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Development of Alternative Glass Formulations for Vitrification of Excess Plutonium - SEM/XRD Analyses

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Abstract:

The Department of Energy Office of Environmental Management (DOE/ EM) plans to conduct the Plutonium Disposition Project at the Savannah River Site (SRS) to disposition excess weapons-usable plutonium. A plutonium glass waste form is the leading candidate for immobilization of the plutonium for subsequent disposition in a geologic repository. A reference lanthanide borosilicate (LaBS) glass was developed and durability tested to provide data to support the Yucca Mountain License Application process. In addition to the reference LaBS glass, alternative frit formulations have also been recently developed. This poster will present SEM/ EDS and XRD data gathered at SRNL in these recent studies on surrogate glasses and actual plutonium-containing glasses. SEM/EDS applications were used to investigate various plutonium and hafnium oxide crystals observed in the glasses melted at 1500 °C and in heat-treated glasses. XRD data was collected on these same powdered glass samples to confirm crystalline phase identification.

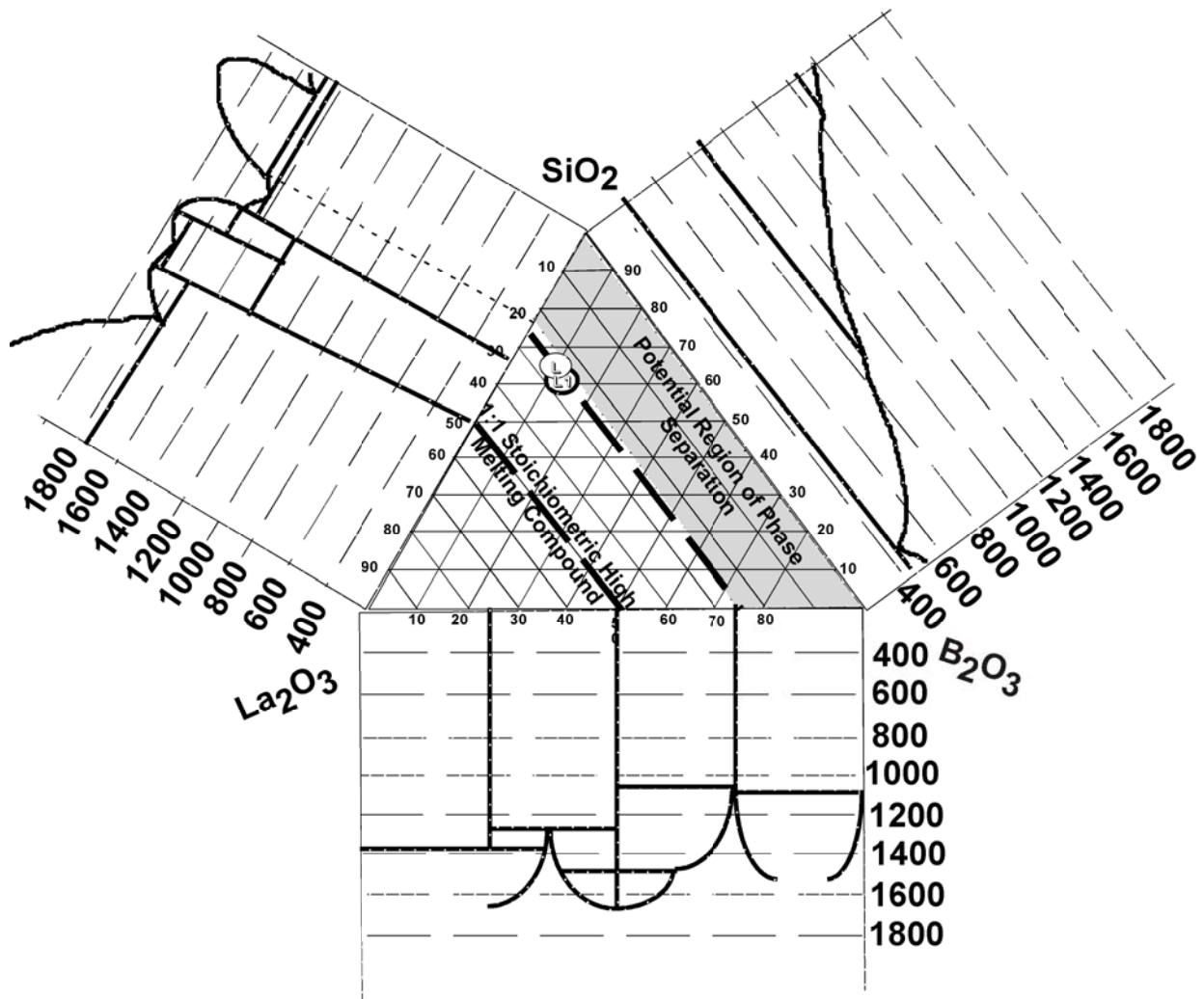
LaBS Glass Compositions

Oxide	LaBS Frit A PuO₂	LaBS Frit B PuO₂	LaBS Frit X PuO₂
Al₂O₃	19.46	19.27	9.05
B₂O₃	10.59	10.50	11.77
Gd₂O₃	7.78	11.58	12.22
HfO₂ (frit component)	-	5.97	6.34
La₂O₃	11.22	7.33	17.20
Nd₂O₃	11.58	7.42	13.58
PuO₂ (Pu₂O₃)	9.50†	9.50	9.50
SiO₂	26.43	26.15	18.10
SrO (CaO+ZnO)	2.26	2.26	2.26
ZrO₂ (frit component)	1.18	-	-
Melt Temp (° C)	1500	1500	1500
Total Ln₂O₃	30.58	26.33	43.00
Ln₂O₃+(Th,Zr,Hf)O₂	40.08	41.80	58.84
SiO₂+Al₂O₃	45.89	45.42	27.15
Reference	WSRC- TR-96- 0322	WSRC- RP-97- 00902	Recent Studies
SUM	100	99.98	100.02

**Ternary Oxide System $\text{La}_2\text{O}_3\text{-B}_2\text{O}_3\text{-(SiO}_2\text{+Al}_2\text{O}_3)$
 Generated from the Binary Oxide Systems $\text{La}_2\text{O}_3\text{-SiO}_2$,
 $\text{Sm}_2\text{O}_3\text{-SiO}_2$, $\text{La}_2\text{O}_3\text{-B}_2\text{O}_3$, $\text{B}_2\text{O}_3\text{-SiO}_2$**

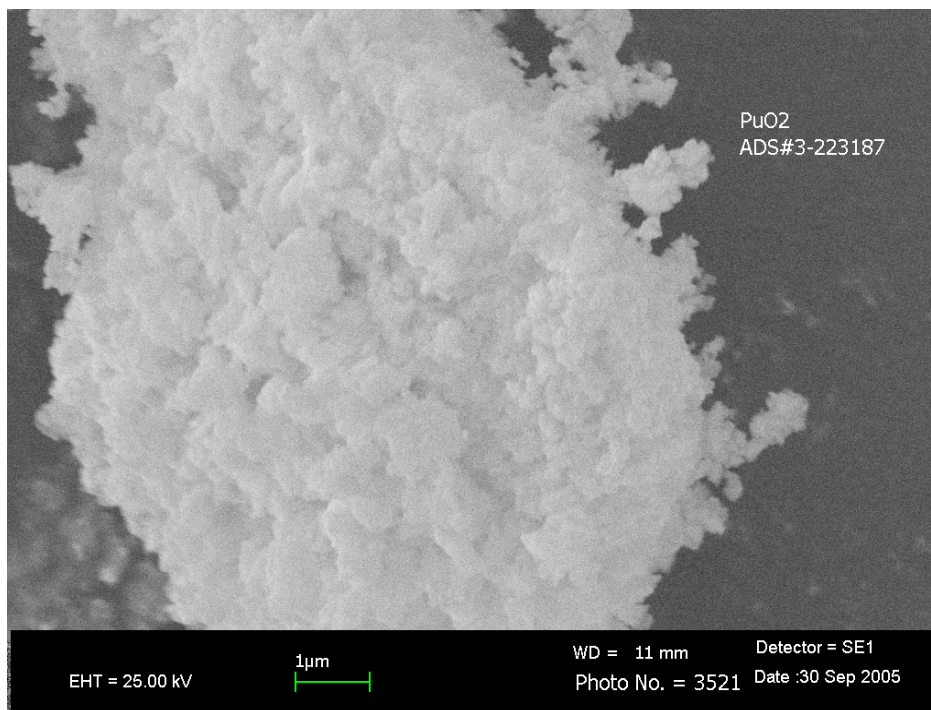
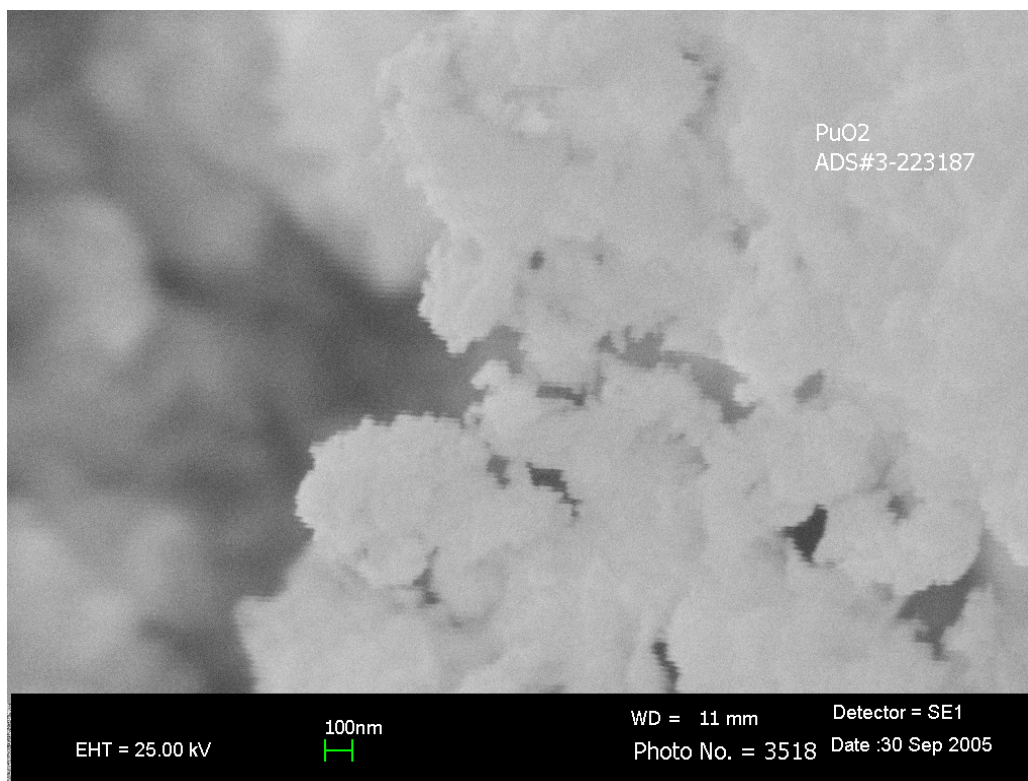
Phase Diagrams for Ceramists, Volume I and II, Amer. Ceramic Society, Westerville, Ohio,

Glass Formulation Team: C. M Jantzen (lead), J. C. Marra, D. K. Peeler

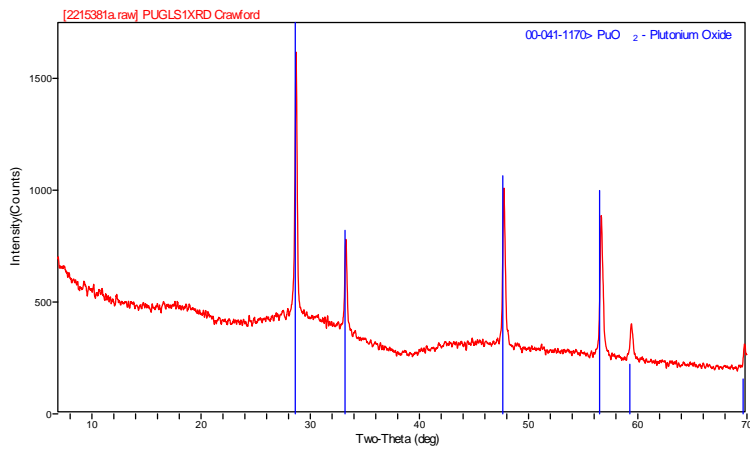
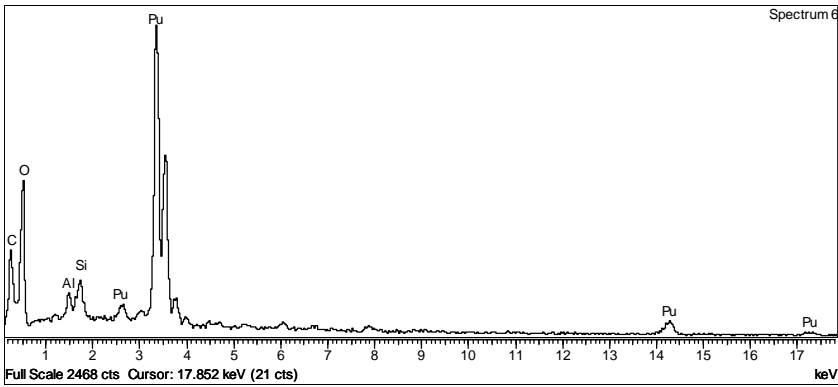
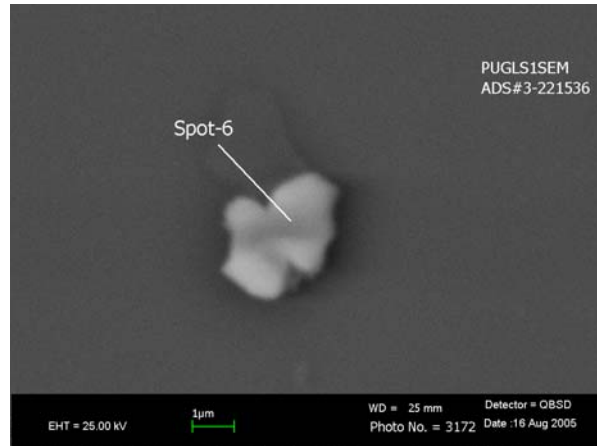


Photographs of Pu LaBS Frit B Glass Used for Performance Testing. Glass Fabrication Performed in SRNL Shielded Cells Facility via Remote Handling



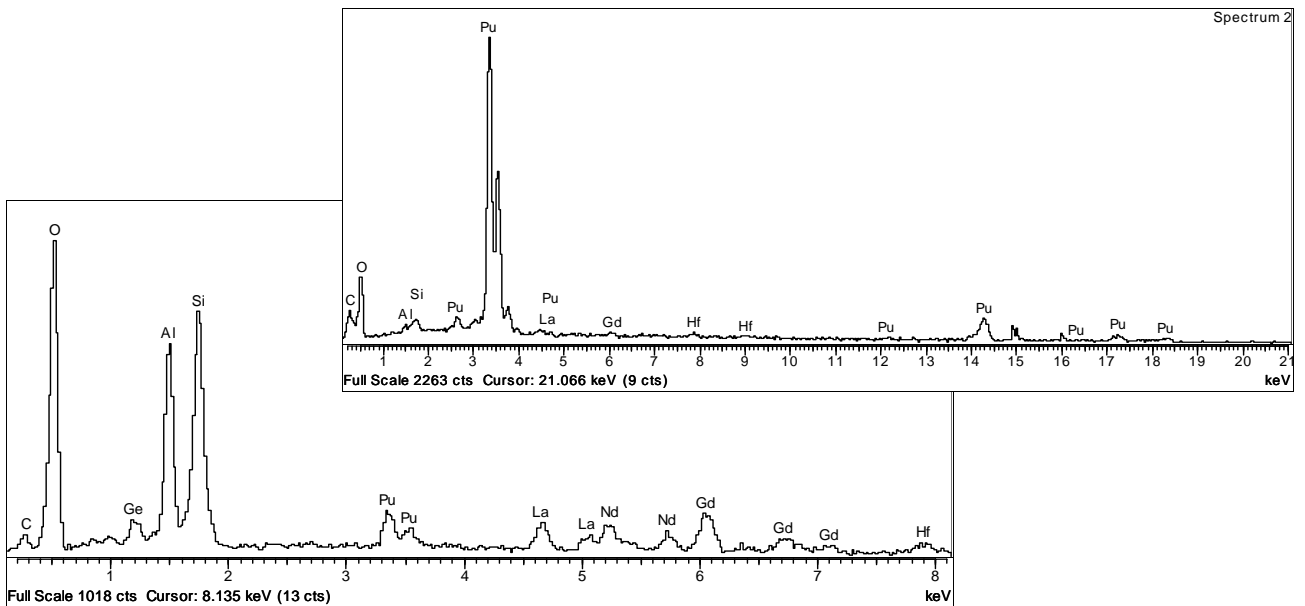
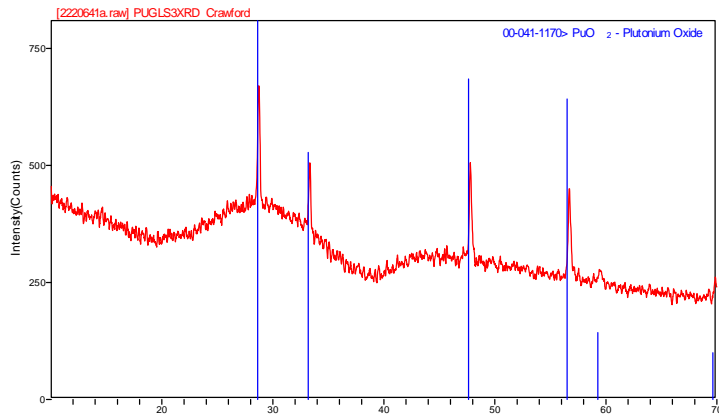
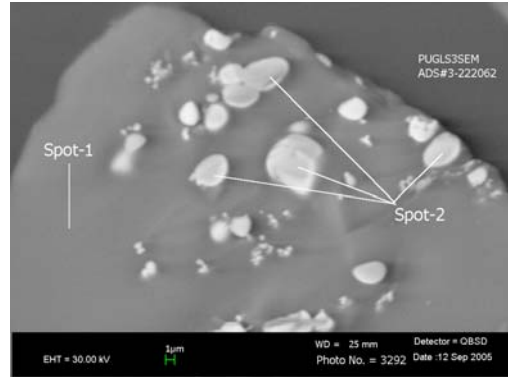
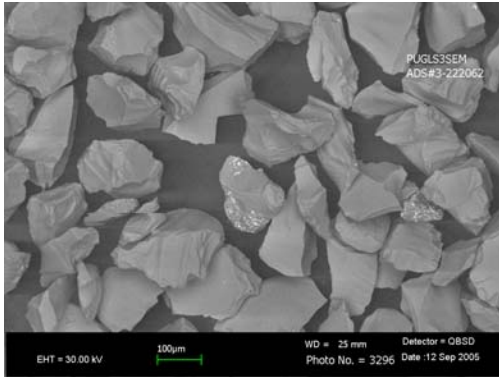


SEM Micrographs of PuO₂ Source Material Showing the Fine Crystallite Size of the Starting Material

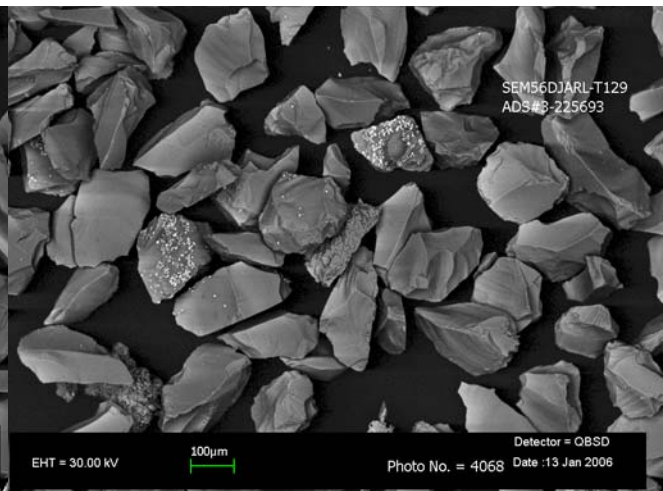
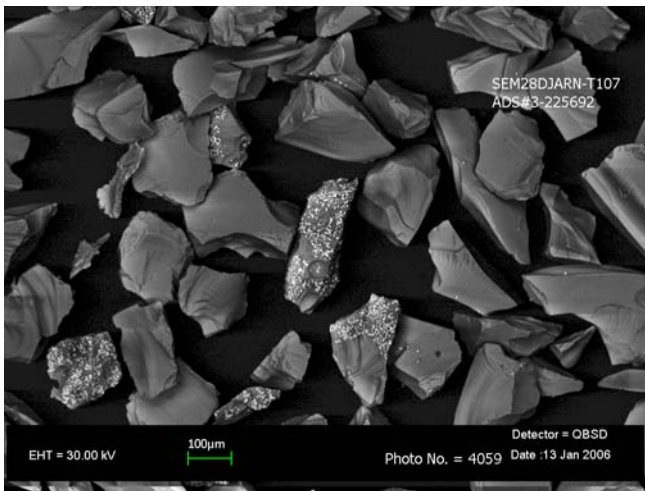
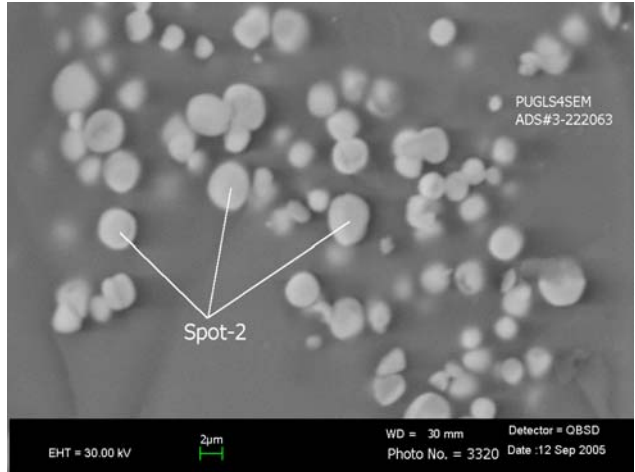
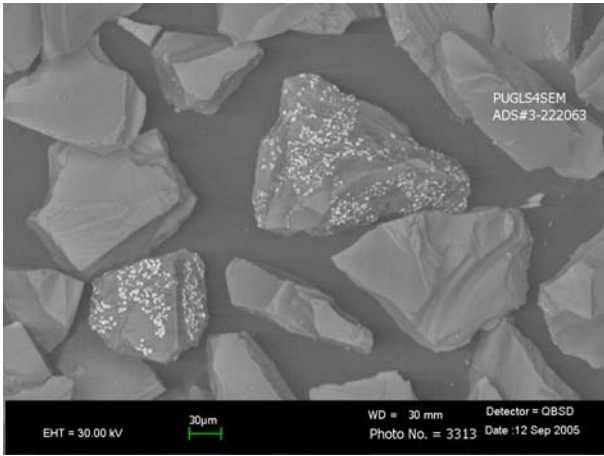
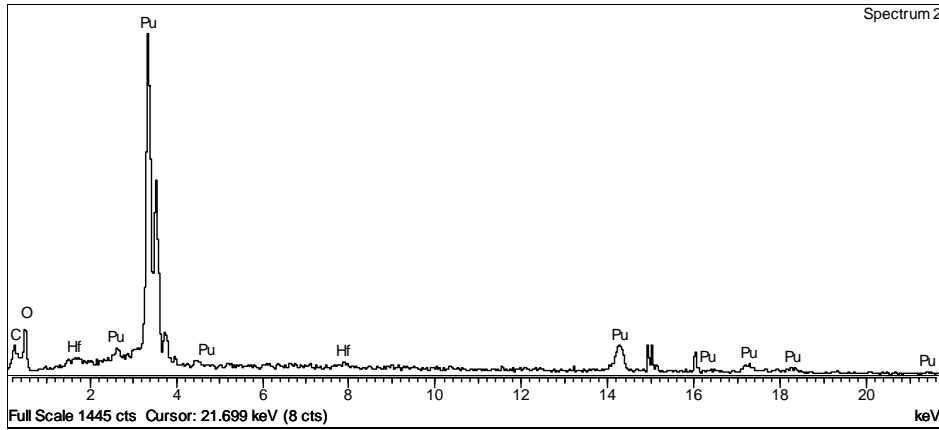


Original Data for Frit B Pu Glass Shards Indicate PuO₂ Crystalline Phase

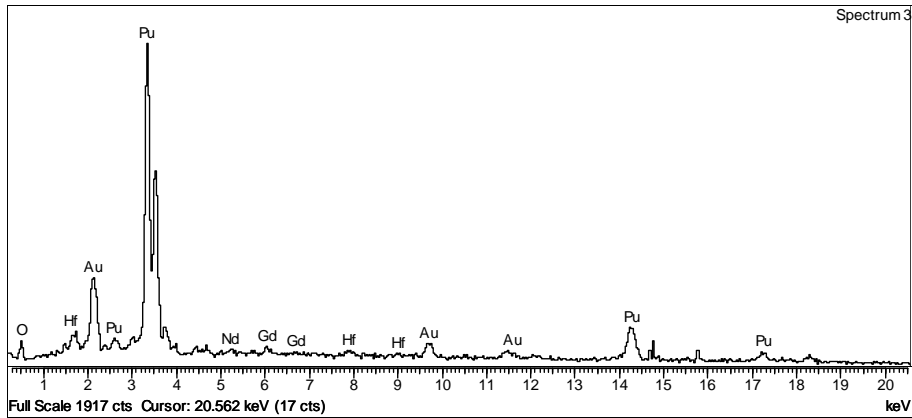
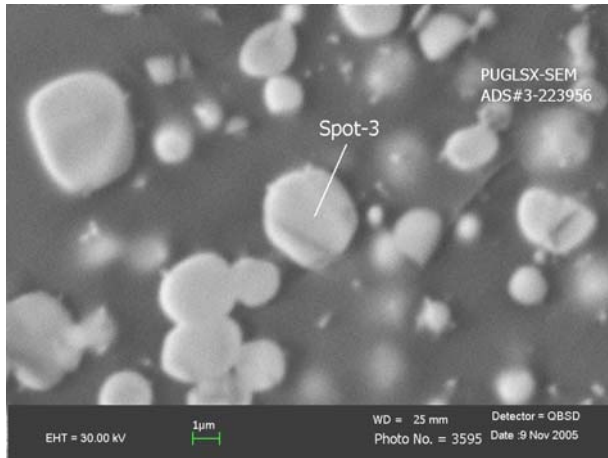
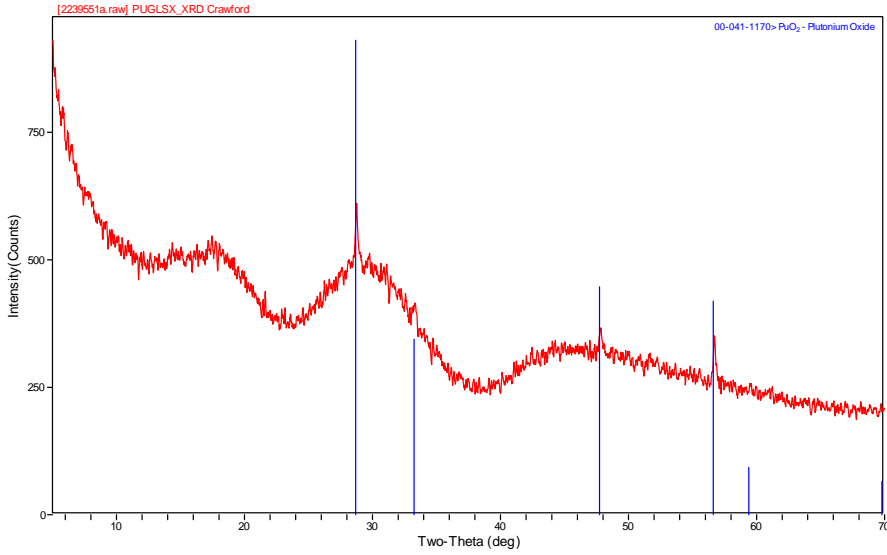
Ground, Washed Frit B Pu-Glass Shows PuO₂ Before Leaching Tests



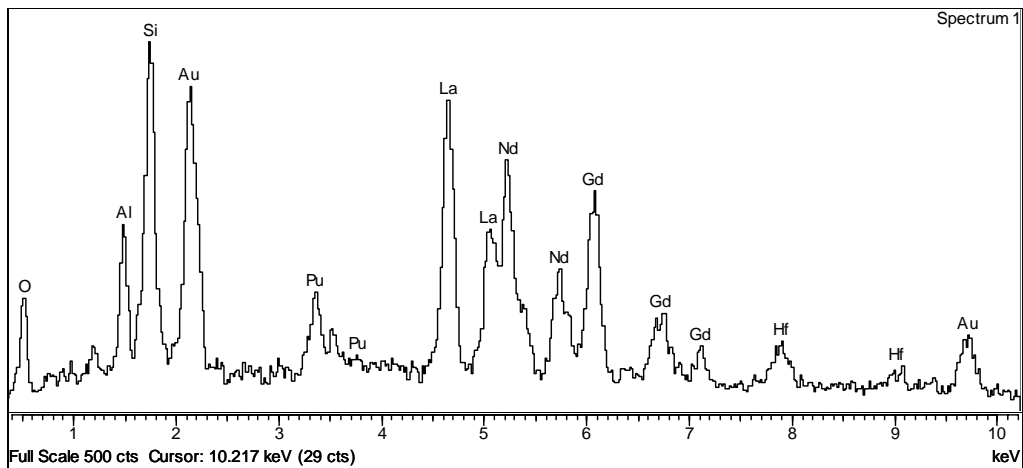
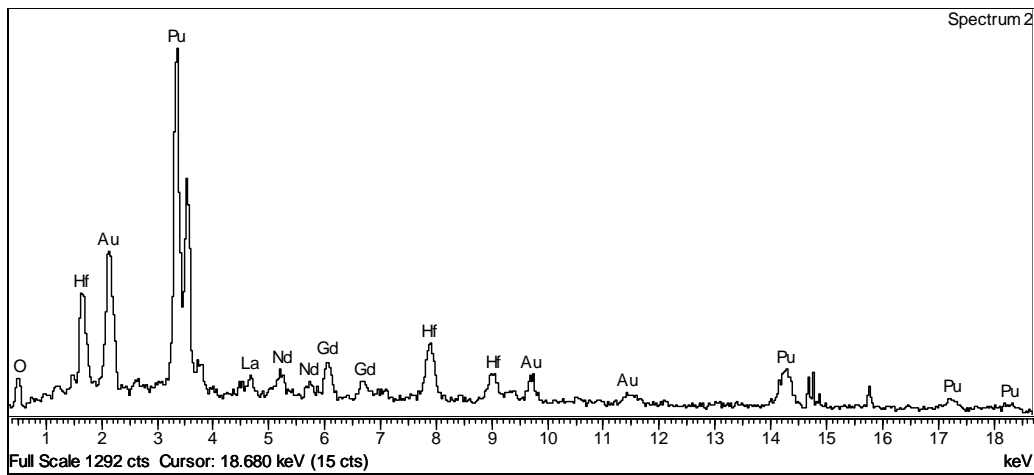
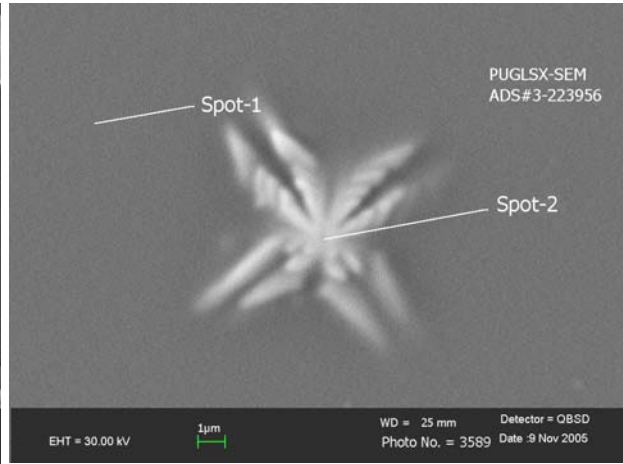
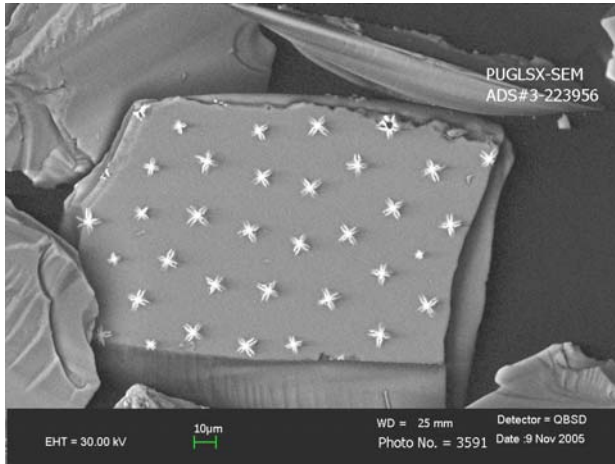
Leached Glass at 7-day, 28-day and 56-day; No Alteration Phases Observed



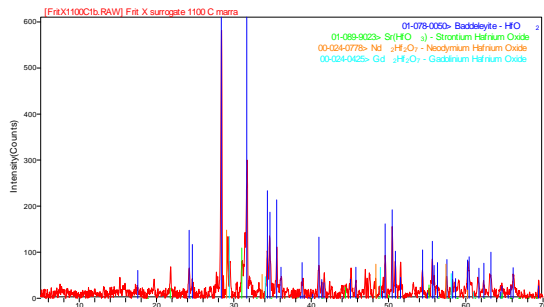
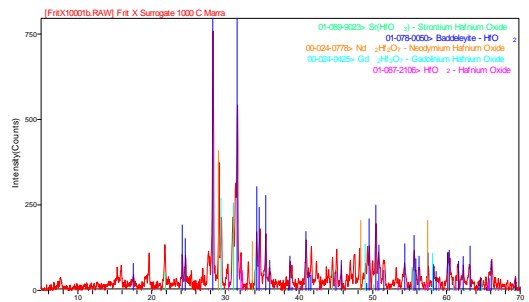
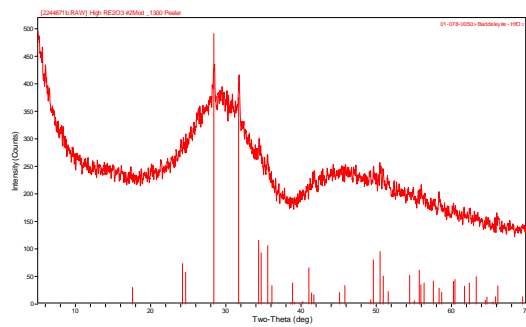
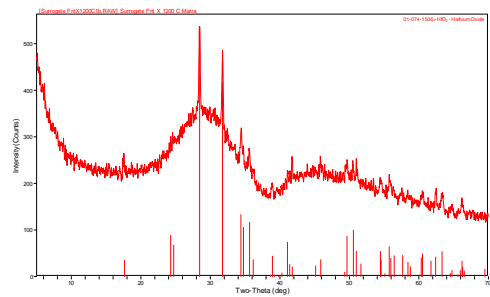
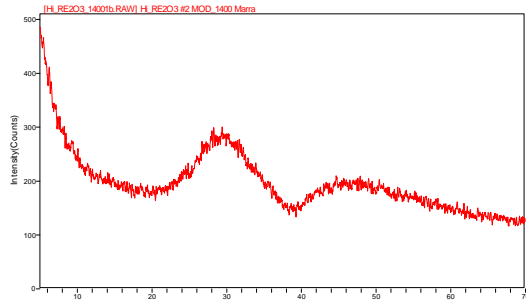
Ground, Washed Frit X Pu-Glass



Ground, Washed Frit X Pu-Glass – Presence of HfO₂/PuO₂ Solid Solution Phase ?



Frit X Surrogate Glass Heat Treatment: 1400°C – No Crystalline Phases; 1300/1200°C Observe HfO₂ Phase; 1100/1000 °C Observe Multiple Hafnium Crystalline Phases and Mixed Rare Earth Oxide Phases



Frit X Pu-Glass Heat Treatment at 1200°C
Spot 5: Mixed Rare Earth Oxide Phases
Spot 6: Amorphous Glass Phase
Spot 7: Some Hf Present in Crystals; Reduced Beam Current (12 keV) Indicates PuO₂/HfO₂ in the Crystal

