Evaluation of a measurement system for Uranium electrodeposition control to radiopharmaceuticals production

Tufic Madi Filho, Adonis Marcelo Saliba Silva, José Patrício Nahuel Cárdenas, Maria da Conceição Costa Pereira, Valdir Maciel Lopes, Alexandre P. S., Diogo F. S., Rafael T. P., Vitor O. A, Anderson F. L., Lucas R. S., Brianna S., Eduardo L. C.

***Abstract*– For 2016, studies by international bodies forecast a crisis in the supply of Molybdenum (^{99}Mo), which is the generator of ^{99m}Tc, widely used for medical diagnoses and treatments. As a result, many countries are making efforts to prevent this crisis. Brazil is developing the Brazilian Multipurpose Reactor (RMB) project, under the responsibility of the National Nuclear Energy Commission (CNEN). The RMB is a nuclear reactor for research and production of radioisotopes used in the production of radiopharmaceuticals and radioactive sources, broadly used in industrial and research areas in Brazil. Electrodeposition of uranium is a common practice to create samples for alpha spectrometry and this methodology may be an alternative way to produce targets of low enriched uranium (LEU) to fabricate radiopharmaceuticals, as ^{99}Mo, used for cancer diagnosis. To study the electrodeposition, a solution of 10mM uranyl nitrate, in 2-propanol, containing uranium enriched to 2.4% in ^{235}U, with pH=1, was prepared and measurements with an alpha spectrometer were performed. These studies are justified by the need to produce ^{99}Mo since, despite using molybdenum in bulk, Brazil is totally dependent on its import. In this project, we intend to obtain a process that may be technologically feasible to control the radiation targets for ^{99}Mo production.**

I. Introduction

In recent years, nuclear medicine diagnostic imaging has become a powerful noninvasive tool, providing information on anatomic and metabolic processes of human body and showing local biochemistry of ill or damaged tissues This type of diagnosis plays an important role in the identification of diseases in an early stage so that, before the appearance of anatomic changes, they allow the illness evolution to be followed so as to evaluate different alternatives of treatment and more precise diagnoses.

The main radiopharmaceutical for diagnostic purposes is the ^{99m}Tc. Technetium-99m (^{99m}Tc, with a radioactive half-life of about 6h) is generated by decay of Molybdenum-99 (^{99}Mo, radioactive half-life of about 66h) and it is produced in nuclear research reactors. Figure 1 shows the ^{99}Mo production and the decay of ^{99m}Tc.

Molybdenum-99 (^{99}Mo$_{42}$)
(Fission product)

Half-life : 65.94 h

Production scheme : ^{235}U (n,f) ^{99}Mo

Decay products : ^{99m}Tc ($t_{1/2}$ 6.01 h) and ^{99}Tc ($t_{1/2}$ 2.11 × 10^5 a)

Type of decay and energy

β	1.2145	(82.4%)	γ	0.7395	(12.13%)
	0.4366	(16.4%)		0.181	(6.0%)
				0.1405	(4.52%)
				0.7779	(4.26%)

Production : thermal neutron induced fission of ^{235}U; fission products contains 6.1 mass-% ^{99}Mo

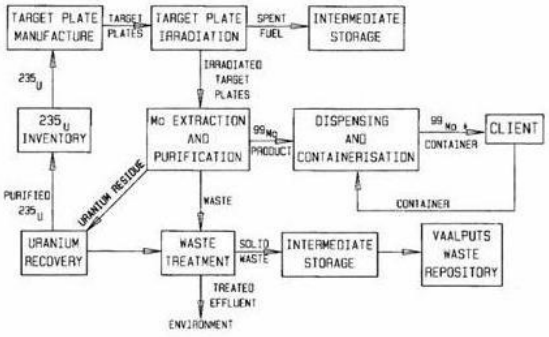

Fig. 1 ^{99}Mo process flow sheet [1]

In order to prevent problems with ^{99m}Tc production, Brazil is developing the Brazilian Multipurpose Reactor (RMB) project. The RMB is a nuclear reactor for research and production of radioisotopes used in the production of radiopharmaceuticals and radioactive sources. The plates for irradiation will be produced by electrodeposition of low enriched uranium (LEU). The first quality control will be made using alpha spectrometry.

The alpha particles detection can be realized with a semiconductor detector Silicon Surface Barrier. The range of α particle in silicon is 10 microns to 1000 microns, to energies of 3.0 MeV to 50 MeV. This detector type offers advantages

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All Authors: Nuclear and Energy Research Institute, IPEN–CNEN/SP
Instituto de Pesquisas Energéticas e Nucleares – IPEN-CNEN/SP
Av. Prof. Lineu Prestes 2242 Cid Univers. CEP: 05508-000– São Paulo-SP.
Tufic Madi Filho, tmfilho@ipen.br *correspondent author.*
Adonis Marcelo Saliba Silva, saliba@ipen.br
José Patrício Nahuel Cárdenas, ahiru@ipen.br
Maria da Conceição Costa Pereira, macoper@ipen.br
Valdir Maciel Lopes, vmlopes@ipen.br
Alexandre P. S., alexpovoa@yahoo.com.br
Diogo F. S., diogofa@msn.com
Rafael T. P., rtpurgato@ipen.br
Vitor O. A, vitoraredes@gmail.com
Anderson F. L., anderson_figueredo@hotmail.com
Lucas R. S., dossantos.lucasrodrigues@gmail.com
Brianna S., briannabs@yahoo.com.br
Eduardo L. C. educorrea1905@gmail.com

in its use: high density of silicon and, consequently, the ionizing particle loses all the energy in a short path, high capacity of a pair electron-hole production, ease of construction and operation at room temperature [2].

Alpha spectrometry has shown growing importance in diverse nuclear fields. This technique is commonly used to obtain the uranium isotopic composition in different matrices. Therefore, uranium isotopes measurements at low-level activity concentration are possible. These measurements are of great interest in nuclear industry, health physics, waste management, radiological protection and environmental science [3]

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II. MATERIAL AND METHOD

The used sources were electrodeposited: two radioactive sources for system calibration were used: ^{241}Am with an activity of 13.86Bq, and natural uranium source with mass of 7.44 10^{-3}g. In order to evaluate the system, a source of 2.4% uranium enriched was used.

Electrodeposition of uranium is a common practice to create samples for alpha spectrometry [4] and this methodology could be an alternative way to produce low enrichment of uranium (LEU) irradiation targets [5] and fabricate radiopharmaceuticals, such as ^{99}Mo, used for cancer diagnosis [6]. The low temperature electrodeposition in ionic solutions was the utilized process, mainly using RTIL procedure with moderate success, as accounted in recent papers in the literature [7].

II.1. MEASUREMENTS

The sources were placed in the vacuum chamber of an Alpha King Spectrometer 676A Ortec in drawers with, approximately, 2 mm spacing. Figure 2 shows a photo with the source inside the detector.

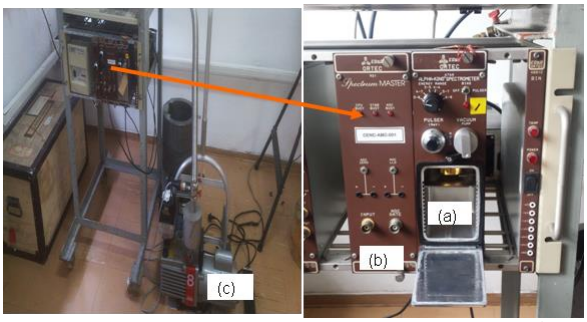


Fig. 2 Alpha spectrometry system: (a) Spectrometer with surface barrier detector inside the vacuum chamber, (b) Spectrum Master ADC device, (c) vacuum pump

The spectra obtained for 241Am source and natural uranium with Maestro software are shown in figures 3 and 4. Figure 3 shows the peak to 241Am and Figure 4 for the natural Uranium.

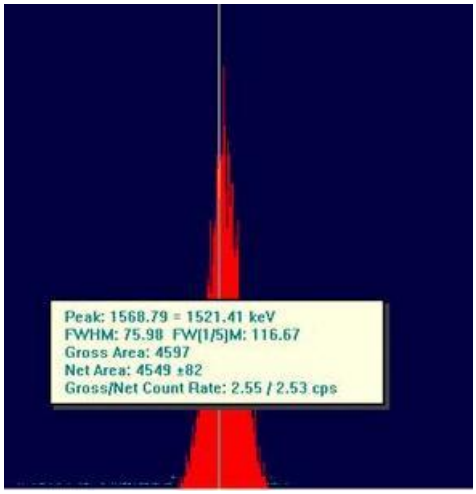


Fig. 3 ^{241}Am source spectrum.

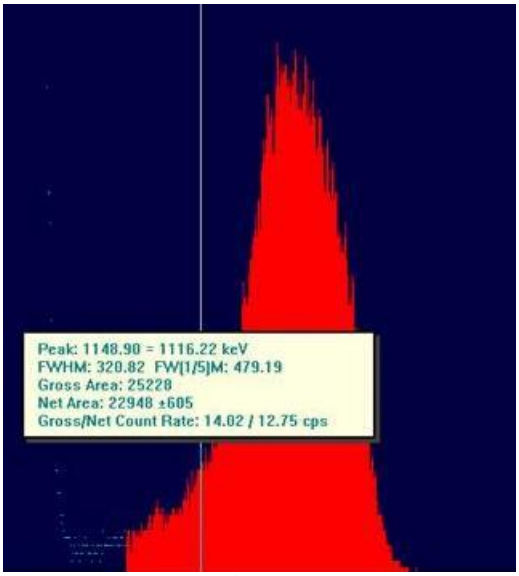


Fig. 4 Spectrum of natural uranium source.

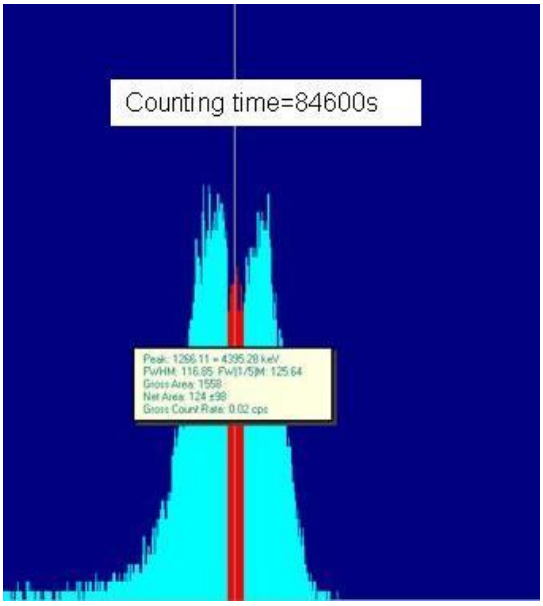


Fig. 5 - Alpha spectrum of uranium enriched to 2.4% in ^{235}U.

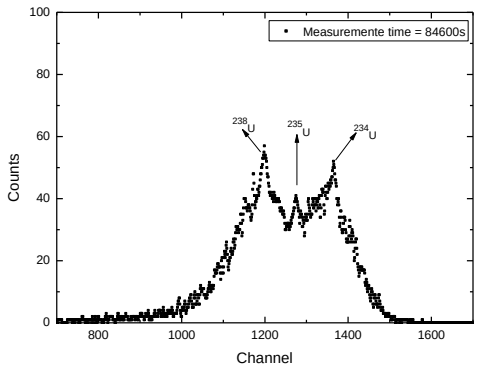


Fig. 6 Uranium enriched to 2.4% in ^{235}U Alpha spectrum, after smooth.

Figure 7 presents an alpha spectrum obtained by Dumitru et all, for a uranium sample electrodeposited, similar to that used in this work.

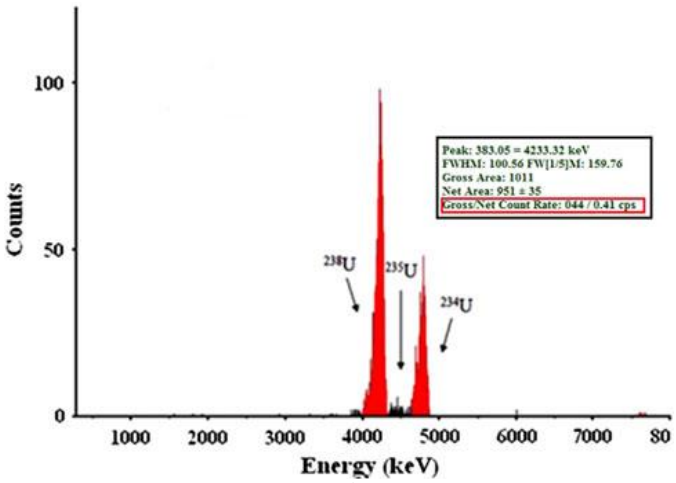


Figure 7 - Alpha spectrum of uranium sample obtained by electrodeposition [8]

II.2. CALCULUS

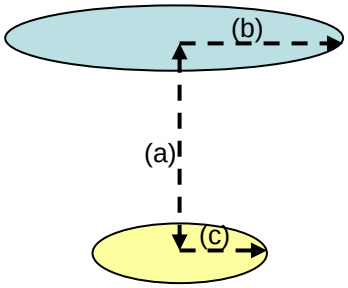
FIRST METHOD

First, the system efficiency using the Am-241 source was determined and the value obtained was used for calculating the mass of natural uranium of the used source, with known value. Then, the mass of uranium enriched source was calculated and compared with the electrodeposition value.

The distances source-detector and radius of these sources were used to determine the geometric factor, equation 1, [9].

$$G = 0.5\left\{1-\frac{1}{(1+\beta)^{1/2}}-\frac{3}{8}\frac{\beta\gamma}{(1+\beta)^{5/2}}-\gamma^2\left[-\frac{5}{16}\frac{\beta}{(1+\beta)^{7/2}}+\frac{35}{64}\frac{\beta^2}{(1+\beta)^{9/2}}\right]-\gamma^3\left[\frac{35}{128}\frac{\beta}{(1+\beta)^{9/2}}-\frac{315}{256}\frac{\beta^2}{(1+\beta)^{11/2}}+\frac{1155}{1024}\frac{\beta^3}{(1+\beta)^{13/2}}\right]\right\} \quad (1)$$

where,



G = Geometric factor;
$\gamma = c^2/a^2$;
$\beta = b^2/a^2$
a = distance between the source and the detector;
b = radius of the detector;
c = radius of the source.

The values of the experimental measurements, of G factor calculated and data about the ^{241}Am sources were used to determine the system efficiency, using the equation 2:

$$\varepsilon = \frac{C_{Net}}{A \cdot Y \cdot G \cdot t_c} \quad (2)$$

where,

C_{Net}= Net Counting,
A= source activity,
Y= % of alpha decay
G = Geometric factor,
t_c = counting time.

Therefore, the equation of source activity may be writing:

$$A = \frac{C_{Net}}{\varepsilon \cdot G \cdot Y \cdot t_c} \quad (3)$$

with:

$$m = \frac{N \cdot M}{A_V} \quad (4)$$

$$A = \lambda N \quad (5)$$

where,
m = mass,
M = Mol,
A_V = Avogadro number,
A= activity;
λ = decay constant;
N = number of radioactive nuclei;

The mass of Uranium is possible to be calculated. The specific activity that is defined as the activity per unit mass of the radioisotope sample:

$$\frac{A}{m} = \frac{\lambda \cdot N}{N \cdot M / A_V} = \frac{\lambda \cdot A_V}{M}$$

Since the half-life is: $T_{1/2} = \frac{\ln 2}{\lambda}$,

Then:

$$m = \frac{A \cdot M \cdot T_{1/2}}{\ln 2 \cdot A_V}$$

$$m = 2.3965 \cdot 10^{-24} \cdot A \cdot M \cdot T_{1/2} \quad (6)$$

Thus, the mass of each uranium isotope: U-234, U-235 and U-238, in the source may be calculated using de eq. 6

SECOND METHOD

Albert CAU (10) proposed a method for calculating the isotope ratio, in this work it was used, with some modifications.

Isotope ratio (IR) = $^{235}U/^{238}U$ (7)

$$IR = \frac{N_{235}}{N_{235} + N_{238}} \rightarrow IR = \frac{1}{1 + \frac{N_{238}}{N_{235}}}$$

As seen from eq. 5: $A = \lambda N$

then,

$^{235}A = \lambda_{235} N_{235}$ $^{238}A = \lambda_{238} N_{238}$

So

$$\frac{A_{235}}{A_{238}} = \frac{\lambda_{235} \cdot N_{235}}{\lambda_{238} \cdot N_{238}} \rightarrow \frac{N_{235}}{N_{238}} = \frac{\lambda_{235} \cdot A_{235}}{\lambda_{238} \cdot A_{238}}$$

Being $T_{1/2} = \frac{\ln 2}{\lambda}$

It may be written:

$$\frac{N_{235}}{N_{238}} = \frac{T_{1/2}(235)}{T_{1/2}(238)} \frac{A_{235}}{A_{238}}$$

then,

$$\frac{N_{235}}{N_{238}} = 0.1575 \cdot \frac{A_{235}}{A_{238}}$$

So, eq. 7 can be written:

$$IR = 0.1575 \cdot \frac{A_{235}}{A_{238}} \quad (8)$$

For the data obtained in the same measure system, A_{235} and A_{238} may be considered as the net counting obtained. Observing that ^{234}U is daughter of ^{238}U and is in secular radioactive equilibrium in the source studied, then $\mathbf{C_U = C_{238} + C_{235} + C_{234}}$, in alpha spectrometry. Thus, the enrichment in U_{235} may be written:

$$Enrichment = 0.1575 \cdot \frac{C_{235}}{C_{238} + C_{234}} \quad (9)$$

The enrichment, result of the second method, it is not affected by the geometric factor or the measurement system efficiency.

III. RESULTS

Using (1), the geometric factors to the sources used in this work were calculated. Table 1 presents the geometric factors calculated.

TABLE I. GEOMETRIC FACTORS

Source	Radius (cm)	Geometric Factor (G)
^{241}Am	0.50	0.416
U natural	1.50	0.316
U enriched	0.50	0.416

With the data obtained for the source of ^{241}Am, using (2), it was possible to determine the measurement system efficiency of 0.44 ± 0.01, whereas Y = 1, due to the poor resolution of the equipment, all of the alpha energies were considered at the peak.

With the efficiency determined and the values obtained in the references plus the values of the geometric factor in (3), the activities calculated are presented in Table 2:

TABLE 2 – VALUES OF THE CALCULATED ACTIVITY

Material	U natural source	^{238}U	Enriched source ^{235}U	^{234}U
Activity (Bq)	87.98	$37.88\ 10^{-3}$	$5.68\ 10^{-3}$	$35.59\ 10^{-3}$

Table 3 presents the mass values calculated using the first method (6).

TABLE 3 – VALUES OF THE CALCULATED MASS IN THE USED SOURCES

Material	U natural source	^{238}U	Enriched source ^{235}U	^{234}U
Mass (g)	$7.44\ 10^{-3}$	$3.21\ 10^{-6}$	$7.10\ 10^{-8}$	$1.56\ 10^{-10}$

Considering that the natural uranium source was deposited, m= $6.66\ 10^{-3}$ g and the obtained value was m= $7.44\ 10^{-3}$g (Table 3), the error between the deposited and calculated values was:

$$E = \frac{6.66 \cdot 10^{-3} - 7.44 \cdot 10^{-3}}{6.66 \cdot 10^{-3}} = 7.66 \cdot 10^{-2}$$

E = 7.66%

The uranium enriched source has 2.4 in ^{235}U, with Table 3 data:

$$\text{Enrichment} = \frac{m_{235}}{m_{238} + m_{234}}$$

Enrichment = 2.21%

The error relative to the deposited value (2.4%) is:

$$E = \frac{2.4 - 2.21}{2.4} = 7.9 \cdot 10^{-2}$$

E= 7.9%

Using the second method (9):

$$Enrichment = 0.1575 \cdot \frac{274}{980 + 890} = 0.0231$$

Enrichment = 2.31%

The error relative to the deposited value (2.4%) is:

$$E = \frac{2.4 - 2.31}{2.4} = 3.75 \cdot 10^{-2}$$

E = 3.75%

CONCLUSION

Comparing the spectrum of Fig 5 with that of Fig 6, it is possible to verify that the used system has poor resolution.

The result of the calculations presents error compared with the values of the sources preparation. Some of this error is due to the measurements, but the main part of the error is owing to the poor resolution of the surface barrier detector. The alpha energy peaks are not well separated and this produced a pile of pulses, during the measurements.

There are a lower percentage of errors in the second calculus method, but both are affected by errors of the measurement due to the resolution. The Enrichment result in the second method is not affected by geometric factor and efficiency of the measurement system, but it needs a better resolution to separate the ^{234}U, ^{235}U and ^{238}U alpha energy peaks.

The two methods used in this work can be applied to calculate the uranium enrichment, but a detector with better resolution and efficiency is required.

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