ABSTRACT Contact: James Jordan, President and CEO RIC, LLC Phone: 703-241-8711 Fax: 703-241-8714 Email: james.jordan@ricllc.com

#### AVS: EXPERIMENTAL TESTS OF A NEW PROCESS TO INDUCTIVELY VITRIFY HLW INSIDE THE FINAL DISPOSAL CONTAINERS AT VERY HIGH WASTE LOADINGS

A series of Advanced Vitrification System (AVS) tests using DOE-provided waste simulant were completed in 2001. These tests produced borosilicate glasses at waste loadings of 35% and 50%, not accounting for the reduced usable volume of the AVS canister. A recent independent evaluation sponsored by the DOE unanimously found that these test glasses produced by RIC in 2001 were in compliance with the waste leachability and durability requirements, however they did not meet chemical composition and phase stability requirements.

**J. Powell, Presentor**, M. Reich, J. Jordan, L. Ventre, R. Barletta, B. Manowitz, M. Steinberg, W. Grossman, G. Maise and F. Salzano of RIC, LLC and C. Hess, Burns & Roe; and, W.G. Ramsey and M.J. Plodinec, DIAL, Mississippi State University

The design and performance capabilities of the Advanced Vitrification System (AVS) are described, together with the results of experimental tests.

The AVS is an in-can melting system composed of several nested containers. The completed canister assembly is called a module. The outer shell of the module is a typical outer stainless steel container. The next inner container is a layer of insulation. Within that is an alumina-lined graphite susceptor or crucible. The graphite crucible is inductively heated to very high temperatures (up to 1500°C) by an external low frequency (30 Hertz) AC transformer coil. The air-cooled outer stainless canister remains at near ambient temperature. After the melted HLW has solidified and cooled to ambient temperature, the HLW feed and off-gas pipes are disconnected from the top head of the module, which is then sealed and readied for shipment or storage. All radioactively contaminated melter components inside the disposal module are disposed of along with the vitrified waste. The graphite crucible also provides a geologically stable barrier for the vitrified product.

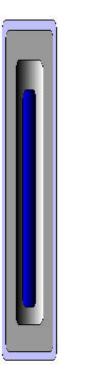
The AVS potentially can double HLW loading over that obtained from Joule melters; lower vitrification costs by about half; reduce the number of disposal canisters required by about half; handle diverse waste feeds with high concentrations of problem elements such as chromium and zirconium; and reduce the time needed to vitrify a given inventory of HLW.

The AVS can operate at temperatures up to 1500 °C because its refractory materials are at melt conditions for only a few hours, instead of years required for conventional melters. This permits making a wide range of vitrified products from borosilicate glass to highly leach resistant glass ceramics. The AVS process minimizes the necessity for waste feed blending and eliminates the cold cap, the need to control melt viscosity and electrical conductivity and the requirement for pouring. Since the AVS is a modular system with each canister-processing cell offering operational redundancy, it minimizes technical risk and the possibility of a single point failure stopping operations. Because each AVS module contains a single-use melter, there is no subsequent D&D requirement.

Bench scale tests of the AVS were carried out at the DIAL Laboratory at Mississippi State University. The tests produced 5-inch diameter AVS borosilicate glass logs with simulated HLW loadings of 35 and 50% by weight. The simulated HLW material was supplied by DOE and corresponded to Hanford Envelope D waste. The product glasses met or exceeded PCT and TCLP requirements at both HLW loadings. Within the accuracy of measurement, cesium was retained in the product glasses. No appreciable corrosion of the alumina liners inside the graphite crucibles was observed. The amount of crystallinity was small, less than 1% at 35% HLW loading, and less than 5% at 50% loading. Crystallinity tests on the 50% HLW loading showed satisfactory stability upon heat treatment after vitrification. Small-scale lab tests indicate that even higher HLW loadings in excess of 60%, appear possible. Additional bench-scale tests have been carried out on problematic wastes containing very high concentrations of chromium, zirconium, and bismuth, with excellent durability behavior.

The process parameters, design, and costs of a full-scale AVS facility to vitrify the Hanford HLW waste are described. The time to process a given AVS module is 3 days, including heatup, vitrification and cooldown. A total of 6000 AVS modules would be required to vitrify all Hanford tank waste, a much smaller number than that required using a conventional Joule melter. The AVS facility, with a projected capital cost of \$236 million dollars, would have 12 process cells, with a capability of producing 4 disposal modules per day. Assuming waste retrieval or other operations were not a constraint—which is probably not the case—the AVS facility could vitrify all Hanford HLW in about 10 years.





Argus Remediation, Inc., Burns & Roe Enterprises, Inc. James Jordan James R. Powell, Ph.D. Morris Reich, Ph.D. UCAR Carbon Company, Inc. Louis Ventre, Jr. Esq.

#### AVS ADVANCED VITRIFICATION SYSTEM Additional Testing Project DE-AC26-00NT40801

Presentation to Industry Partnerships for Environmental Science and Technology Conference

November 1 at NETL, Morgantown, WV James Powell, Vice President and Chief Scientist

## Acknowledgements

#### • NETL

- Denise Riggi, Contracting Officer's Assistant
- George Bellas, Contracting Officer's Technical Representative
- TFA
  - Ted Pletrok
- CTC, Inc. (Independent Review Team)
  - Steve Ostheim,
- DIAL
  - John Plodinec
  - Gene Ramsey

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#### What is the AVS system?

- HLW is vitrified inside the final disposal canister, using inductive heating of an inner graphite/alumina crucible.
- After cooling, the closed canister is sent to the repository for disposal.
- No separate melter, no pouring, no need for D&D of melter equipment.
- 1350 degree centigrade graphite/alumina crucible is thermally insulated from cool outer stainless can.
- AVS can vitrify wide range of waste compositions with minimum pretreatment, and can achieve much higher waste loadings in canisters than possible with conventional melters.



## AVS Technology Overview





Top Head

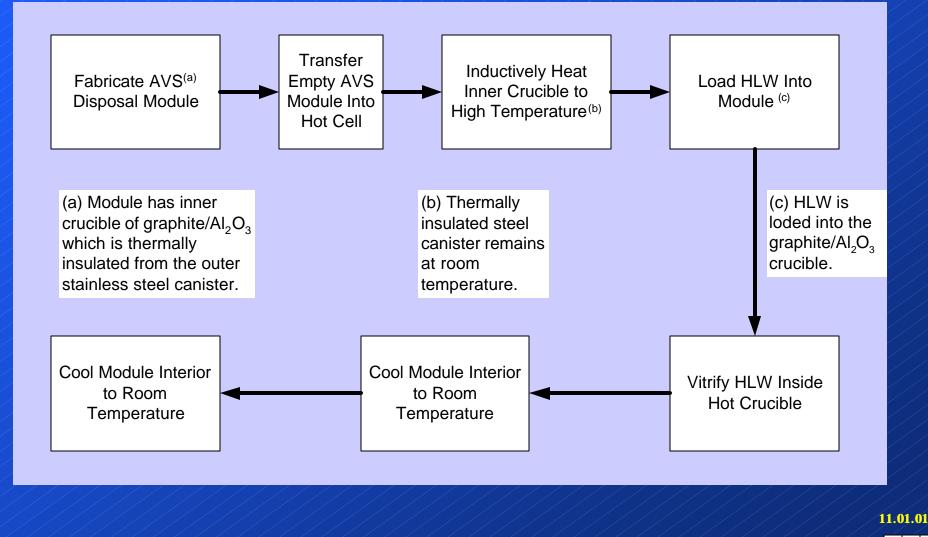
Alumina Liner = .5cm

Graphite Crucible =2.0 cm

Graphite Fiber Insulation = 1.0 cm

Stainless Steel Canister = 1 cm

## The AVS System: How Does It Work?



## Why AVS?

Compared to the Conventional Melter Systems for Vitrifying High Level Nuclear Waste (HLW) the AVS has the Potential to be:

- Cheaper in Capital and O & M Costs
- More Reliable
- Adaptable to Problem HLW Feeds
- Able to Load Much More HLW into a Disposal Canister
- Less Need for Blending and Pre-Treating the HLW Feed
- Quicker in Carrying Out a HLW Disposal campaign



#### Principal Features of the AVS Module

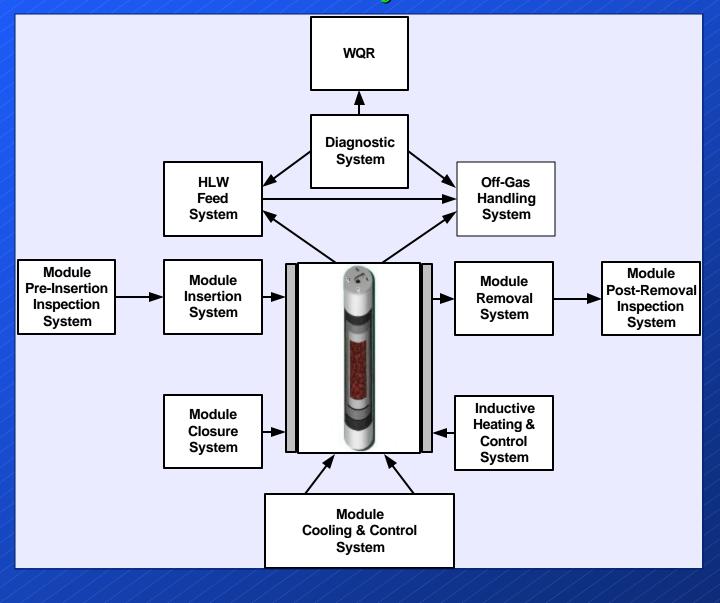
- Inner High Temperature Graphite Crucible
- Outer Stainless Steel Canister at Near Ambient Temperature
- Thermal Insulation Between Hot Graphite Crucible from Cool Stainless
  Canister
- Low Frequency (30 Hertz) AC Inductive Coil Heats Graphite Crucible and Stainless Canister (Forced Air Cooled)
- Dewatered HLW Sludge/Frit Mixture is Extruded at Ambient Temperature Into Hot Graphite Crucible
- Residual H<sub>2</sub>O in HLW/Frit Mix Evaporates as Module is Filled Over 1 Day
- HLW/Frit Mixture is Vitrified inside the AVS Module in a Four Stage Process
- Total Process Time is ~3 Days per Module
- Stainless Canister is Closed After Canister Cools to Room Temperature
- Cooled Module is Removed for Final Disposal

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### AVS Module Parameters

- 4.5 Meters Length
- 0.61 Meter Outside Diameter
- 1 Centimeter Thick Stainless Steel Canister
- 1.0 Centimeter Thick Graphite Fiber Insulation
- 2.0 Centimeter Thick Graphite Crucible (Plasma Sprayed Al<sub>2</sub>O<sub>3</sub> Coating)
- 0.5 Centimeter Thick Alumina Crucible
- 1 m<sup>3</sup> Volume Inside Crucible
- ~3 Day Process Time
- 1,200 Kilograms Empty Weight
- 3,300 Kilograms Filled Weight (70% Fill Efficiency, D<sub>gLASS</sub> = 3,000kg/m<sup>3</sup>)
- 2/1 Ratio, A/C Heating in stainless/graphite
- ~\$1,000 for Electrical Energy to Process Module (assume 10<sup>¢</sup>/kw-hr)

AVS Subsystems



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### Preliminary RIC-AVS Facility Design and Operational Concept



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## Experimental Tests of the AVS Process and Vitrified Waste Form

#### • Phase 1

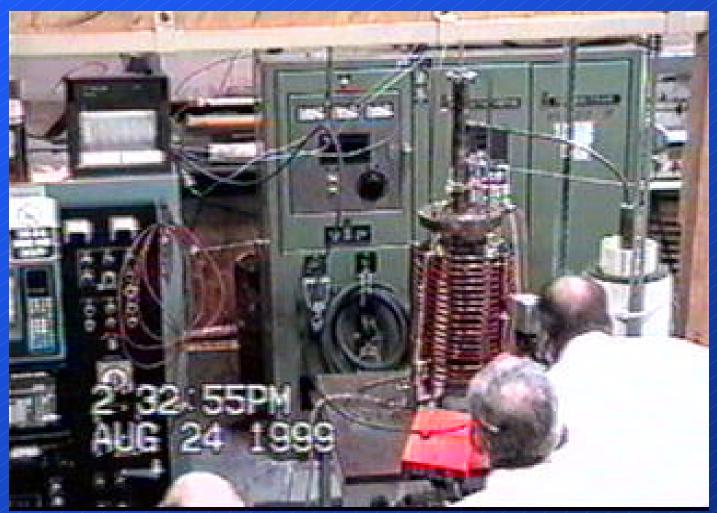
- 5 Inch Diameter Bench-Scale Vitrified Logs
- Variety of HLW Compositions Tested (High Bismuth, High Chromium, High Zirconium)
- Range of HLW Loadings Tested up to 100% HLW Feed (No Added Glass Formers)
- PCT Durability

#### • Phase 2

- Laboratory-Scale (2-½ Inch) and 5-Inch Diameter Bench-Scale Borosilicate Glass Vitrification
- Envelope D HLW Simulant
- 35% and 50% HLW Loadings (SiO<sub>2</sub> and Na<sub>2</sub>O Subtracted)
- PCT, TCLP, SEM, XRD, DTA/TGA, etc Tests
- Continuous Fill Tests

### Phase 1 Results

## **AVS Bench Scale Tests**



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# Log of Phase 1 Tests

Experiment	Waste Loading	Special Chemistry	Chemical Analysis	РСТ	TCLP	T <sub>g</sub>	Crystallinity	
Number							X-Ray	SEM/
	(wt %)	enemistry	7 mary 515				Diffraction	EDX
1	29.5		Х					
2	30.0		Х					
3	28.8		Х					
4	28.8		Х				Х	
5			Х					
6	30.0		Х	Х			Х	
7	49.8	20% Fe <sub>2</sub> O <sub>3</sub>	Х	Х			Х	
8	30.0		Х	Х			Х	
9	36.7		Х	Х			Х	
10	55.7	10% Bi <sub>2</sub> O <sub>3</sub>	Х	Х				
11	89.1		Х	Х				
12	87.5	14.1% ZrO <sub>2</sub>	Х	Х				
13	87.8	4% Cr <sub>2</sub> O <sub>3</sub>	Х					
14	88.1		Х	Х				
15	88.6	2.2% Ag <sub>2</sub> O	Х					
16	88.6	2.2% Ag <sub>2</sub> O	Х	Х				
17					Х			
18			Х	Х	Х	Х	Х	Х
19			Х	Х	Х	Х	Х	Х
X = performed, = No Pre-Treatment/Glass Formers Added								

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## Calculated Glass Product Composition

Pre-Treatment		Washed aste	Enhanced Caustic Leached Waste				
Waste Concentration In Glass → Component in Glass	25%	35%	25%	35%	45%	70%	
SiO <sub>2</sub>	57.5	52.5	57.7	52.8	47.9	35.6	
$Na_2 \tilde{O}$	7.3	6.5	7.0	6.2	5.3	3.3	
$B_2 \tilde{O}_3$	10.5	9.1	10.5	9.1	7.7	4.2	
$Al_2O_3$	8.9	12.5	3.2	4.5	5.8	8.9	
$Fe_2O_3$	2.9	4.1	4.7	6.7	8.5	13.3	
Li <sub>2</sub> O	3.7	3.2	3.7	3.2	2.8	1.5	
CaO	1.0	0.9	1.1	1.2	1.2	1.3	
MgO	1.2	1.2	1.4	1.6	1.7	2.2	
K <sub>2</sub> O	0.3	0.4	0.4	0.6	0.8	1.2	
Cr <sub>2</sub> O <sub>3</sub>	0.3	0.4	0.1	0.2	0.2	0.3	
Ce <sub>2</sub> O <sub>3</sub>	0.5	0.6	0.8	1.1	1.4	2.2	
La <sub>2</sub> O <sub>3</sub>	0.3	0.4	0.4	0.6	0.8	1.2	
Bi <sub>2</sub> O <sub>3</sub>	2.2	3.1	3.6	5.1	6.6	10.2	
BiPO <sub>4</sub>	2.9	4.1	4.7	6.7	8.6		
$P_2O_5$	0.7	1.0	1.1	1.6	2.0	3.1*	
PbO <sub>2</sub>	0.4	0.6	0.7	0.9	1.2	1.9	
Ag <sub>2</sub> O	0.4	0.6	0.7	0.9	1.2	1.9	
ZnO	0.4	0.6	0.7	0.9	1.2	1.9	
MnO <sub>2</sub>	0.4	0.6	0.7	0.9	1.2	1.9	
ZrO <sub>2</sub>	0.4	0.6	0.7	0.9	1.2	1.9	
SrO	0.4	0.6	0.7	0.9	1.2	1.9	
$Na_2SO_4$ as $SO_3$	0.3	0.5	0.1	0.1	0.1	0.1	
Total: 100.0 100.0 100.0 100.0 100.0 100.0							
$P_2O_5$ added as $Na_3PO_4$							

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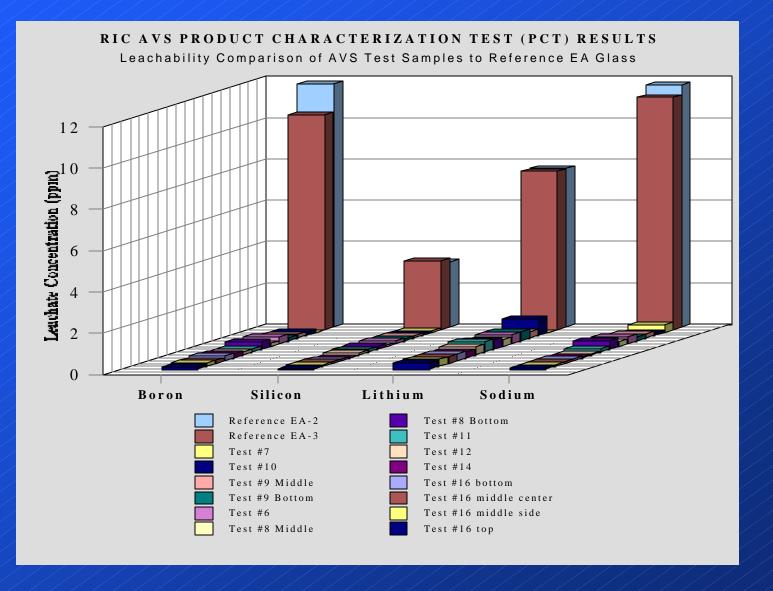
# AVS Bench Scale Test Results **Test 10**

- 70% TFA Waste Simulant
- 30% Frit
- 6 hrs @ 1450°C
- ASTM Leach Results: Better than standard





#### Environmental Protection Advantage



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### Phase 2 Results

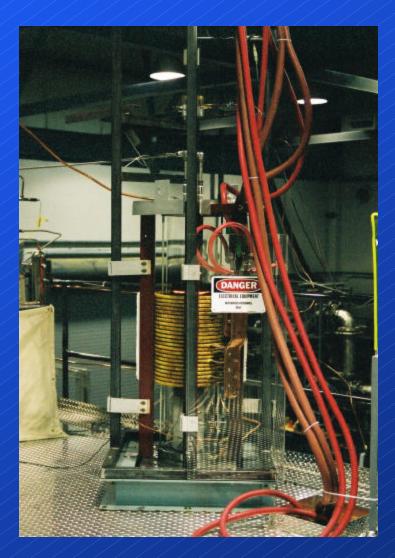
Additional Tests of the AVS Program Objectives and Scope

- Demonstrate that the AVS process can produce vitrified borosilicate waste glass product that meets the WAPS criteria for Hanford waste
- Two waste glass formulations to be produced at Bench Scale (5 inch diameter glass logs)
  - 35 % waste loading
  - Maximum waste loading that meets WAPS criteria
- Envelope D waste simulant and definition of waste loading provided by DOE
- Describe implications of results for full scale AVS process.
- Compare AVS process with conventional joule melter.

### Additional Tests of the AVS Overview of Project Technical Approach

- Perform laboratory scale experiments to define parameters, protocols, and performance relevant to bench scale experiments, including:
  - Process Times, Temperatures, Heating/Cooling Rates, etc.
  - Effect of waste loading fraction on glass durability, crystallinity, etc.
  - Method of filling
  - Potential limitations on process (foaming, corrosion, etc.)
- Perform bench scale experiments using
  - Inductively heated, insulated, graphite/alumina crucibles
  - Reconstituted test apparatus derived from previous AVS experiments at Argus Remediation, Inc.
- Perform battery of analytic tests on feeds and products (chemical composition of solids and off-gasses, DTA/TGA, PCT, TCLP, XRD, SEM, etc)

Additional Tests of an Advanced vitrification system AVS AVS Test Stand



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### Additional Tests of the AVS Summary of Project Results

- Two Bench Scale Borosilicate Glass Logs Produced
  - 35% and 50% waste loading
- Glass Logs Satisfied PCT, TCLP, Crystallinity & Homogeneity Criteria
  - PCT leach rates less than 1/10<sup>th</sup> of standard EA glass
  - TCLP Tests met UTS standards
  - Low crystallinity (1- 5%)
  - Uniform composition and properties measured throughout glass product

#### Bench and Laboratory Scale Tests Demonstrated

- No appreciable corrosion of alumina crucible at anticipated process conditions (1350 degree centigrade refining temp)
- No foaming problems
- Ability to continuously fill crucibles to high L/D ratios (>10/1)
- High retention of cesium in glass product

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## Additional Tests of the AVS Project Results

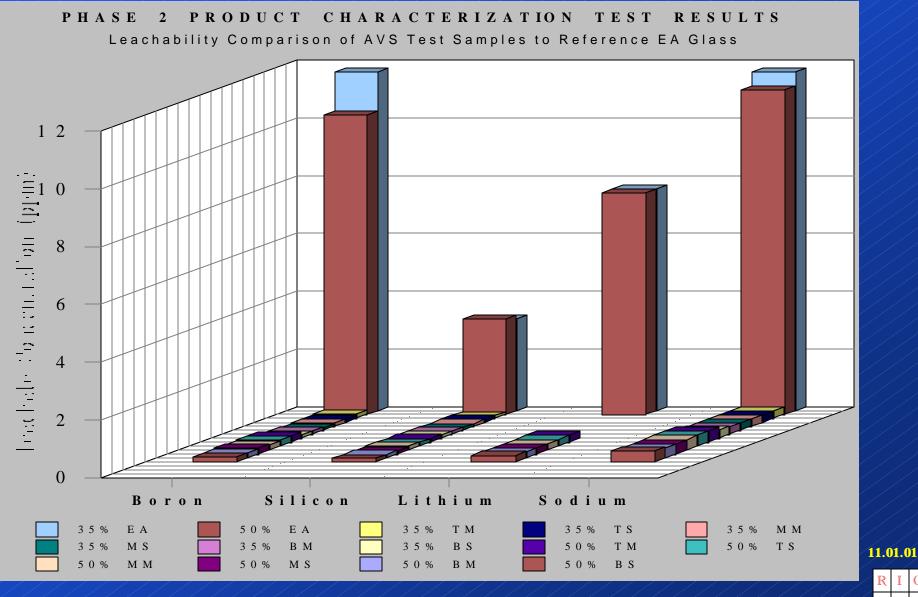
- Maximum weight of 50% HLW AVS canister is 3630 Kg. Well within 4200 Kg Limit
- AVS canister with 50% HLW loading holds 70% more HLW than standard poured canister with 25% waste loading (Savannah River Experience) and 14% more HLW than a standard pour canister with a 35% AVS canister loading
- Laboratory scale experiments indicate that 62% waste loading should be achievable-HLW loading advantage then increases to 214% and 144%
- AVS system also appears to have significant production advantages, including: reduced sensitivity to variations in feed, much shorter time at temperature, simpler operation, enhanced reliability, and potentially much less D&D

## Log of Tests Performed

Sample Number Waste		Chemical	PCT TCLP		Тg	Crystallinity	
	Loading	Analysis		•	_	X - ray	SEM/EDX
~	(wt %)					Diffraction	
Laboratory Scale							
R - 1	3 5	Х	Х	Х			
R - 2	4 5	Х	Х	Х			
R - 2 a	4 5	Х		Х			
R - 2 i	4 5						
R - 3	5 5	Х	Х	Х			
R 3 a	5 5						
R - 4	67.7	Х	Х	Х			
R - 5	3 5	Х	Х	Х			
R - 6	4 5	Х	Х	Х			
R - 7	5 5	Х	Х	Х			
R - 8	3 5	Х	Х	Х			
R - 9	4 5	Х	Х	Х			
R - 1 0	5 5	Х	Х	Х			
R - 10a	5 5						
R - 1 1	60	Х	Х	Х			
R - 1 2	4 5	Х	Х	Х			
R - 1 3	5 0	Х	Х	Х			
R - 1 4	6 2	Х	Х	Х			
TTT Samples	5 0		Х			Х	Х
"Blind" Sample	-	Х	Х	Х			
COFFEE-1	3 5	Х	Х	Х			
Bench Scale							
Shakedown	3 5	Х	Х	Х			
Run-1	3 5	Х	Х	Х			
R u n - 2	3 5	Х	Х	Х	Х	Х	Х
R u n - 3	5 0	Х	Х	Х	Х	Х	Х
R u n - 4	5 0	Х	Х	Х		Х	Х
R u n - 5	3 5	Х	Х				
R u n -6	5 0						
X = performed							

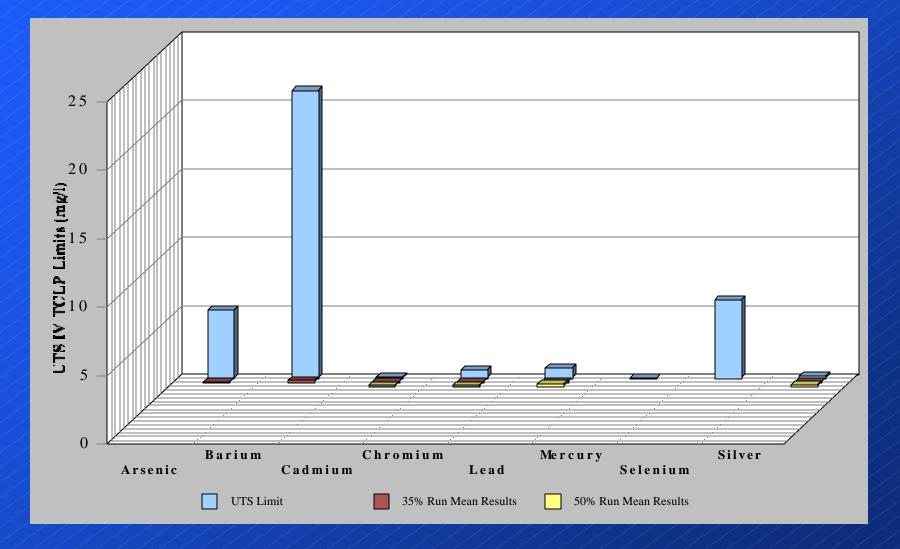
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#### PCT Results



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### TCLP Results



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### Continuous Feed Fill Experimental Equipment (COFFEE) Tests

#### Objectives

- Demonstrate the feasibility of continuously filling an AVS module at temperature with dry waste oxide
- Demonstrate that foaming is not an issue when feed is added at the fill temperature where the melt is fully fluid.

Method

- Small batches (55 g or 0.5") added to melt at refining temperature of 1350
  °C
- Observe melt surface between additions
- Continuous addition until a melt aspect ratio (h/d) of greater than 7.5 achieved
- Results
  - Uniform, acceptable glass
  - No foaming during additions
  - High fill rate ~ 5" liquid glass/hour demonstrated, 10" per hour appears possible
    - Equivalent to 18 (demonstrated) 36 hour fill for 4.5 m height AVS canister
  - High aspect ratio

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## Comparison of HLW Capacity in AVS Module to Conventional Canisters

- AVS Modules have Less Fill Volume than Conventional Pour Type Canisters
  - (V<sub>f</sub>)<sub>AVS</sub> » 0.8 (V<sub>f</sub>)<sub>Conv</sub>
- However, AVS Modules can hold more HLW than Conventional Canisters if:
  - $(\%WL)_{AVS}(\mathbf{r}_{G})_{AVS}(V_{f})_{AVS} > (\%WL)_{Conv}(\mathbf{r}_{G})_{Conv}(V_{f})_{Conv}$
- HLW Capacity of AVS Modules is Compared to that of Conventional Canisters for:
  - 25 and 35% Waste Loading in Conventional Canisters
  - 35, 50, and 62% Waste Loading in AVS Modules
- AVS Modules Can Hold Considerably More HLW than 25% Loaded Conventional Canisters.

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#### Comparison of Available Fill Volumes and Overall Weights for AVS Module and Stainless Steel Pour Canisters

Parameter	Stainless Steel Pour Canister	AVS Module
Weight of HLW Inside Canister or		
Module, kg		
25% Waste Loading	730	
35% Waste Loading	1088	831
50% Waste Loading		1242
62% Waste Loading		1566
Ratio of HLW Inside AVS		
Module to Stainless Canister with		
25% Waste Load		
35% Waste Loading in AVS		1.14
50% Waste Loading in AVS		1.70
62% Waste Loading in AVS		2.14
Ratio of HLW Inside AVS		
Module to Stainless Canister with		
35% Waste Load		
35% Waste Loading in AVS		0.76
50% Waste Loading in AVS		1.14
62% Waste Loading in AVS		1.44

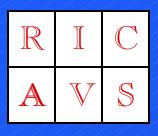


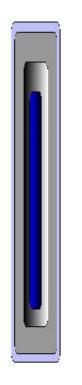
#### Weight of AVS Module

- AVS Module Weight of Components
  - 702 kg Stainless Steel Container
  - 17 kg Insulation
  - 277 kg Graphite Susceptor
  - 148 kg Alumina Crucible
  - 1144 kg Empty Weight
- Module Filled to 90% Level with 50% HLW Glass
  - 2484 kg of Glass
- Total Module Weight is 3628 kg
  - Within WAPS Limit of 4200 kg

## Summary and Conclusions

- AVS Module Uses Well Known Commercial Materials
  - Stainless Steel Canister
  - Graphite and Alumina Crucibles
  - Graphite Fiber Insulation
- Dewatered HLW/Frit Mixture Extruded Into Inductively Heated Hot Graphite Crucibles Inside Module
- ~3 Days to Process a Module (Includes Cooling)
- Modest Electrical Requirements for AVS Process
  - 30 Hz AC Frequency
  - 150 kW(e) Average Power for Module
  - ~\$1000/Module Energy Cost (10°/kw-hr)
- AVS Product Glass has:
  - High HLW Loading (50% or More Compared to 25% for Conventional Melter)
  - Capability to Handle Wide Range of HLW Composition (High Bi, Cr, Zr, etc.)
  - Excellent Durability and Low Crystallinity. PCT Results Very Favorable.
- One AVS Module Can Dispose of Much More HLW Than One Canister from a Conventional Melter.





#### Thank You

RIC LLC Argus Remediation, Inc Burns and Roe Enterprises, Inc. UCAR Carbon Company, Inc.

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