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Inventories Estimation Method in Solid Residuals of a Irradiated HEU (from Mo-99 Production Targets) Purification and Down-blending Process

J.O. Vaccaro, P.M. dos Reis, M.S. Alí, V.C.Ferrero, M.R.Varone
Laboratorio Facilidad Radioquímica (LFR)
National Atomic Energy Commission (CNEA), Av. Del Libertador 8250, C1429BNP Buenos Aires – Argentina

ABSTRACT

The National Atomic Energy Commission of Argentina has carried out a campaign in order to recover and down-blend its remaining irradiated highly enriched uranium (HEU, from Mo-99 production targets) inventories to low enriched (19,75%±0,2%). This material was mainly in the form of solids on stainless steel filters and was purified in a hot cells operated process and the solutions (products of this purification) were down-blended with natural uranium. The first step of this process involves the nitric acid dissolution of the uranium content of each filter. The yield of this dissolution process has to be determined in order to accomplish process losses estimations and safeguards. A method based on high resolution gamma spectrometry (with Hyper Pure Germanium detectors, HPGe) and dose rate measurements has been developed and in the present work this method to determine the solid residuals is described, analyzed and its results presented. This method has been accepted by the Regulatory Authorities and has been used during the whole campaign.

1. Introduction

In the context of irradiated highly enriched uranium recovery from stainless steel filters used in Mo-99 production facility, it has been necessary to determine, in order to accomplish safeguard requirements, the differences between the content of uranium declared by the sender and the actual amount of uranium.

The filters to separate Mo-99 after irradiation, using an alkaline agent, are made of a cup with a plate of synthesized stainless steel in the bottom, that retains all insoluble impurities (principally uranium, another actinides and lanthanides) and leave pass through the soluble compounds (molibdenum, which will be separated downstream, and soluble impurities)[1].

These filters were stored, over more than ten years, as waste. In March 2010, a contract between National Nuclear Security Administration (NNSA-DoE) and the Comisión Nacional de Energía Atómica (CNEA) was signed in order to minimize HEU remaining inventories in Argentina. The campaign to recover and down-blend HEU inventories coming from Mo-99 production has been carried out in the Laboratorio Facilidad Radioquimica (LFR), a hot cells facility located in the Ezeiza Atomic Center (CAE).
The process to recover and down-blend the highly enriched uranium was based on an acid dissolution with nitric acid, chromatographic purification (TBP/XAD-7 column) and mixing with natural uranium solutions to down-blend.
Davies-Gray titration method was employed to determine the total amount of uranium dissolved, and uranium isotopic analysis was performed using ICP-MS technique.
Differences between actual and declared amounts were, in some cases, not negligible. So it was indispensable to determine whether these differences came from the dissolution process yield or from true discrepancies between declared and actual amounts of uranium on the filters.

2. Methodology

To estimate the undissolved uranium on the filters a method based on a combination of measurement and calculation of the gamma/X-rays emitted was employed.
The filters were put in an stainless steel vessel or “cartridge” and its dose rate was measured (at a distance of 1 meter).
After that a gamma ray spectrometry of the cartridge was performed (using HPGe detector - 20% relative efficiency).

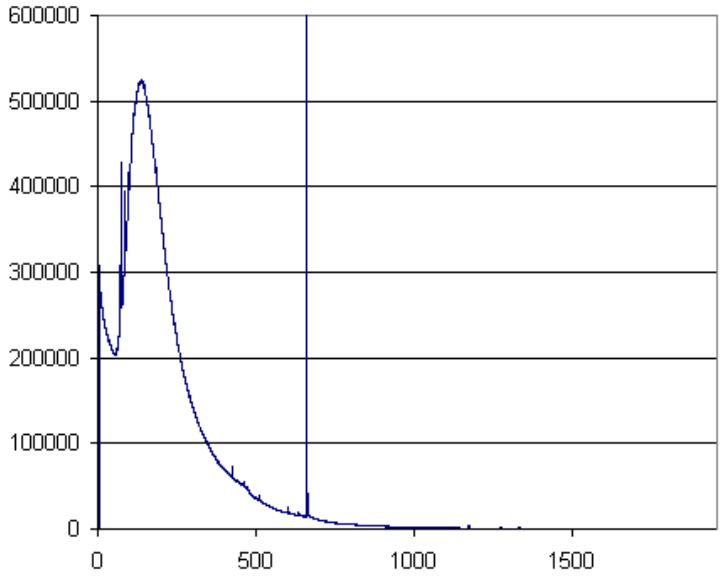

Fig. 1. Spectrum obtained with Genie-2000 using a HPGe detector. The data obtained was transferred to a spreadsheet in order be able to analyze it.

Fig. 2. HPGe detector used in the measurement of filters post-dissolution.

Using the Sb-125 fission product peaks, a relative efficiency calibration curve was fitted. The spectrum was corrected with that efficiencies and normalized to obtain a “fractional spectrum”. This “fractional spectrum” multiplied the total photons emission rate gives a good approximation of the cartridge photons emission spectrum.

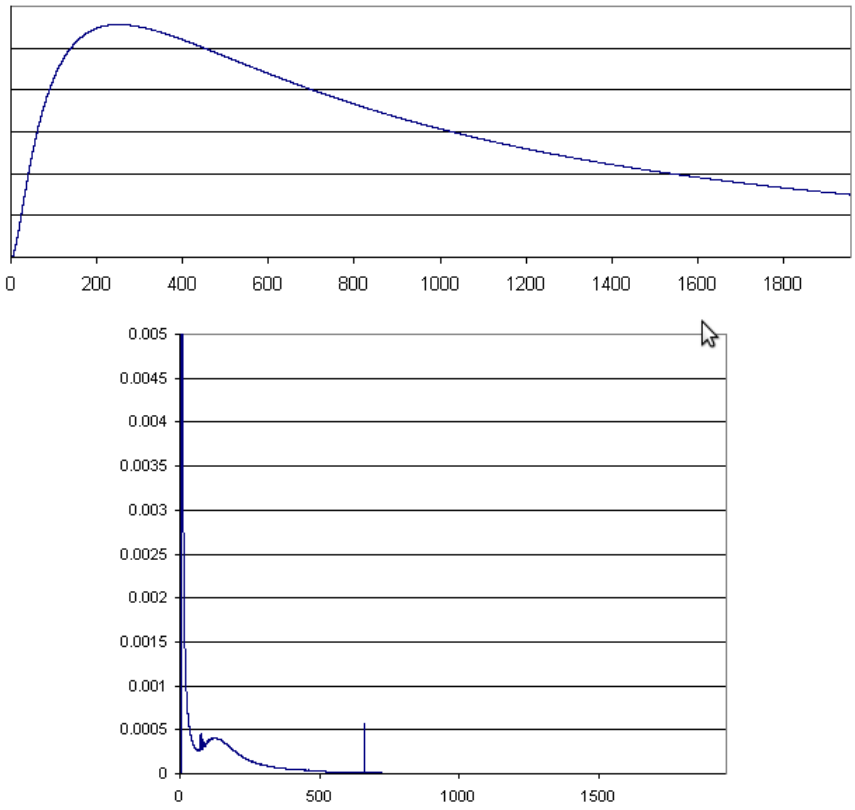

Fig. 3. Relative efficiencies obtained from the peaks of Sb-125 and efficiency-independent emission spectrum. It can be viewed the relative importance of low energy emission due to bremsstrahlung of beta emitters interacting with the wall of the container cartridge.

Main Sb-125 (2.75855 a) gamma spectrum peaks[2]	
Energy (keV)	**I%**
427.874	29.2
463.365	10.36
600.597	17.55
635.950	11.19

Ch. 1. Gamma spectrum peaks of Sb-125. These four peaks were used to fit a relative efficiency curve.

Moreover, using Microshield v5.03 software a cylindrical geometry was created to obtain the dose rate measured. The source of the simulation was performed with the corrected spectrum of photons obtained previously, allowing to reach an emission rate that explains the measured dose rate.

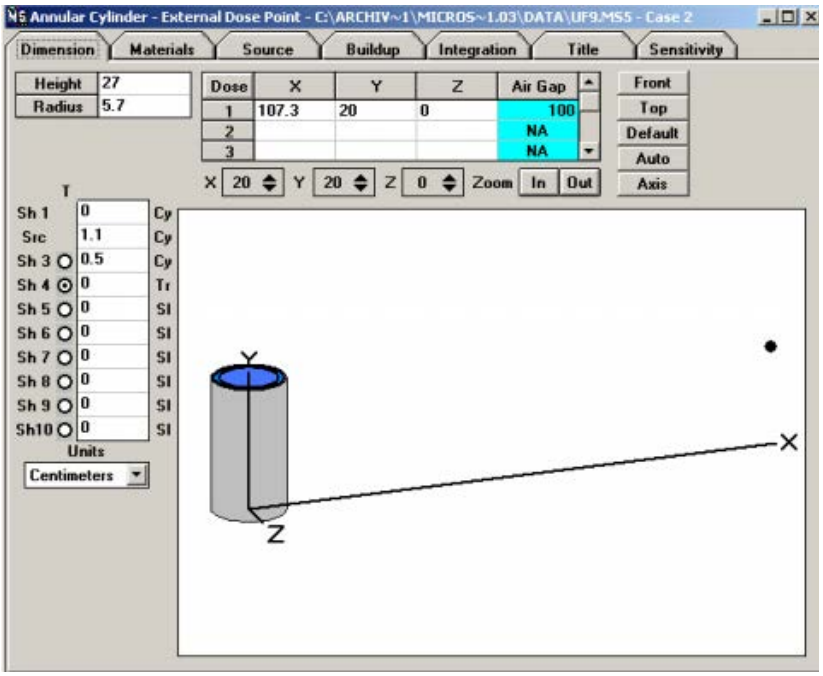

Fig. 4. Microshield chase. A cylinder including the source distributed in it was used as model.

With the total emission rate of photons distribution and the 185.712 keV U-235 peak, an estimation of the U-235 emission rate can be obtained and, therefore, the mass of this nuclide considering the absolute intensity corresponding to that peak (57.25%).
The measurement was made in the cartridge named U#F-9, containing 3 previously processed filter cups and re-inserted after.

Fig. 5. Filter cup inside the hot cells.

3. Results

The results of this measuring-calculation technique indicate that the mass of U-235 remaining in this item is about 0.4 g.

Dose rate (1 meter)	3.43 microSv/h
Emission rate (total)	4.91 x 10^8 photons/s
U-235 remaining mass	0.4 g

Ch. 2. Results of measurements and calculations.

The total mass found after dissolution was 34.49 g. Therefore, the loss in dissolution process is about 1.16% of the diluted mass.
This percentage was rounded to 1.5% and used to determine the remaining masses in the rest of the irradiated HEU items processed in the LFR facility considering that the dissolution process had not varied significantly its yield.

4. References

[1] Operación de la instalación para producción de Mo-99 partiendo de productos de fisión; Marqués R. O., Cristini P. R., Marziale D. P., Furnari E. S., Fernández, H.; Dirección de Radioisótopos y Radiaciones; Comisión Nacional de Energía Atómica; Ezeiza, Argentina.; Actas de la XV Reunión Científica, IV Encuentro Latinoamericano, I° Jornadas sobre Centrales Nucleares; San Carlos de Bariloche, Río Negro, Argentina; 2 al 6 de Noviembre de 1987.

[2] Table of Radionuclides (Vol. 1 - A = 1 to 150) – BIPM (Monographie BIPM-5); M.M. Bé, V. Chisté, C. Dulieu, E. Browne, V. Chechev, N. Kuzmenko, R. Helmer, A. Nichols, E. Shönfeld, R. Dersch; Bureau International des poids et mesures; 2004.