Development of a fecal method for actinides

Alexandre Gagné, Xiongxin Dai, Sheila Kramer-Tremblay, Joel Surette, Candice Didychuk and Dominic Larivière*

Atomic Energy of Canada Limited

*Laval University

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Plan

- Introduction
  - Context
  - Objectives
- Analytical development
  - Strategy
  - Fusion
  - Flocculation
  - Coprecipitation
  - Actinide separation
  - Results
- Conclusions
  - Acknowledgements

Context

Refurbishment and refection of aging nuclear reactor may results in internal contamination by α emitters (actinides) without proper detection by routine dosimetric procedure.

Context

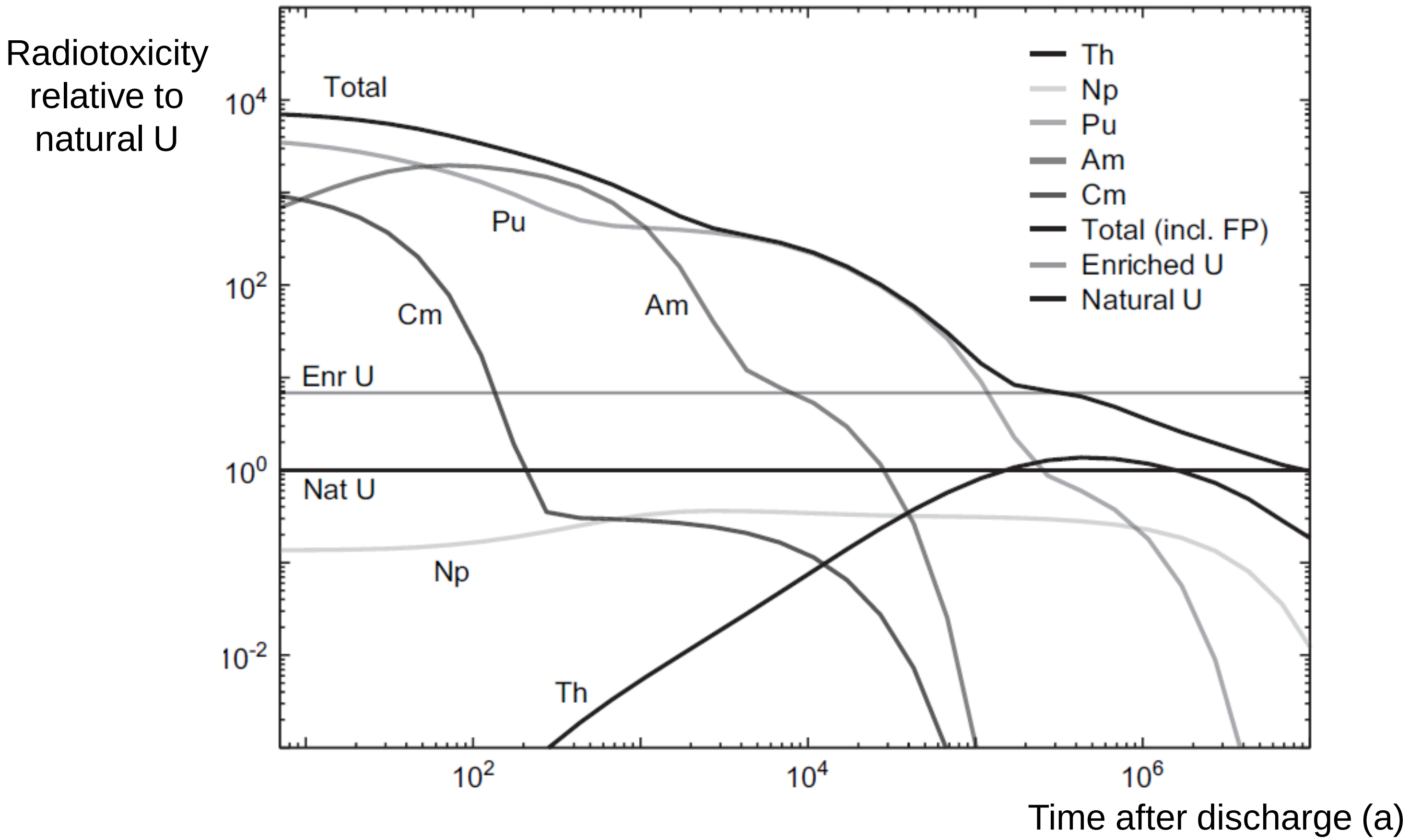


Figure 1 : Evolution of the radiotoxicity from actinides in spent fuel from a 41 GWd/tHM (UOX) reactor [1]

[1] Westlén D., *Prog. Nucl. Energ.*, 2007; 49: 597-605.

Objectives

To develop an analytical procedure for complete digestion of fecal sample followed by the separation and the analysis of occurring actinides at low level.

Factors to consider:

- Matrix composition
  - **Silica content**
- Rapidness
- Efficiency and reliability (ANSI N13.30)
- Conviviality

Strategy

Urine = soluble actinides

Fecal mater = refractory actinides

Sample size [2]

150 g / day for adult male

120 g / day for adult female

Diet

- Organic content
- Daily intake of Si : 20 - 63 mg [3-4]
- Presence of phosphates, sulfates and sulfides

[2] Valentin J., ICRP Publication 89, 2003; 129. [3] Jugdaohsingh R., Anderson S.H.C. , Tucker K.L., Elliott H., Kiel D.P., Thompson R.P.H. and Powell J.J., *Am. J. Clin. Nutr.*, 2002; 75: 887-893. [4] European Food Safety Authority (EFSA), *The EFSA Journal*, 2009; 1132: 1-24.

Strategy

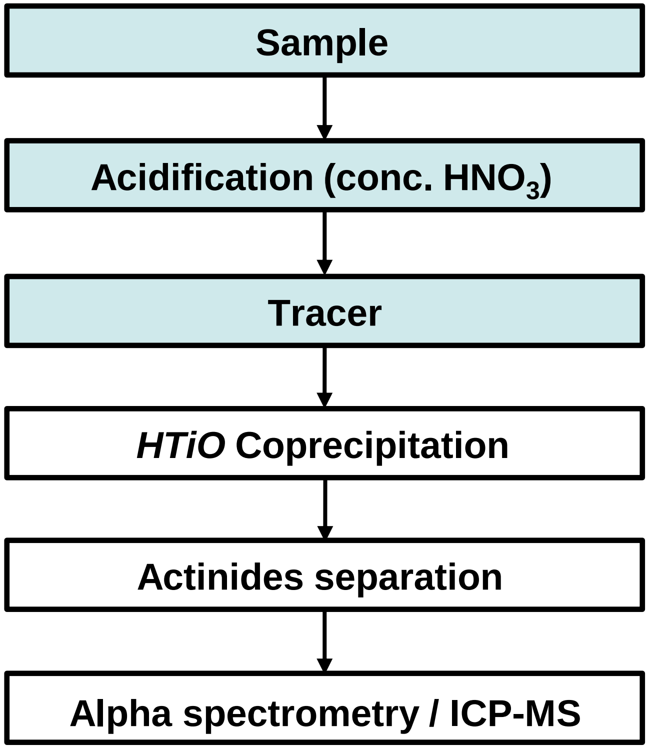


Figure 2 : Diagram of the urine methodology [5]

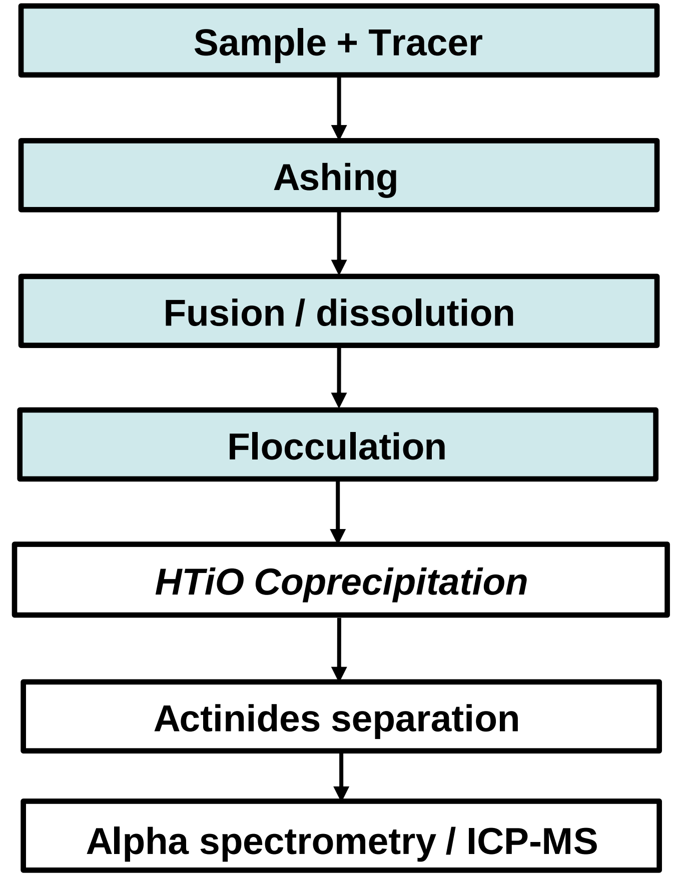


Figure 3 : Diagram of the proposed fecal methodology

[5] Dai X. and Kramer-Tremblay S., *Health Phys.*, 2011; 101: 144-147.

Strategy

Table 1 : Composition of synthetic feces [6]

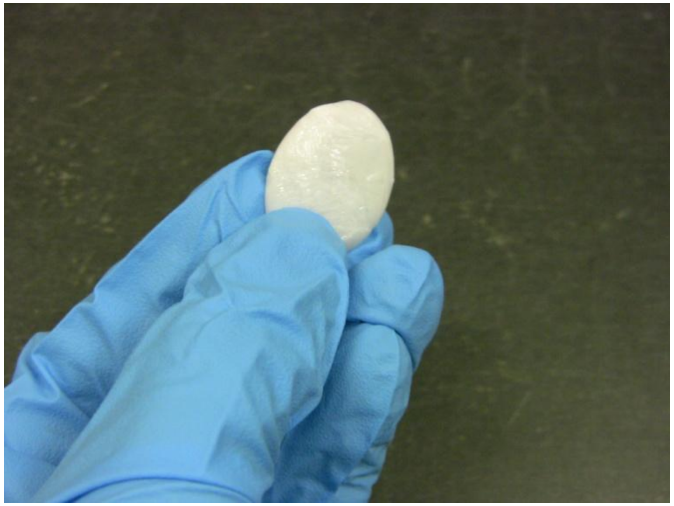
Reagents	Mass (g)
Ammonium dihydrogenophosphate	2.10
Calcium nitrate	0.97
Potassium carbonate	0.83
Magnesium carbonate	0.61
Sodium sulfate	0.37
Silica	**0.15**
Ammonium chloride	0.04
Ferric ammonium sulfate	0.04
Stannous chloride	0.03
Zinc sulfide	0.01
Organic compounds	**31.50**
Water	**63.35**
Total	**100.00**

Organic compounds	Mass (g)
Leucine	7.10
Lysine	5.10
Gelatin	5.00
Cellulose	4.00
Palmitic acid	3.00
Threonine	2.00
Stearic acid	2.00
Methionine	0.80
Peanut oil	1.50
Oleic acid	1.00

[6] Payne G.F., Bores N., Melton K.K. and Rankin J.M., U.S. Bioassay Intercomparison Studies Program at Oak Ridge National Lab, U.S. Department of Energy, 1998; 8.

Fusion

Fusion is performed in a graphite crucible at 1000°C with an electrical furnace for 30 minutes.

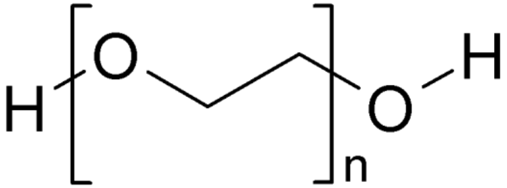


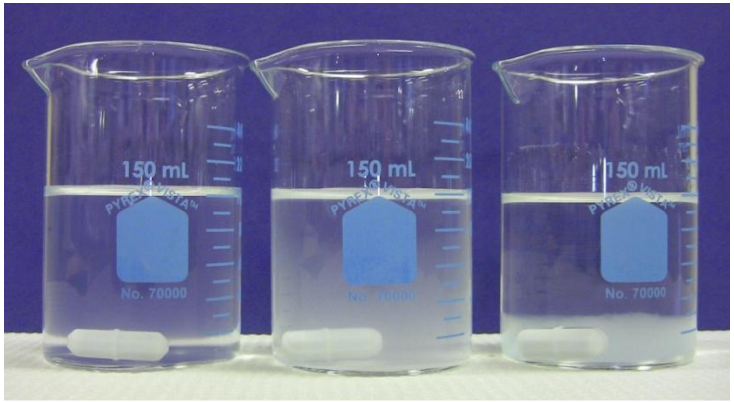
Fusion glass → (30mL *aqua regia*; Reflux for 1 h) → (Dil. to 90mL) → **Dissolved fusion glass** (~ 3-4M H^+)

Flocculation

Table 2 : Elimination (%) of silica by different PEG via flocculation

PEG	Elimination
1500	93 ± 2
2000	93 ± 3
6000	93 ± 2
35000	93 ± 2





Coprecipitation

Dissolved sample $\xrightarrow[\text{pH 9, } NH_4OH]{\text{1mL } TiOCl_2 \text{ (7\%)}}$ **↓ HTiO** [Actinides]

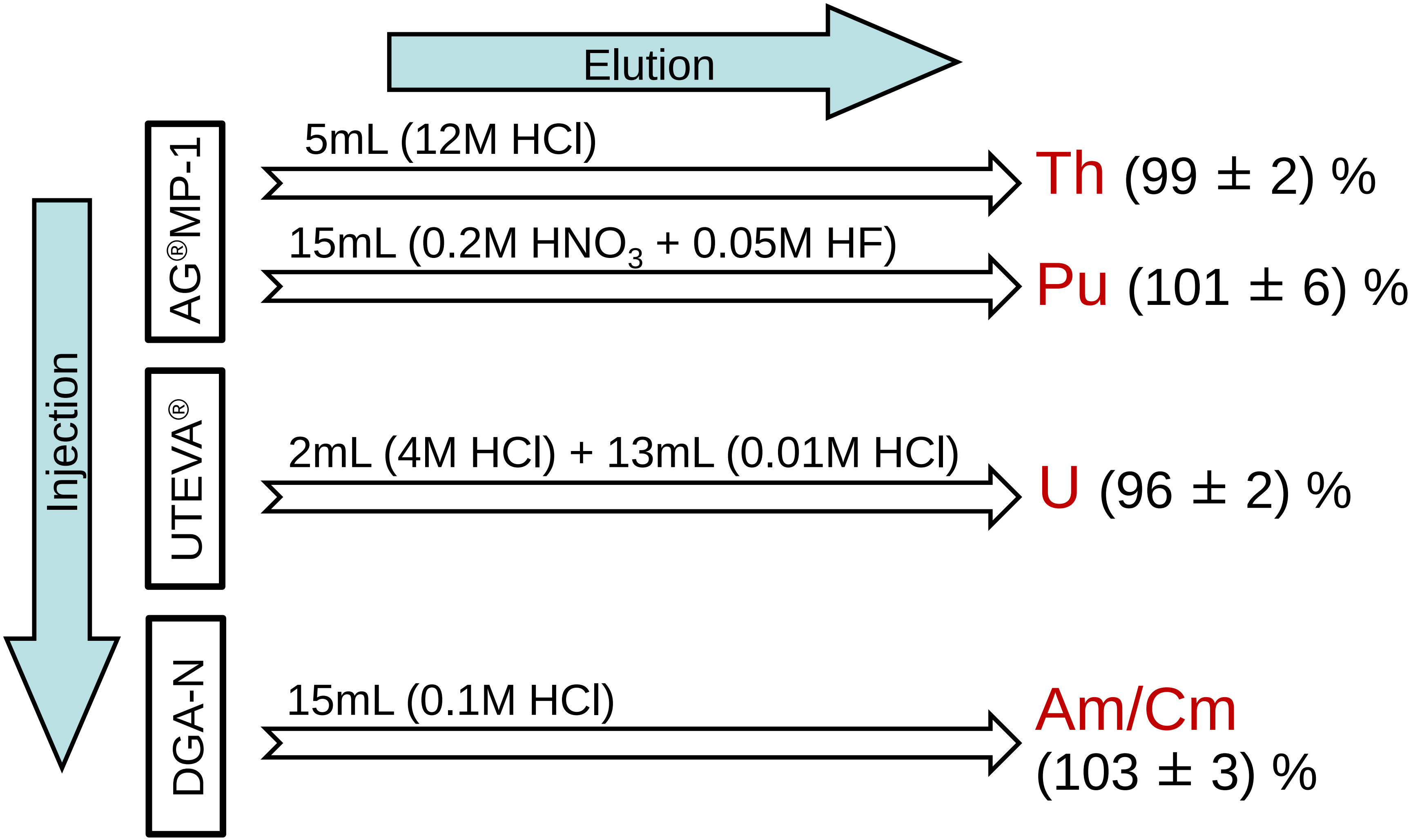
HTiO [Actinides] $\xrightarrow[0.03M\ NaNO_2]{8M\ HNO_3,\ 0.3M\ H_2O_2}$ **$[Ti(O_2)OH]^+NO_3^-$ + Dissolved actinides**

HTiO - Advantages:

- Polyvalent (*e.g.* phosphates, sulfates)
- Negligible retention on AG® MP-1M [7], UTEVA® [8] and DGA-N [9].

[7] Strelow F.W.E., *Anal. Chim. Acta*, 1981; 127: 63-70. [8] Horwitz E.P., Dietz M.L., Renato C., Diamond H., Essling A.M. and Graczyk D., *Anal. Chim. Acta*, 1992; 266, 25-37.[9] Horwitz E.P., McAlister D.R., Bond H.A. and Barrans R.E.Jr., *Solvent Extr. Ion Exch.*, 2005; 23: 319-344.

Actinide separation



Results

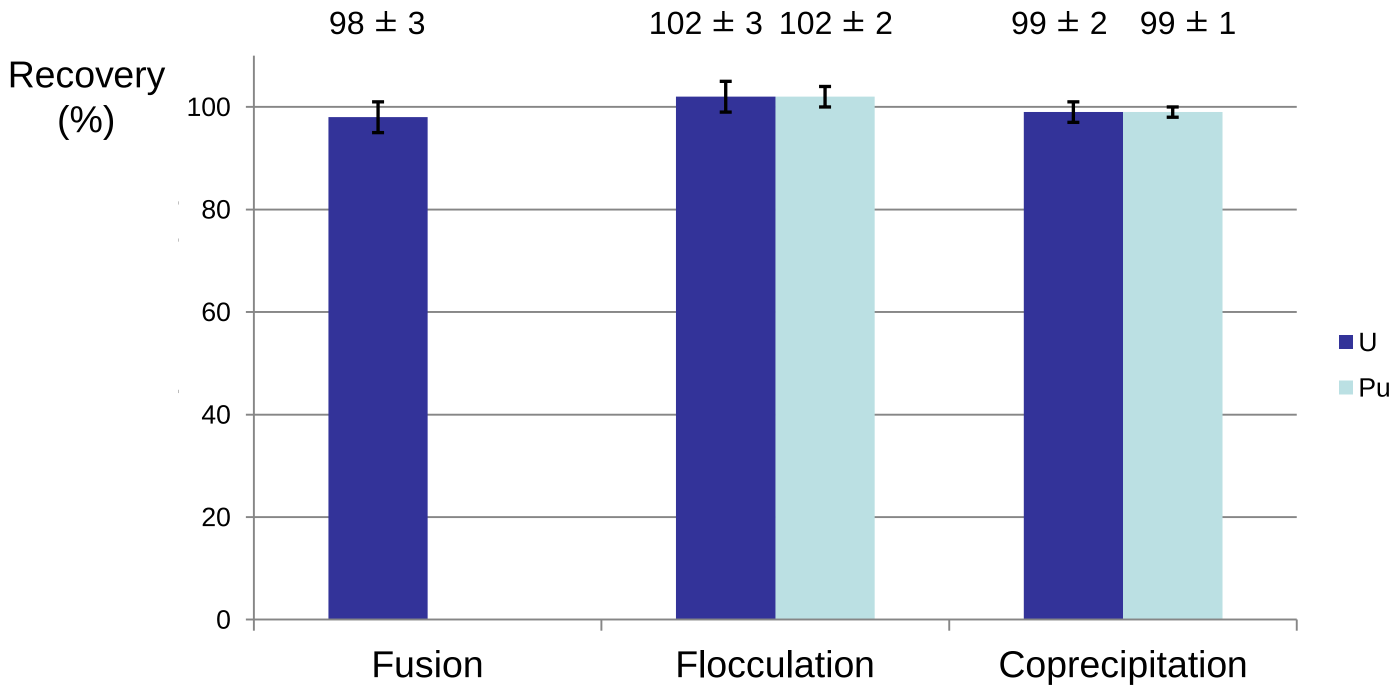


Figure 4 : Recovery of U and Pu for various pre-separation steps

Results

Table 3 : Minimum Detectable Activity (MDA) of the developed Bioassay by alpha spectrometry

Actinide	Mean MDA (mBq)
^{241}Am	**0.44 ± 0.17**
^{242}Cm	**0.16 ± 0.12**
$^{243/244}Cm$	**0.38 ± 0.13**
^{238}Pu	**0.62 ± 0.22**
$^{239/240}Pu$	**0.44 ± 0.29**

Conclusions

- Borate fusion completely decompose and dissolve the artificial fecal ashes (successful on human feces).
- Flocculation and coprecipitation are fast and efficient for fecal bioassay.
- The overall chemical recoveries for the actinides of interest (Th, U, Pu, Am and Cm) are acceptable (> 70 %).
- Developed method is valid by ANSI N13.30 criteria.

Acknowledgements

Research Team

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