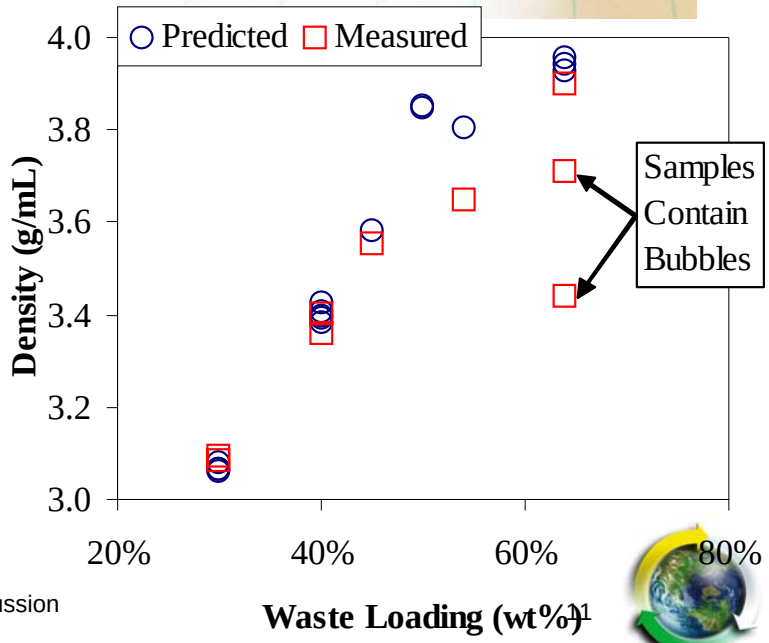
Cs/Sr Glass

- **Based on well developed and demonstrated technology**
- **Alumino-silicate glasses and glass ceramics**
  - Fe_2O_3 or SnO_2 used for charge compensation, e.g.:

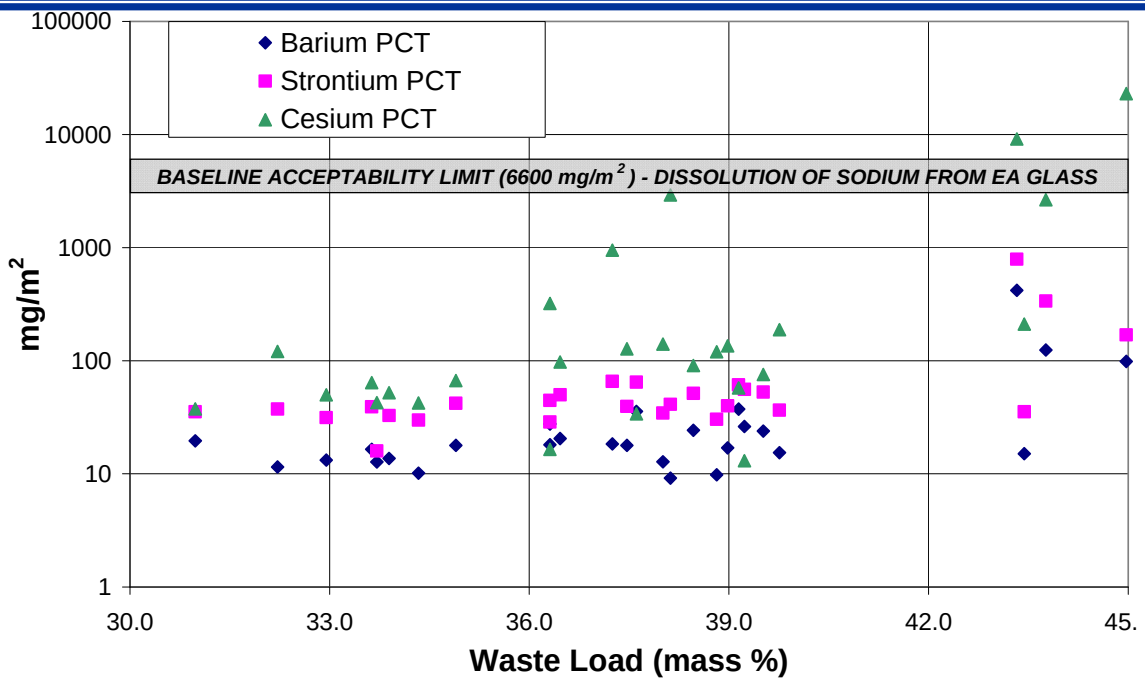
$$SrO + Fe_2O_3 \xrightarrow{\beta\beta} ZrO_2 + 2FeO$$

$$Cs_2O + Fe_2O_3 \xrightarrow{\beta\gamma} 2BaO + 2FeO$$

  - 30 to 64 wt% waste loading
  - density between 3 and 4 g/mL



Cs/Sr Glass/Glass Ceramic



- **PCT responses well below EA glass**
- **Between 0 and 52% crystals* (primarily pollucite, celsian, and other alumino-silicates)**
- **TCLP responses as a function of waste loading pending**

* a roughly 80% crystal melt was made, but, wouldn't pour

Aqueous Transition Metal Fission Products

TRUEX Raffinate

Reduce — *Electroreduction*

Blend — *Tc, Fe, Zr, UDS; in crucible*

Melt — *Inert or reducing cover, ≤1600°C*

Package — *Remove crucible, load canister, decon canister, seal*

Dispose — *Repository*

Transition Metal Fission Products Metal

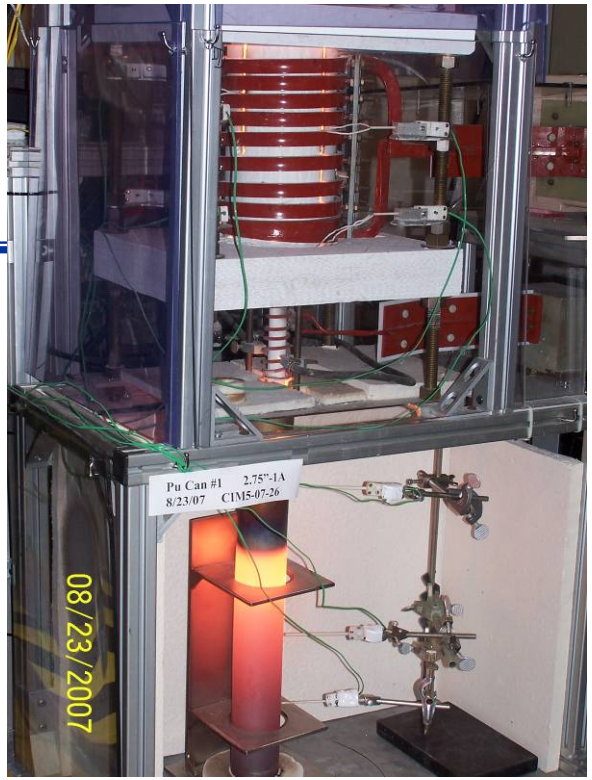
- **Contain significant concentrations of platinum group metals (PGM) not soluble in glass**
- **Alloy properties the same as earlier described metal**
- **Adds little to volume of metal waste form volume, but, would increase glass volume by ~3×**
- **Requires reduction from nitrate solution → electrochemical reduction is being investigated**

Aqueous Lanthanide Fission Products Glass

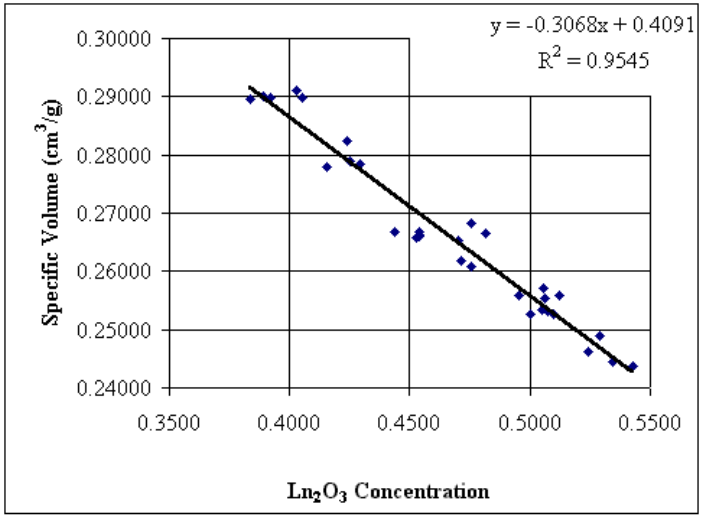
TALSPEAK Product

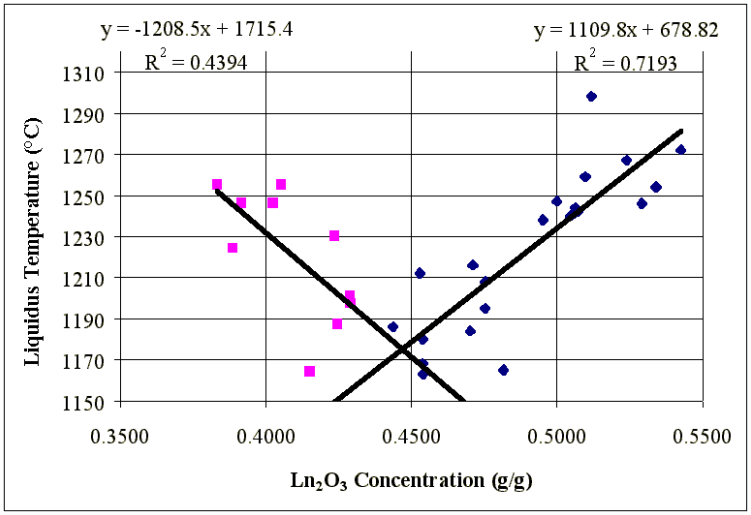
Blend	*Glass forming chemicals, (potentially Cs/Sr and/or TMFP)*
Feed Melter	*HWIM, 1350°C (or CCIM if w/ Cs/Sr and/or TMFP)*
Cast	*Disposal container*
Package	*Seal, decon*
Dispose	*Repository*

Lanthanide Glass



- **Based on well developed and demonstrated technology**
  - developed for SRS Am/Cm treatment
  - chosen and demonstrated for PIP
- **Loadings at 60 wt% (oxide basis)**
- **Outstanding durability**





Property Data for Simulated Americium/Curium Glasses, PNNL-13009, Pacific Northwest National Laboratory, 1999

Electrochemical Process & Waste Streams

Electro-Chemical Metallic Wastes

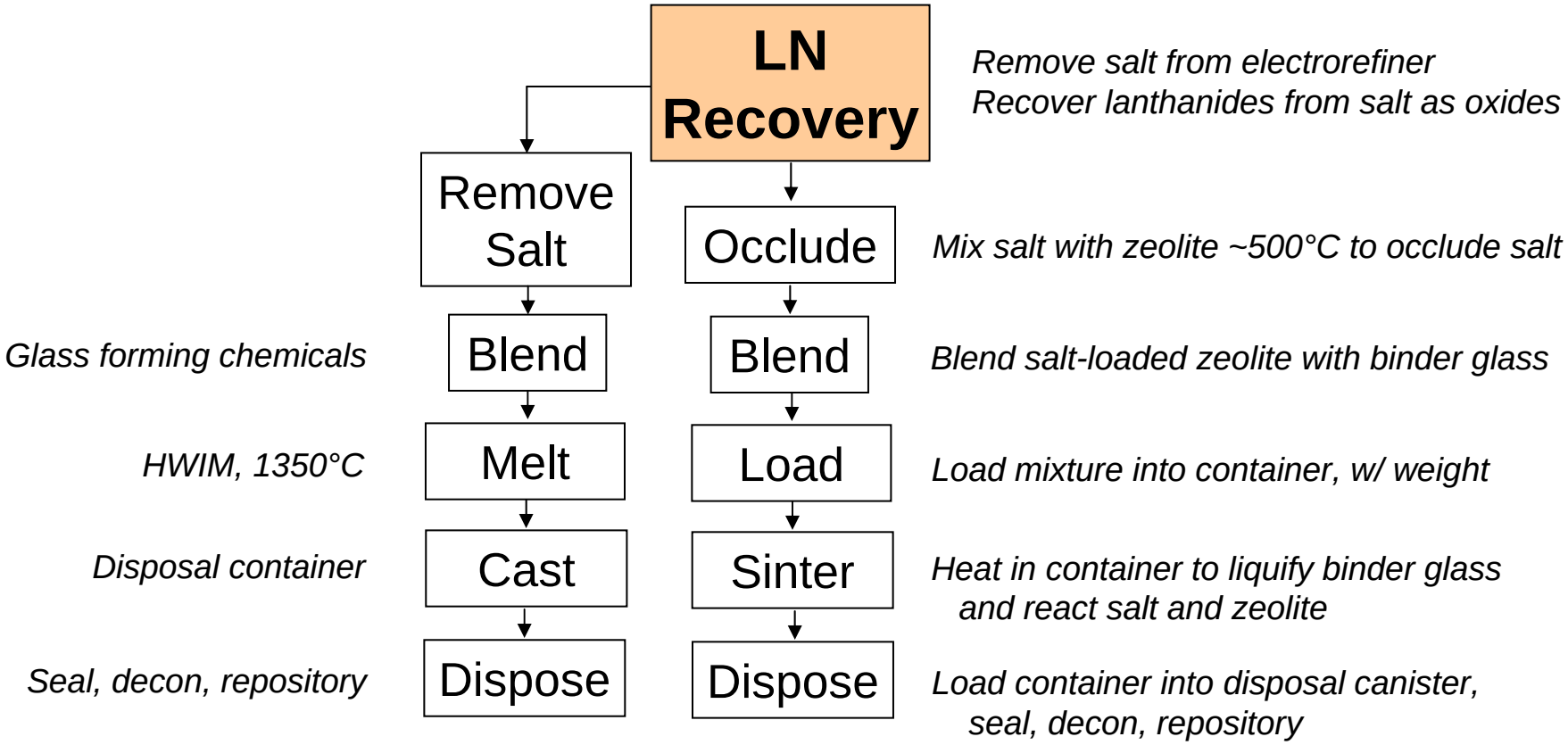
Electro-Refiner

Drain	*Remove anode basket from electrorefiner and drain salt*
Distill	*Transfer to crucible furnace and heat to distill residual salt*
Additives	*Add trim chemicals needed for desired alloy composition*
Melt	*Heat to ~1600 °C under reducing conditions*
Package	*Load ingot into disposal canister, seal, decontaminate*
Dispose	*Repository*

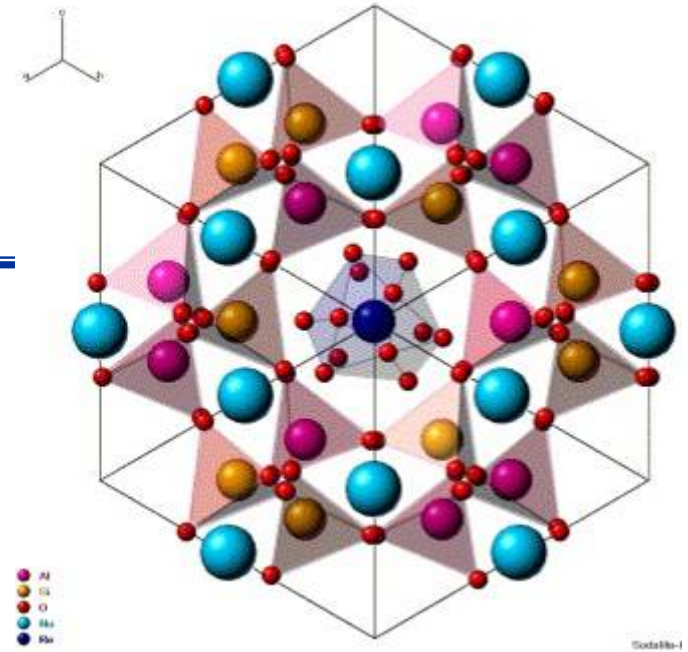
EChem Metal Waste Form

- **Developed for EBR-II fuel and blankets**
- **Demonstrated at full scale**
- **Same SS-15 Zr discussed for aqueous wastes**
  - high durability
  - qualification testing well underway

Electro-Chemical Fission Products



Glass Bonded Sodalite



- **Developed for EBR-II fuel/blankets**

$$Na_{12}(AlSiO_4)_{12} + 4NaCl \rightarrow 2Na_8(AlSiO_4)_6Cl_{12}$$

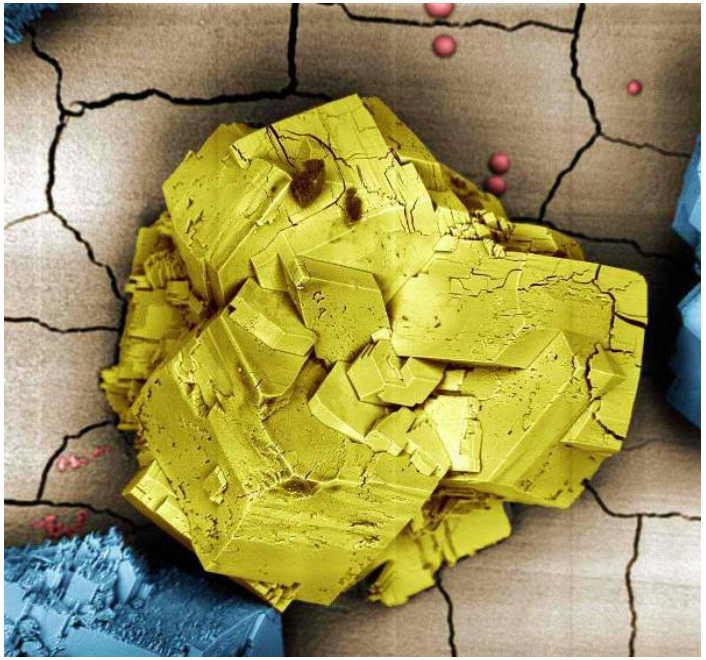
- **Demonstrated at full scale**
- **Durability determined by glass corrosion rate → HLW rate-law used**

Waste Form Testing

- **Waste form testing performed to meet three objectives**
  - model/predict long-term degradation
  - demonstrate qualification
  - define acceptable operating range
- **The tests best suited to meet these objectives are in various states of development, acceptance, and use**
  - appropriate test for more mature waste forms (e.g., HLW glass) are readily available
  - testing protocol for less common nuclear waste forms (e.g., encapsulated zeolite with AgI) have not been as well established
- **Panel evaluated the test methods appropriate to meet each of the objectives**

Testing Protocols for GNEP Waste Forms

- Test method selection and testing protocols based on consensus standards (ASTM, ANS, …)
- Panel recommended initial testing for each baseline waste form for each disposal condition
- More complete list for glass and ceramics
- Acceptable test responses depends on a host of assumptions
- General repository release models being developed to aid in determining acceptable test responses for various environments



Hanford Immobilized LAW Product Acceptance Testing: Tanks Focus Area Results, PNNL-13744, Pacific Northwest National Laboratory, 2001.

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

Technology Readiness Assessment

- **Screening technology readiness assessment performed for each of the key waste streams**
- **Certain technology elements for each waste form rank low on the readiness scale**
  - Generally resulting in low TRL for all waste forms
  - Misleading to take TRL out of context
  - Qualitative judgments were made based on EM TRA process
- **Screening TRA being reviewed by an independent expert panel and is being updated before issue**
- **Complete TRA to be performed after resolution of some waste form baseline issues**

Technology Readiness Assessment

Stream	Description	Process/Form	Maturity
Hulls (LWR)	Zirc-alloy	Leach, compact	H
^{3}H (both)	HTO	Cement	H
I (Aq)	AgZ	Encapsulated AgZ	L
Kr (both)	AgZ	Compressed gas	M
^{14}C (Aq)	CO_3 in NaOH soln	Cement	H
UDS (both)	PGM, Mo, Tc, Zr, (O in Aq, SS in Echem)	Metal alloy	L
^{99}Tc (Aq)	IX eluate	Metal alloy	M
Cs/Sr/Ba/Rb (Aq)	HNO_3 soln	Aluminosilicate ceramic or glass	M
LNFP (both)	LN in HNO_3 soln (Aq) LN_2O_3 (Echem)	Glass	H
TMFP (Aq)	Fe, S, PGM, Zr, Mo in HNO_3 soln	Metal alloy	L
FP (Echem)	Cs, Sr, Ba, Cl, K, Li	Glass bonded sodalite	M

Waste Form Strategy Evolution

- **Critical evaluation of waste management baseline currently underway**
- **New baseline will strive to**
  - minimize complexity and number of waste processes
  - minimize amounts of various waste types
  - use nature to guide → match waste form to waste and disposal chemistries
- **Key unknowns/uncertainties**
  - repository type (Yucca Mtn currently limited to 70,000 MTHM)
  - waste type classification (NWPA based on PUREX)
  - requirements for capture of fission product gases
  - GTCC disposal

Treatment of Aqueous Wastes

Hulls **Hardware**	Compact	Metal for GTCC or Repository Disposal
HTO **$^{14}CO_2$**	Capture Cement	Cement for LLW Disposal
Iodine	Capture Encapsulate	Zeolite in Geo-polymer for Repository Disposal
UDS **Tc** **TMFP**	Reduce Melt	UDS, Tc, TMFP Alloy for Repository Disposal
Cs/Sr **LN**	Vitrify Decay store	Cs/Sr, LN Glass for Repository Disposal

Treatment of Electrochemical Wastes

HTO	Capture Cement	Cement for LLW Disposal
UDS	Remove salt Melt	Alloy for Repository Disposal
LNFP	Separate Remove salt Vitrify	High Ln Glass for Repository Disposal
Cs/Sr **OFP**	Remove Cl Immobilize	Glass or Ceramic for Repository Disposal

Conclusions

- **GNEP waste treatment is on sound foundation**
  - waste forms selected to match waste chemistry and disposal environments → nature guides selection
  - many waste processes are based on mature technologies
  - no technical *show stoppers* identified
- **Waste form formulation and testing is well underway**
  - preliminary testing on all key waste forms
  - test methods being developed to meet three testing criteria
  - program in place to fill technology and qualification gaps
- **Waste form campaign is evolving**
  - reduce complexity and amount
  - take advantage of unique waste characteristics
  - focus efforts on increasing technology readiness