Characterization of Reprocessed Fuel by Wide Environmental Sampling

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Under the International Atomic Energy Agency's (IAEA) Additional Protocol wide environmental sampling is considered part of the novel safeguards technology for future use by the IAEA inspectorate. This research will investigate if this monitoring technique could also be applied to c haracterize reprocessed fuel and localize the corresponding place of radiochemical processing. Therefore, correlations between the environmental signature (measured volatile fission product isotope ratio's) and fuel characteristics (burn-up and isotopic composition as well as produced Pu) shall be determined. Then, the study will examine how these correlations can be used for detection, localisation and identification of undeclared activities.

Objectives

The study aims to determine the signature of different types of spent fuel by measuring the isotopic abundances of volatile fission products that are released during reprocessing. A thorough literature study and first theoretical analyses show that released noble gases, other volatile fission gases of chalcogenide, halogen and alkali metal type are the best candidates for this type of monitoring. Three types of measurement stations are considered: **1)** a noble gas measurement station at long distance (500 km from the reprocessing plant), **2)** a measurement station at the border of the reprocessing site (5 km from the stack), **3)** a measurement station for all volatile fission products in the stack.

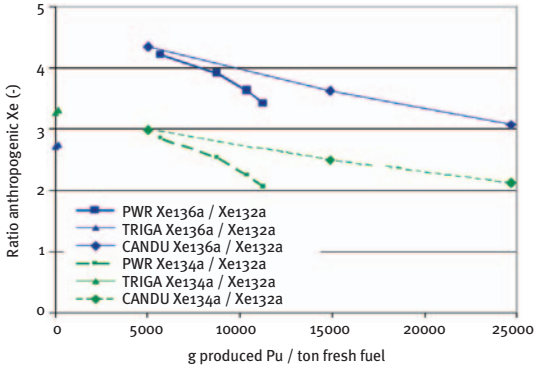

Fig. 1
Xe isotopic ratios for PWR, TRIGA, CANDU in function of produced Pu

For nuclear safeguards purposes detection of undeclared processing of low burnup fuel is important Therefore continuously refuelled reactors such as CANDU are of particular interest. Discrepancies between the declared activities and the measured releases could provide a basis for an early warning system.

This study examines if data that come off the different measurement stations can be correlated to determine the origin and nature of the source.

The fixed measurement stations that are placed by e.g. the Comprehensive Test Ban Treaty Organisation can provide input signals of the Kr-85 and the Xe-131m, Xe-133m, Xe-133, Xe-135 isotopes. When these stations measure deviations from natural abundancy the original source term must be located. To do so, the place of detection must be linked to the origin of the release under the given weather conditions. It will be examined if atmospheric modelling can be used for back tracing of the released noble gases.

Then the concentrations of other fission gases such as I-127 and I-129 can be estimated at the two other measurement points. It will be examined if these data confirm the type of reactor fuel and if they reveal additional information on the isotopic composition. For independent observations it is recommended to measure also other volatile fission products such as Te-130 and Te-128.

			NUBASE 2003	PWR (47520 MWd/ton)	CANDU (6000MWd/ton)	TRIGA (1742 MWd/ton)
			Nat. is. Abdc	gram% of inventory HM	gram% of inventory HM	gram% of inventory HM
Noble gas	gas	Kr-83	0.1149	4.90E-05	9.91E-06	3.55E-06
		Kr-84	0.5700	1.55E-04	2.07E-05	7.15E-06
		Kr-85	0.0000	2.57E-05	4.70E-06	1.14E-06
	Xe	Xe-132	0.2689	1.46E-03	1.53E-04	4.56E-05
		Xe-134	0.1044	1.98E-03	2.58E-04	8.36E-05
		Xe136	0.0887	2.93E-03	3.58E-04	6.95E-05
Halogens	I	I-127	1.0000	5.49E-05	5.41E-06	1.28E-06
		I-129	0.0000	2.33E-04	2.71E-05	7.73E-06
Alkalimetals	Cs	Cs-133	1.0000	1.47E-03	2.12E-04	7.20E-05
		Cs-135	0.0000	5.38E-04	7.76E-05	6.90E-05
Chalcogenides	Te	Te-128	0.3174	1.17E-04	1.29E-05	3.60E-06
		Te-130	0.3408	4.75E-04	5.85E-05	1.82E-05

Table 1
Fission gas nuclides, selected for isotopic correlation to characterise reprocessed spent fuel in function of burnup and fuel type.

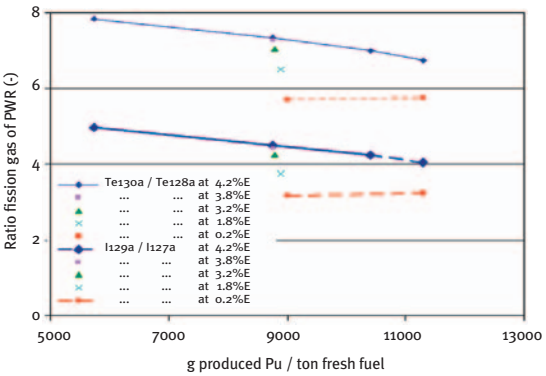

Fig. 2
Te and I isotopic ratios for PWR fuel for different enrichment grades (% E) in function of Pu

Results

A complete study of the different isotopes has been carried out. Burnup calculations with the SCALE code for PWR, CANDU and TRIGA yield significant isotopic ratios. The choice of the "Pu quantity" as variable showed to be beneficial because of a reduction in parameters and the direct safeguards interest. Moreover the isotopic ratios were converted to represent only the relative anthropogenic contributions (taking into account the natural isotopic abundancy. The evolution of Xe-136 / Xe-132 with quantity of produced Pu for different enrichment grades showed that this ratio is independent from the enrichment grade.

The Xe isotopic ratios are found to be reactor-type dependent as shown in Fig. 1 for the different reactors PWR, CANDU and TRIGA. With Kr-83 / Kr84 and Kr-85 / Kr-84 ratios, we can specify the enrichment grade of the PWR fuel.

These calculations have to be compared with experimental data, which have been collected and converted in the mean time. An extension to other reactor types is only useful if experimental data are available.

However, with these preliminary results the following procedure to characterize spent fuel using noble gas measurements at long distance may be promising.

a. Measure Xe-132, Xe-134, Xe-136
b. Derive anthropogenic ratios Xe-134 anthropogenic / Xe-132 anthropogenic and Xe-136 anthropogenic / Xe-132 anthropogenic
c. Selecting the appropriate reactor type based upon the indicate amount of Pu that was derived in (b)
d. Measure Kr-83, Kr-84, Kr-85
e. Derive anthropogenic ratios Kr-83 anthropogenic/Kr-84 anthropogenic and Kr-85/Kr-84 anthropogenic
f. Determine the matching enrichment grade (in case of PWR-fuel. In addition the volatile fission products that are not noble gases for the same type of reactors have been calculated with SCALE and some results are shown in Fig. 2. Using the same methodology we derived the following procedure that might be used for spent fuel characterization using stack measurements of other volatile fission products
g. Measure I-127, I-129
h. Derive anthropogenic ratios I-129 anthropogenic / I-127 anthropogenic
i. Verify the consistency with the previous selected reactor type.
j. Measure Te-128, Te-130
k. Derive anthropogenic ratios Te-128 / Te-130
l. Determine the matching enrichment grade (PWR-fuel). Verify if these enrichment determinations confirm the previous ones.

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