Alternatives for Low-Temperature Waste Immobilization

Cementitious Materials Technical Exchange December 12-14, 2006 J. H. Westsik, Jr., R. J. Serne Pacific Northwest National Laboratory

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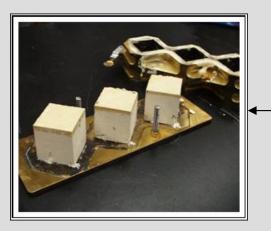
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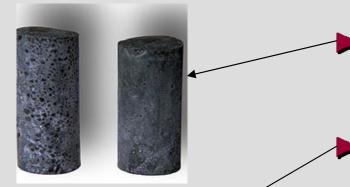
Project Scope

- Seek solutions from private sector to demonstrate lowtemperature immobilization technologies for mixed radioactive and hazardous waste
 - Low-temperature waste forms to contain volatile radionuclides including Tc and I
 - Mature technologies with feasibility of deployment within 1 to 2 years
- Conduct screening tests using low-temperature (<150°C) immobilization technologies on Hanford WTP caustic scrubber and Idaho Sodium Bearing Waste (SBW) simulants



Waste Forms Selected for Evaluation





PURALITH"

GEOPOLYMER LOW TEMPERATURE WASTE FORM

- Alkali Aluminosilicate Hydroceramic Cement: Diagnostic Instrumentation and Analysis Laboratory (DIAL) at Mississippi State University
 - **Phosphate Bonded Ceramic** (Ceramicrete - magnesium potassium (Mg-K-PO₄) system): CH2MHILL

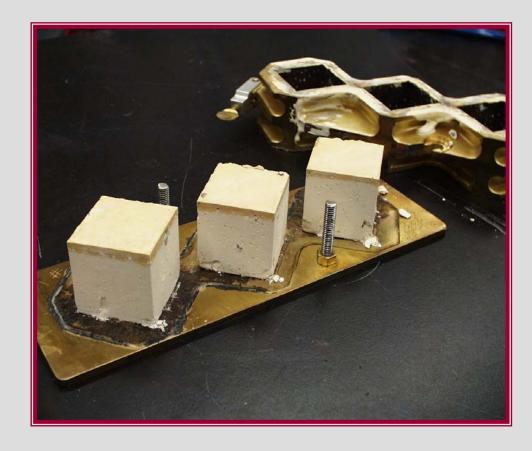
Alkali Aluminosilicate Geopolymer ("DuraLith"): The Catholic University of America Vitreous State Laboratory (VSL)



Alkali Aluminosilicate Hydroceramic Cement

MSU-DIAL

- Ingredients:
 - Sodium Hydroxide
 - Metakaolin
 - Vermiculite
 - Silica
 - Sodium Sulfide
 - Waste
- Developed as higher temperature process yielding insoluble sodalites and zeolites



Phosphate Bonded Ceramic

CH2MHILL

Ingredients:

- Magnesium oxide
- Potassium acid phosphate
- Calcium silicate
- Waste
- MWFA-developed technology



Ceramicrete Idaho Sodium Bearing Waste



Alkali Aluminosilicate Geopolymer

CUA-VSL

Ingredients

- Silica and alumina source (proprietary)
- Alkaline solution
- Forms amorphous or partially microcrystalline geopolymer



Hanford Secondary Waste Simulant

Element	Target (moles/L)	Target (g/L)
Na	2	46
AI	0.011	0.299
Cr	2.80E-04	0.0145
Ag	2.20E-04	0.0237
Cd	1.40E-05	0.00157
Re (Tc)	6.00E-07	1.12E-04
I	2.90E-06	3.68E-04
Hg	2.40E-06	4.81E-04
Pb	1.50E-04	0.031
CO ₃ -	0.96	57.6
NO ₃ -	0.018	1.116
OH-	0.094	1.598
TOC	0.18	13.86
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Sodium Bearing Waste Simulant

Element	Target (moles/L)	Target (g/L)	
Na	1.88	43.24	
AI	0.575	15.5	
K	0.175	6.825	
Cr	0.0033	0.172	
Cd	0.0007	0.0786	
Hg	0.002	0.401	
Pb	0.0013	0.269	
Re (Tc)	3.13E-06	5.83E-04	
I	5.66E-05	7.18E-03	
SO ₄ -	0.0491	4.71	
NO ₃ -	4.91	304.4	

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Waste Form Characterization

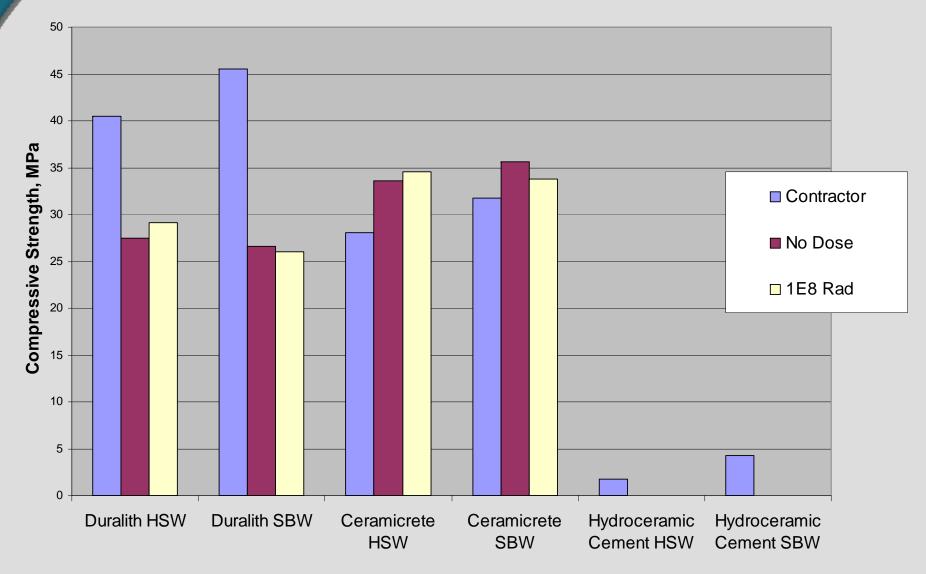
Contractors prepared and characterized waste forms using PNNL provided simulant

- Chemical composition
- Waste loading
- Toxicity Characteristic Leaching Procedure
- Compressive strength
- PNNL conducted additional testing on Contractorprovided specimens
 - Product Consistency Test
 - ANSI/ANS 16.1 Leachability
 - Compressive strength after exposure to radiation

TCLP Results, mg/L

	Dur		Carran	ievete	Hydroceramic	
	Duralith		Ceramicrete		Cement	
	HSW	SBW	HSW	SBW	SBW	UTS
Ag	<0.07	<0.07	0.008	0.003		0.14
Cd	< 0.03	<0.03	<0.0003	< 0.0003	0.6	0.11
Cr	0.015	0.04	0.05	<0.002	0.003	0.6
Hg	<0.01	<0.01	<0.008	<0.008	0.1	0.025
Pb	<0.1	<0.1	0.01	0.001		0.75

Compressive Strength



PCT Results

- Using standard PCT sample preparation, observed evidence of sample dissolving or breaking up
- Tried washing with acetone and using larger particles
- Results presented in final report (PNNL-16052 1)
- Significant work needed to understand non-glass waste form performance in PCT and to interpret in terms of disposal facility

ANS-16.1 Leachability Index Results

		Leachability Index
Sample Material	Waste Type	Na > 6
Ceramicrete	HSW	8.1
	ПЭМ	8.2
		7.6
	SBW	7.6
DuraLith	HSW	8.6
	ПЭМ	8.6
		8.3
	SBW	7.7



ANS-16.1 Leachability Index Results (Re and I)

Duralith

- Bettered LI target (Tc LI > 9) for both simulants
- Did not meet LI target (I LI >11) for either simulant

Ceramicrete

- Did not meet target for Tc
- No I detected in leachate so could only report less than values below target
- CH2M Hill report 7-day LI >11 for both I and Re in highly spiked simulants

Testing with improved analytical sensitivity required to have confidence in Re and I results

Waste Forms After 90 Days in ANS-16.1



Ceramicrete

Duralith

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Summary

- Three low-temperature waste forms selected for scoping studies for Hanford secondary waste and Idaho sodiumbearing waste
- "Ceramicrete" phosphate bonded ceramic and "Duralith" alkali aluminosilicate geopolymer show potential based on TCLP, compressive strength and Na leachability index requirements
- ANS-16.1 immersion test revealed formulation issues that will need to be addressed
- Re (Tc) and I immobilization not easily demonstrated using leach tests at expected low waste concentrations---analytical detection limit problem

Next Steps

Address the formulation issues identified through the ANSI/ANS 16.1 testing and perhaps also in the PCT testing.

- Using waste simulants spiked with higher concentrations of I and Re, determine whether the waste forms can achieve sufficient reduction in the release of I and Tc.
- Based on observations from ANSI/ANS 16.1 testing and contractor thermal cycling tests on Ceramicrete, expand compressive strength testing to include impacts of thermal cycling and immersion in water.
- Optimize the quantities of binder materials to improve waste loading. If acceptable, this may include the removal of water to concentrate the wastes to be immobilized
- Examine other binder materials that may be less costly.
- Demonstrate the long-term effectiveness of sequestering agents added to reduce the mobility of iodine and technetium in the wastes.
- Demonstrate the preparation of the waste forms on a production scale.
- Determine long-term waste-form-performance characteristics to support disposal-system performance assessments.

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Project and Technical Risks/Issues/Opportunities

Technologies at different stages of maturity

- Waste form development / optimization
- Process development / demonstration
- Waste form characteristics to support disposal system performance assessments

Out-year activities will be defined based upon ORP and ID decisions whether or not to pursue alternative low-temperature immobilization technology---are any of these LT waste forms really adequate?