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The impact of transuranics and key fission products on emissions limits & waste declarations

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The impact of transuranics and key fission products on emissions limits & waste declarations

OVERVIEW

Introduction

Estimation of higher actinides in Pu-containing fuels with burn-up

Estimation of gaseous (C-14) source term in non-oxide fuels

Estimation of the radium/radon equilibrium in contaminated cells

Practical application of the results

Conclusions and outlook for the future

The impact of transuranics and key fission products on emissions limits & waste declarations

Introduction

With ageing hot cells, waste and its disposal is becoming a greater proportion of the hot cell work as renovation is a continuous part of the work.

Meanwhile the control of such waste is becoming ever more careful and exact. Safety & Licensing Authorities are requiring more charactorisation & declaration of the individual waste barrels and a more careful monitoring of the emissions from hot cells (& other laboratories).

There are problems with declarations of higher actinides to compare with total neutron counting performed as checks at the storage facility.

There are concerns of potential emissions peaks during decontamination or other activities

A review of the current emissions limits is currently underway with the regulator‘s agent (TÜV).

The impact of transuranics and key fission products on emissions limits & waste declarations

Specific Problems in this presentation:

There are problems with declarations of higher actinides to compare with total neutron counting performed as checks at the storage facility.

There are concerns of potential emissions peaks during decontamination or other activities

A review of the current emissions limits is currently underway with the regulator‘s agent (TÜV) therefore estimates of certain gaseous inventories need to be made.

The impact of transuranics and key fission products on emissions limits & waste declarations

Current Emission Limits for ITU

This is based on the estimation of the most active fission products based on a modelling of a UO_2 commercial fuel with 30GWd/tU burn-up and a cooling time of 3 years. This was modelled using KORIGEN code.

Although they are all β-active nuclides, 80% are also important γ-emitters (Cs-134/137 and includes an α-emitter (Pu-241)- 10%. Assumes some aerosol escapes (hence solid fission products included)

Kr-85, Rn-222 & C-14 emissions are separately considered

Beta radionuclide source term (mix no. 6)	
Cs-137	20%
Cs-134	18%
Ce-144	14,50%
Ru-106	15%
Sr-90	10%
Pu-241	10%
Pm-147	9%
Eu-154	1,30%
Sb-125	1%
Eu-155	0,50%
Te-125m	0,40%
Sm-151	0,30%
Total	100%

Activity	% of total
β (+α,γ)	10
β only	10
γ + β	80

Mix no.6 of beta-emitting isotopes showing their radionuclide composition or Source term

Review of Emission Limits for ITU

Source term limits currently under review.

The first point decided is to extend the cooling time since the fuels have all aged considerably. Minimum average of 5-7 years seems likely, alternatively calculate limits with variable cooling time.

This will mean increased proportion of the longer lived isotopes (ie Cs-134, and Sr-90 will represent lower proportions of the updated mix.

C-14 & tritium need also to be specifically considered.

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Origin of C-14 - comes from 2 routes: fission yield and irradiation. The latter becomes more important if there is carbide and nitride fuels particularly in fast spectra

C-14: $t_{½}$=5730 y (β emission)

1) Irradiation production of C-14 from C & N

Element	Isotopes	Content (%)	Reaction	Reaction x-section σ (thermal spectrum) (barns (10^{-24}cm)	Product	Half life
Carbon	C-12	98.93	n capture	0.0035	C-13	
	C-13	01.07	n capture	0.0014	C-14	
	C-14	0				
Nitrogen	N-14	99.636	n capture, n, p reaction	0.080 1.93	C-14	
	N-15	0.364	n capture	0.0004	N-16	

2) Fission yields of tritium & C-14 from a fast neutron spectrum

Isotope	Pu-239	U-235
fission yield of H-3 (%)	0,0142	0,0108
fission yield of C-14 (%)	3,01E-04	1,58E-04

The production of C-14 is approx. 10^5 times greater from N-14 (by n,p reaction) than from C-12
Therefore nitride inventory decisive.
For mixed carbonitride fuels 100% nitride will be a conservative assumption

Current Emission Limits for ITU

Fission Gas Emissions

Fission gas analyses include the Kr isotopes which can be released during puncturing tests. Here the main isotope is Kr-85 which is a β,γ emitter (the unstable Xe isotopes are very short-lived (eg .Xe-135).

The amounts from a single puncturing are far below the emission limits.

Kr-85 (t½=10.76y)
β: 700keV,
γ: 514keV, ~3%

Other Kr iostopes: Kr-83, 86
Other Xe iostopes: Xe-129,131,133, 134 & 136

Emission Limits for ITU - 1

Radium/ Radon equilibrium

The monitoring system of the Hot Cells ventilation showed an α-activity due to aerosols and/ or gases during cleaning of a cell that had been used for experiments with Ra-226 (t½= 1600yr). The daughter isotope: Ra-222 has short t½ of 3.825 days.
The cleaning of the cell did not bring much reduction in the activities/dose rates. Therefore the question: is Ra/Rn contamination in the cell or is it outside the cell. And if so how much Ra226 would give such activities. This required the equilibrium to be modelled and then measurements to be made from the cell's ventilation and from the general hot cells flow rates by switching the flows over the monitors

Alpha activities:	**Rd-226 (t½= 1600yrs) => Rn-222 (t½= 3.825 d)**
Alpha particle energies:	**Rd-226 (4.78, 4.60 MeV), Rn-222 (5.49 MeV)**

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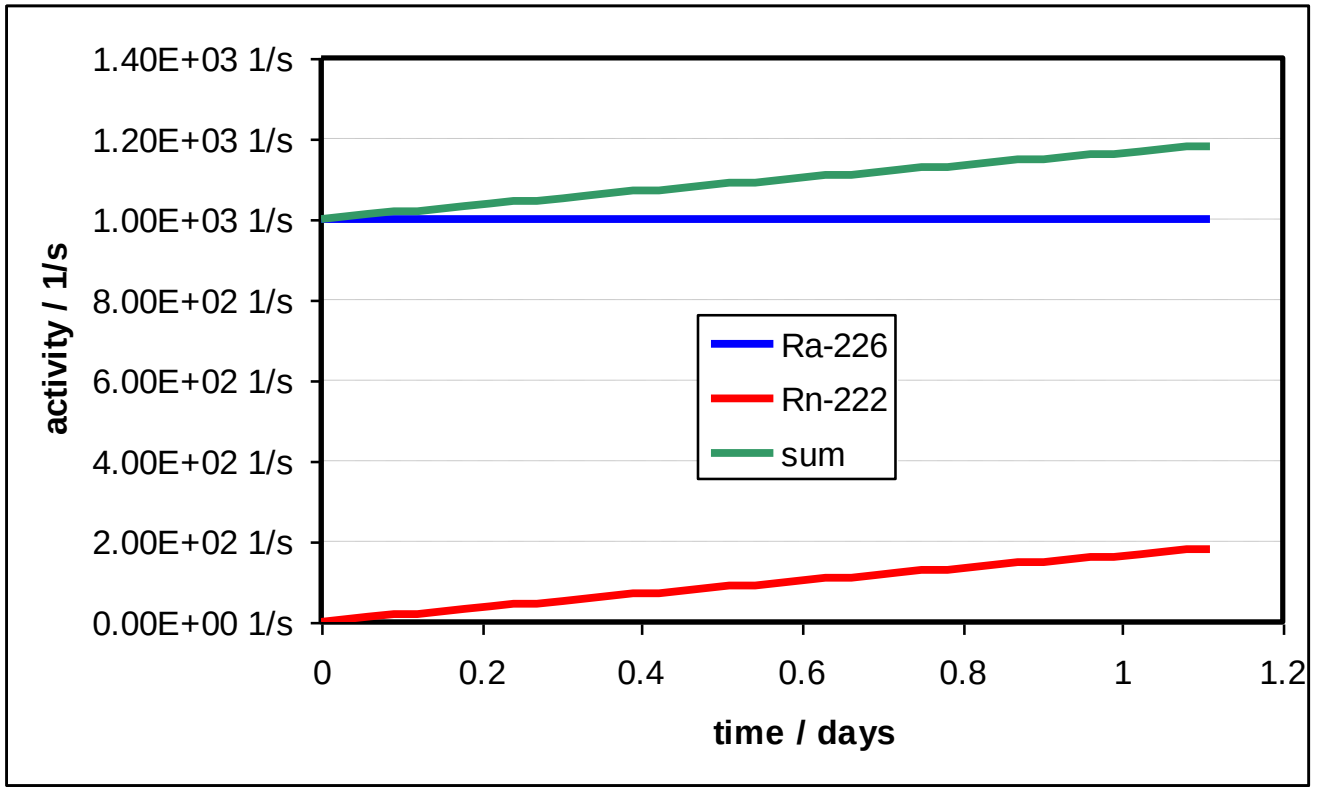
Emission Limits for ITU

Radium/ Radon equilibrium -2



Graph showing the equilibrium of radon-222 gas formed from radium-226 isotope with time

variation of the ventilation system used to see if the readings were from Rn or mixed Rd/Rn (ie Rd contamination in the ventilation system). If all from the cell then an estimation of the residual Rd could be made

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Fuel declarations at ITU

Pu and Cm concentration build-up with burn-up

Waste declarations at the intermediate storage facility often have problems to make complete isotopic declarations, particularly the higher actinides because of total neutron measurements at the facility

Isotope	Total Half-Life	Spontaneous Half-Life, years	Spontaneous Fission Yield, n/sec-gram
233U	1.59×10^{5} y	1.2×10^{17}	8.6×10^{-4}
235U	7.04×10^{8} y	3.5×10^{17}	2.99×10^{-4}
238U	4.47×10^{9} y	8.20×10^{15}	1.36×10^{-2}
238Pu	87.74 y	4.77×10^{10}	2.59×10^{3}
239Pu	2.41×10^{4} y	5.48×10^{15}	2.18×10^{-2}
240Pu	6.56×10^{3} y	1.16×10^{11}	1.02×10^{3}
241Pu	14.35 y	2.5×10^{15}	5×10^{-2}
242Pu	3.76×10^{5} y	6.84×10^{10}	1.72×10^{3}
241Am	433.6 y	1.05×10^{14}	1.18
242Cm	163 days	6.56×10^{6}	2.10×10^{7}
244Cm	18.1 y	1.35 x 107	1.08×10^{7}
252Cf	2.646 y	85.5	2.34×10^{12}

Cm isotopes have 10^{4}x greater neutron production/ g than Pu isotopes

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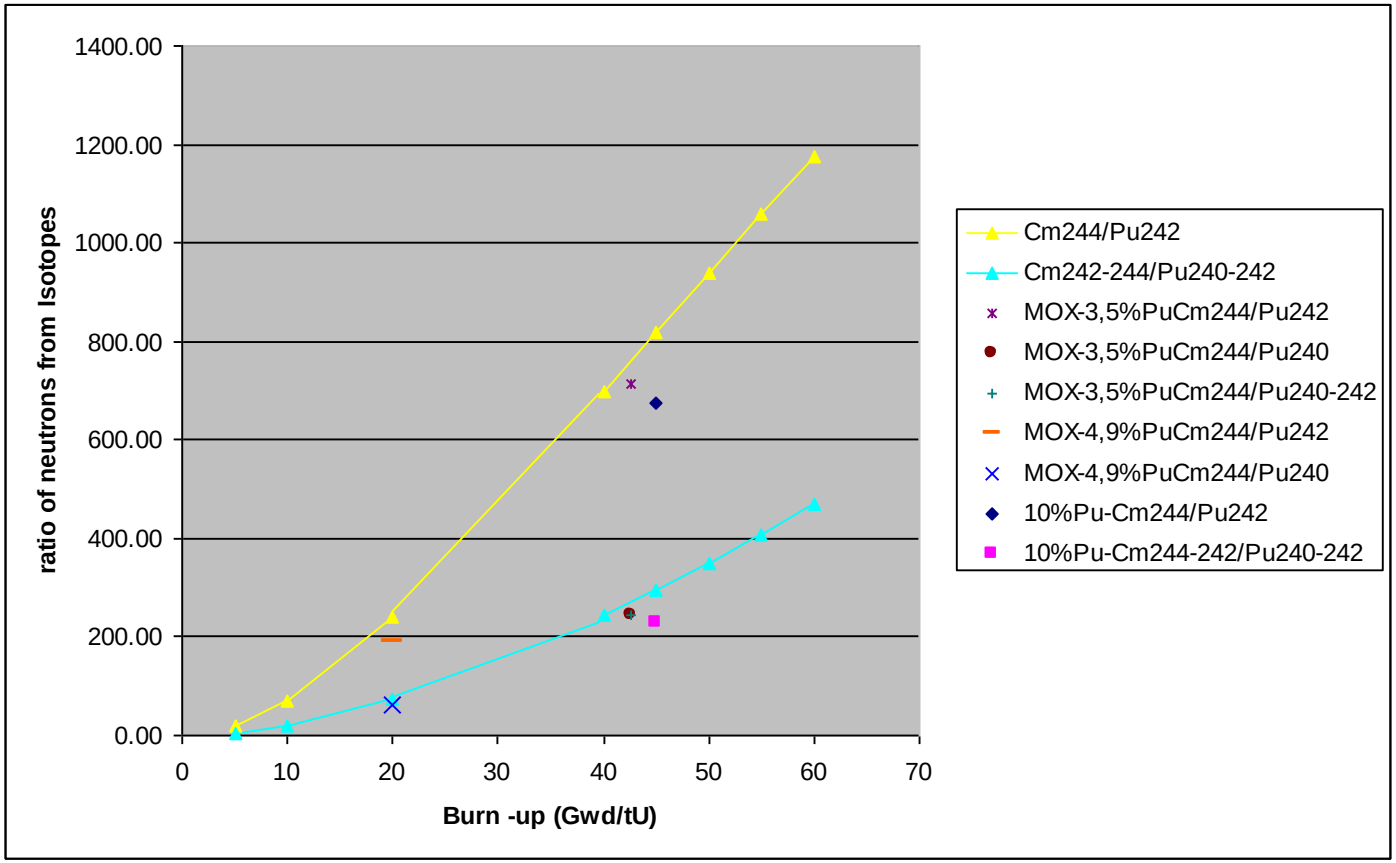
Fuel declarations at ITU

Pu and Cm concentration build-up with burn-up

Best ratio is that total Cm and total Pu, (not extremely high number to amplify errors), however Cm 244/Pu 242 also possible.

Some variation in the Pu content in fuel evident, but not very consistent.



Variation of neutron production ratio with burn-up of 8wt% Pu & 10 %Pu and various MOX fuels for comparison (5years cooling time)

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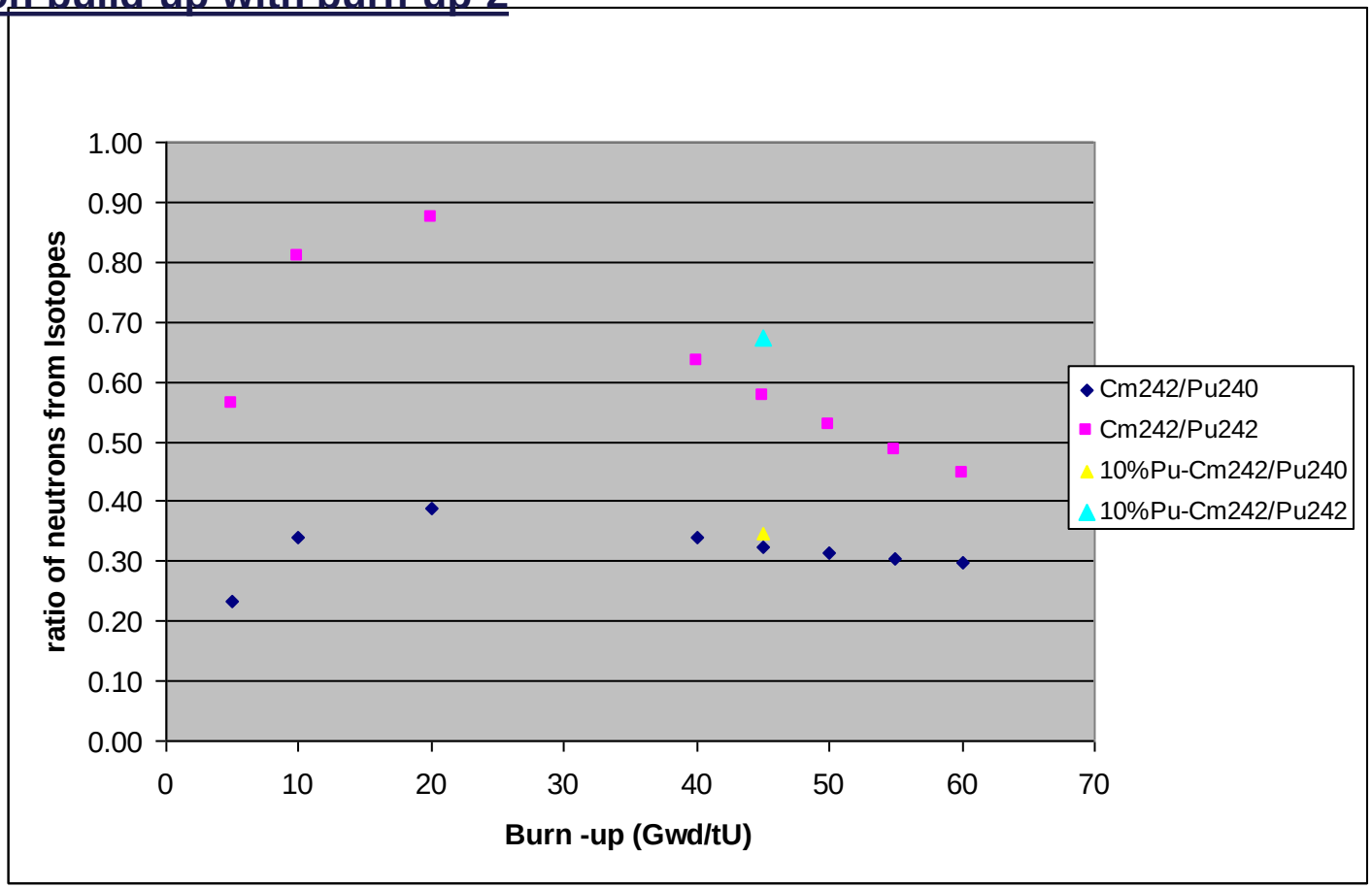
Fuel declarations at ITU

Pu and Cm concentration build-up with burn-up-2

Neutron ratio for the Cm242/Pu240 and CM242/Pu242 very favourable.

However this requires an analytical separation of the isotopes to be able to use Pu-242 as an estimator



Variation of neutron production rates with burn-up of 8wt% Pu MOX fuel (5 years cooling time)

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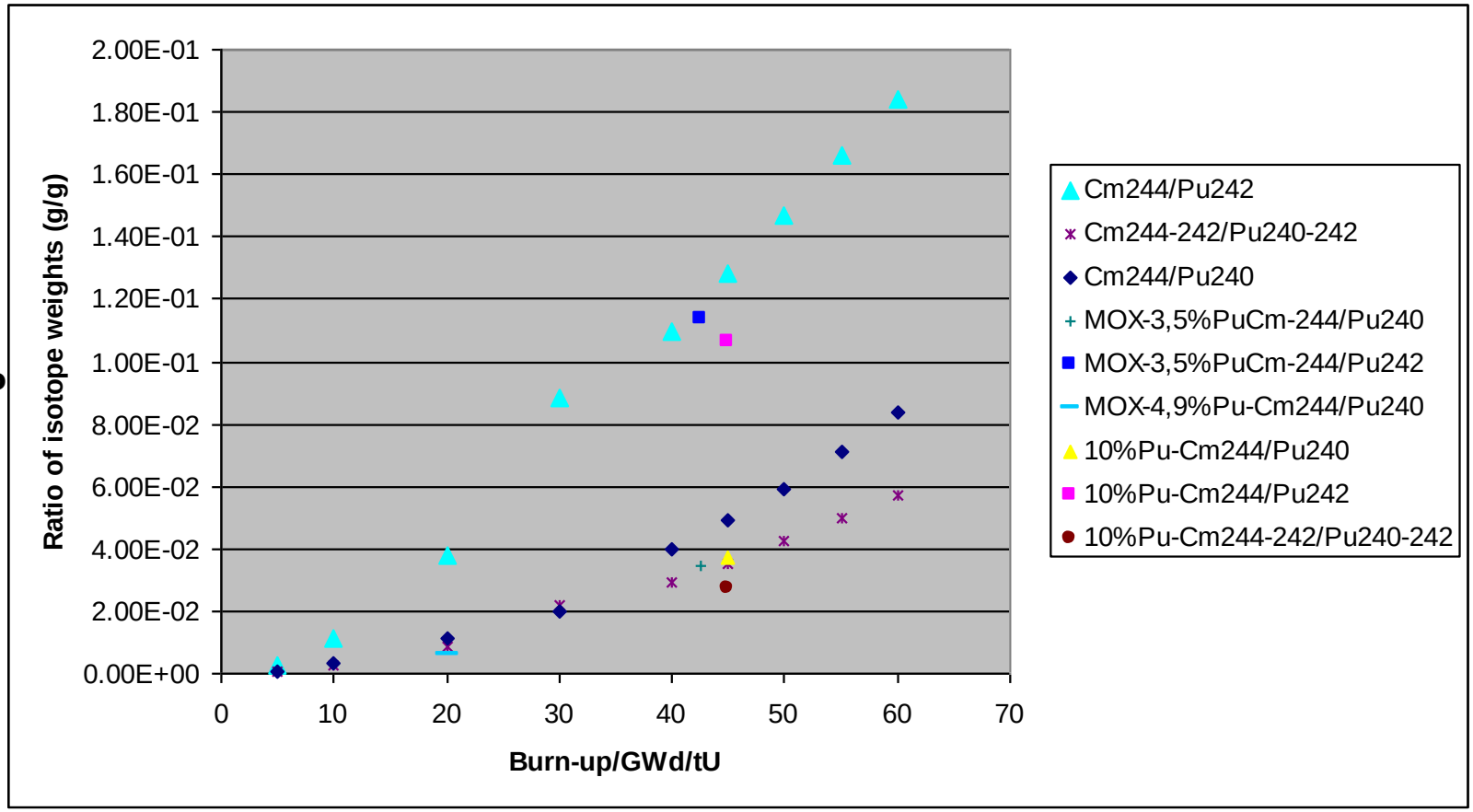
Fuel declarations at ITU

Pu and Cm concentration build-up with burn-up-3

Cm-244/Pu-242 is the smallest (best) ratio.

But Cm-244/Pu240 ratio is also possible



Isotopes weight ratios of 8% Pu Fuel compared with further MOX fuels of lower Pu content (5years cooling time).

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Fuel declarations at ITU

Pu and Cm concentration build-up with burn-up-4

Cm244/Pu242 or Cm244/Pu240 ratios

- **10% Pu content has distinct drop in the ratios compared to the 8wt% Pu MOX.**
- **MOX fuels with 4,9% Pu 3,9% Pu are only very slightly below 8% Pu MOX.**
- **10 % Pu results do not very different from (wt% Pu) - limits of the KORIGEN code?**

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Fuel declarations at ITU

Pu and Cm concentration build-up with burn-up-5

Estimation based on mass spectroscopic analysis.

The optimum is probably Pu-240 to Cm-242 or Cm-244.

Both Cm242 & Cm244 have only a factor 2 between their spontaneous neutrons, as do Pu 240 and Pu 242.

Pu-240 will have less interference (with a reasonably long half-life: 6563y)

Pu-242 needs separation from Cm-242 or Am-242 when determined by mass spectroscopy.

These factors suggest taking modelling estimates based on the weight ratio of Pu240 to Cm244 to estimate the total neutron counts expected from a Pu-containing oxide fuel sample.
(with good MS determination then (Pu-240+242)/(CM242+244) is better)

Conclusions

1) The various waste forms & emissions at ITU have been described along with the problems of operational limits and declaration requirements.

2) The problems relating to i) advanced Pu-containing fuel composition, particularly the higher actinides and their neutron radiation, ii) gaseous irradiation products (H-3 & C14) and their monitoring in relation to the emission controls in exhaust gases.

3) Modelling of Pu-containing fuel composition has been used for estimation and appears to be very reasonable for MOX fuels, but has limitations for the other forms such as metallic fuels.

4) The inventory for C-14 from carbide & nitride fuels has also estimated to give upper bounds for the emissions possible from the hot cells ventilation system

5) This work ensures that the best estimations possible are made for waste declarations and to assist the expedition of waste to intermediate storage, and that the potential emissions are assessed before the limits are approached.

Outlook

To continue the modeling of the gaseous products in order to agree on an update of the emissions limits (eg more realistic base case-include MOX with UO_2, increased average burn-up)

To include inventory of exotic fuels to assess C-14 source term

To standardise estimates of the higher actinides to assess neutron fluxes from waste