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ROSATOM State Corporation



Leaching of plutonium from highly radioactive samples of ceramics, glass and single crystals

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Overview of Russian waste management strategy

and the second



Current state*

Legacy waste

~ 490 million m³ of liquid waste (4,27·10¹⁹ Bq)
~ 90 million tons of solid waste (3,59·10¹⁹ Bq)



Commercial waste after SNF reprocessing



* Official Rosatom information for 2011

Russian waste management: the future

- Closed fuel cycle
 - Experimental facility at Krasnoyarsk region: only HLW and LLW?
 - Using U and Pu after reprocessing in a fresh fuel fabrication: MOX- and REMIX-fuel
- Geological disposal
 - Decision | Granite rock in Krasnoyarsk region
 - Underground facility | Probably at 2060
 - Final waste form for HLW?

Π

Ceramic

Approach to study

- Using accelerated radiation damage activity of ²³⁸Pu ≈ 280 * activity of ²³⁹Pu
- Long-term study of samples depending on accumulated doses
 - The "oldest" samples was synthesized in 2000

- At the moment cumulated dose is more than 10²⁶ alpha-decays/m³

Standard static leach test at 25°C and 90°C according MCC-1

Testing of ceramic based on cubic zirconia and monazite

 Cubic zirconia (Zr,Y,REE,...)O₂ and monazite (REE,...)PO₄ are well known durable host-phases for actinides immobilization

Demonstrate high resistance to self-irradiation and chemical durability in water but behavior under long-term radiation damage is still unclear!

Samples

Cubic zirconia

- ²³⁸Pu-doped Zr_{0.79}Gd_{0.14}Pu_{0.04}O_{1.93} at different accumulated doses (163 and 170 alpha-decays/m³ x 10²⁴) Contains 9.9 wt.% of ²³⁸Pu. Synthesized in 2000.
- ²³⁹Pu-doped Zr_{0.75}Gd_{0.19}Pu_{0.06}O_{1.91} at accumulated dose
 0.6 alpha-decays/m³ x 10²⁴
 Contains 9.9 wt.% of ²³⁹Pu. Synthesized in 2000.

Monazite

²³⁸Pu-doped La_{0.9}Pu_{0.1}PO₄ at different accumulated doses (1 and 100 alpha decays/m³ x 10²⁴)
 Contains 8.1 wt.% of ²³⁸Pu. Synthesized in 2002.

Sample	Cumulative dose, α-decay/m³ x 10 ²⁴	NL, g/m², 28 days, 90 °C				
Cubic zirconia $Zr_{0.79}Gd_{0.14}Pu_{0.04}O_{1.93}$ doped with 9.9 wt.% ²³⁸ Pu						
	1	0.04				
Previous data	6	0.4				
Flevious data	8	0.4				
	13	0.2				
Pellet #1	163	0.3				
Pellet #2	170	0.7				
Cubic zirconia Zr _{0.75} Gd _{0.19} Pu _{0.06} O _{1.91} doped with 9.9 wt.% ²³⁹ Pu						
Previous data	insignificant	0.04				
Pellet #1	0.6	0.1				
Pellet #2	0.6	0.1				
Monazite $La_{0.9}Pu_{0.1}PO_4$ doped with 8.1 wt.% ²³⁸ Pu						
Pellet #1	1	0.42				
Pellet #2	100	1.6				

Normalized Pu mass loss from samples of single-phase Pu-doped ceramics after static leach tests in distilled water. Correction for ceramic porosity was ignored.

Decrease of monazite chemical durability

what's going on?



partial destruction of solid solution with partial or release of Pu into separate phases mechanical destruction of ceramic matrix and increase of leaching surface

What about plutonium dioxide itself?

Static leach test for ²³⁸Pu-doped PuO₂



Polycrystalline PuO₂ doped with 11 wt.% ²³⁸Pu



Single crystals of PuO₂ doped with 1 wt.% ²³⁸Pu



Results of PuO_2 static leach tests: \bigcirc and \bigcirc – polycrystalline samples (the same pellet was used in different tests); \square – single crystals (only separate fresh samples for each test)

III

Glass

²³⁸Pu-doped borosilicate glass

• We are in a starting point

		Wt.%	Mol.%
A MAR AN AL	SiO ₂	47.7	60.04
	Na ₂ O	14.5	13.09
	B ₂ O ₃	21.1	16.70
	Al_2O_3	6.8	3.61
	Eu ₂ O ₃	3.0	0.45
25 mm	CaO	5.8	5.87
	PuO ₂	1.06	0.23

- The same result for 7 and 14 days \rightarrow saturation?
- Testing to be continued...

Chernobyl "lava" – artificial glass-like material

- More than 30 years old
- Can be considered as a vitrified HLW analogue
- Self-destruction caused by radiation damage has been observed at 2011



Samples used for static leach test at 25°C for 7, 14, 28, 56 and 270 days

"lava" type	days	NL, g/m ² (25°C, distilled water)			
		²³⁹ Pu + ²⁴⁰ Pu	²⁴¹ Am + ²³⁸ Pu	¹³⁷ Cs	¹⁵⁴ Eu
Black	7	0.3	0.4	<10-4	<10 ⁻³
	14	3.1	9.4	5.9·10 ⁻³	1.5.10-2
	28	0.2	0.7	8.0.10-4	
	56	1.4	2.0	<10-4	<10-3
	270	0.7	1.2	5.0·10 ⁻²	
Brown	7	0.1	0.2	2.8·10 ⁻³	<10-3
	14	1.9	4.4	1.2.10-2	
	28	0.04	0.14	5.6.10-3	1.2.10-2
	56	1.3	1.2	<10-4	<10- ³
	270	1.4	1.8	0.2	< 10 s

Normalized mass loss of actinides and fission products from static leach test in distilled water at 25°C.

Conclusions

- Normalized Pu mass loss from cubic zirconia ceramic doped with ²³⁸Pu and ²³⁹Pu is similar at extremely different doses of selfirradiation.
- Although ²³⁸Pu-doped monazite, La_{0.9}Pu_{0.1}PO₄, remains crystalline at very high dose of self-irradiation the Pu normalized mass loss from its matrix increases depending on accumulative dose.
- Pu leaching from ²³⁹PuO₂ and ²³⁸⁻²³⁹PuO₂ in distilled water can be comparable at least with some ²³⁹Pu-and ²³⁸Pu-doped ceramic waste forms leached under the same conditions. Therefore, development of more feasible leach tests for Pu-doped ceramics would be useful.
- The use of unique KRI collection of Chernobyl "lava" allows obtaining important data for modeling chemical durability of vitrified HLW. Study of Chernobyl "lava" may be applied to predict some features of Fukushima's corium melt.

Thank you for attention!

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