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#5-70 Discrimination of plutonium and curium in passive neutron multiplicity counting with PVT plastic scintillators

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In the framework of plutonium characterization in radioactive waste drums by passive neutron multiplicity counting, the Nuclear Measurement Laboratory of CEA Cadarache is studying plastic scintillators as a cost-effective alternative to 3He gas proportional counters. Plastic scintillators offer a three order of magnitude faster time response than 3He detectors, large and easy-to-shape detection volumes, and a similar neutron detection efficiency at a much lower cost when using basic PVT scintillators without PSD capabilities, like EJ-200 or BC-408 commercial models. However, their high sensitivity to gamma rays makes it difficult to use plastic scintillators in neutron-gamma mixed fields. Previous studies conducted at the Nuclear Measurement Laboratory of CEA DES IRESNE, at Cadarache, France, have shown that it is possible to measure plutonium spontaneous fission neutrons despite the presence of parasitic (α,n) neutron emitters and gamma-ray sources, using data processing techniques based on the historical shift register method combined with time-of-flight analysis.This work present a new patent-pending data processing, based on several time-of-flight windows combined with multiplicities analysis, to further allow for plutonium quantification even in the presence of perturbing spontaneous fissions from 244Cm. The performances are studied through the development of a full factorial design using MCNP-PoliMi simulations of a measurement system for 118 liters waste drums, composed of sixteen 10×10×100 cm3 EJ-200 plastic scintillators. The drum is homogenously filled with an organic matrix of density 0.3 g. cm-3 contaminated with plutonium and curium quantities equivalent to a 240Pu effective mass between 0 and 500 g. The matrix is also uniformly contaminated by 60Co with an activity between 0 and 2 MBq. In order to account for the multiplication effects associated with high plutonium masses concentrated in clusters, an additional experimental design was developed with spherical PuO2 samples of density 3 g.cm-3. They are located in the center of the organic matrix and cover a 240Pu effective mass from 0 to 650 g, while the curium background still ranges from 0 to 500 g of 240Pueff. Data processing algorithms are used to reconstruct the detectors response from the MCNPX-PoLiMi collision files, including real and accidental coincidences, a low-energy threshold of 50 keVee (electron-equivalent), and a time resolution of 3 ns (FWHM) to each detection channel. The measured quantities of interest, to highlight the contrast between Cm and Pu are for instance, the singles, doubles and triples of traditional multiplicity counting, but here recorded in specific time windows to take advantage of the slight difference in the time-of-flight (ToF) of Pu and Cm spontaneous fission neutrons. Simulations indicate that this time difference can be exploited for Pu/Cm differentiation in Rossi-Alpha and two-dimensional time histograms, based on the relative detection times of the particles involved in double and triple coincidences respectively. All these input data (explanatory variables) are fed into a multilinear regression algorithm, or a machine learning artificial neural network, to obtain predictive models of the 240Pu effective mass. Monte Carlo methods are finally used to account for the impact of explanatory variables uncertainties on the accuracy and robustness of the models. In addition, test configurations are simulated, different from the training set but included in the limits of the factorial design, to assess the predictive capabilities of the numerical models. The best model obtained for the prediction of plutonium mass involves five explanatory variables: Singles (S), doubles counted between 10 and 20 ns (D10-20) after the first detected particle (which is often a fission gamma ray) and between 10 and 60 ns (D10-60), triples between 10 and 20 ns (T10-20) and between 10 and 60 ns (T10-60). This approach allows a prediction of the plutonium mass on the full range of the experimental design, i.e. from 0 to 650 g of 240Pueff, whatever the quantity of Cm between 0 and 500 g of 240Pueff. This is a promising result in view of future experimental validation tests planed with the setup, and this new approach would increase the state-of-the-art limit of Cm/Pu mass ratio by one decade to allow Pu assay.

Primary author: Dr BOTTAU, Vincent (CEA)

Co-authors: Dr PEROT, Bertrand (CEA); Dr ELEON, Cyrille (CEA); Mr D'ALMEIDA, Filip (CEA)

Presenter: Dr BOTTAU, Vincent (CEA)

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