Journal of Physics & Astronomy

Research | Vol 5 Iss 1

Calculation of Activity for Irradiated 110 mAg Foil Using the Los Alamos MCNP5 Code

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Received: January04, 2017; **Accepted:** January05, 2017; **Published:** February06, 2017

Abstract

The main object of this study is the simulation, using the MCNP5 code, of the irradiator 226Ra-Be unit which is available at the physics department of the sciences faculty, Damascus University to calculate the neutron fluxes. The simulation results showed that neutron fluxes, in the energy ranges: thermal (10.9 MeV to 10.6 MeV), fast (0.11 MeV to 12.0 MeV), and medium (10.5 MeV to 10.1 MeV). Where flux percent was about (thermal 70.0%, fast 18.0% and medium 12.0%).

The calculation of activity for a silver foil, in a specific time after irradiation, was simulated and compared with the experimental results. The comparison shows good agreement between the simulated and measured activity.

Keywords: ***MCNP5; Simulation; Neutron spectrum; NAA; Foil activation; Silver 110m; Neutron flux***

Introduction

Neutron irradiation following 9Be(α,n)12C reaction (Q=5.7MeV) is the most commonly used as it gives the highest neutron yield. For many years, Radium 226, with its decay products, has been used as an alpha emitter when long-lived sources are needed. More recently, the availability of isotopes such as 239Pu, 227Ac, and 241Am has made it possible to produce neutron sources which have certain advantages, in particular a less intense gamma emission [1].

Radium 226 was the first alpha emitter used because it was well studied as a radioactive source and it was relatively plentiful compared with other high energy alpha emitting isotopes. Polonium-210, which itself is a decay product of 226Ra , was also used as an isotope in early neutron source. Another early radioactive material used in isotopic neutron source was Actinium 227 (227Ac), but because of its relative scarcity, this source was rarely used.

Citation: Altaani A, Nahili M. Calculation of Activity for Irradiated 110 mAg Foil Using the Los Alamos MCNP5 Code. J Phys Astron. 2017;5(1):105.

It was found that beryllium had the best neutron yields of the light elements, therefore, nearly all isotopic neutron sources after the 1950s were a combination of an alpha emitter and beryllium. However, some isotopic neutron sources used fluorine, boron or lithium instead of beryllium [2].

Neutron activation analysis (NAA), discovered in 1936, is an important technique for quantitative multi-element analysis of major, minor, trace, and rare elements. The initial step in neutron activation analysis is irradiating a sample with neutrons in a nuclear reactor or sometimes in other neutron sources. The stable nucleus absorbs one neutron and becomes a radioactive nucleus. The concentration of the stable element of interest in the sample can be measured by detecting the decay of these nuclei.

The radioactive nuclei emit characteristic gamma rays. Detection of the specific gamma rays (of specific energy) indicates presence of a particular element. Suitable semiconductor radiation detectors may be used for quantitative measurement. The concentrations of various component elements in given samples are found by computer data reduction of gamma ray spectra. Sequential instrumental neutron activation analysis allows quantitative measurement of up to about 35 elements in small samples of 5 mg to 100 mg. The lower detection limit is in parts per million or parts per billion, depending on the element [3]. An illustration in the case of a neutron capture reaction is depicted in FIG. 1 [4].

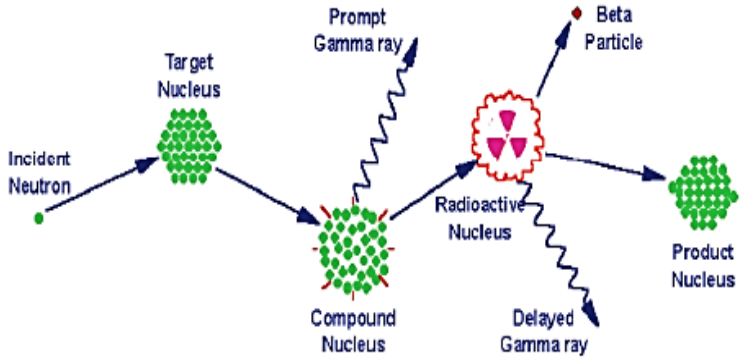

FIG. 1. **Diagram illustrating the neutron capture by a target nucleus followed by the emission of gamma rays.**

The aim of this work is 1) to calculate the neutron fluxes in the channels of 226Ra-Be irradiator unit by using MCNP5 code, and 2) to calculate the activity of a silver foil and compare it with the values measured using a neutron activation dosimeter.

Materials and Methods

Description of 226Ra-Be irradiator unit

The irradiator 226Ra-Be unit (PHYWEB-edienungsanleitung-neutronenquelle 3.5 mCi-09080.01) is available at the Physics Department of the Sciences Faculty, Damascus University. It consists of:

The container: It was made of steel (thickness 4 mm) in the form of a parallelepiped of dimensions 50 × 50 × 60 cm^3 and covered with a rectangular steel cover. The container contains a moderator of paraffin (density 0.904 g/cm^3). There are ten cylindrical channels for irradiation (for each channel: thickness 1 mm, diameter 2.2 cm). There is also a gap to insert a cadmium plate, which is used as an absorber of thermal neutrons.

Five channels (4,5,6,7,8) are distributed on the circumference of a circle of radius 10 cm around the source (226Ra-Be). The other five channels are located differently away from the source at 15 cm (channel 9), 20 cm (channels 11 and 12), and 25 cm (channels 10 and 13), as shown in the TABLE 1 and FIG. 2 [5].

Name	Shape	Dimensions	Notices
Steel container	Cubic	$60 \times 70 \times 50$ cm^3	Contain paraffin moderator
Cadmium	Plate	$40 \times 10 \times 0.2$ cm^3	On Ox axis
^{226}Ra-Be source	Cylindrical tube	7×2 cm^2	-

TABLE 1. **Some general properties for neutron irradiator 226Ra-Be**.

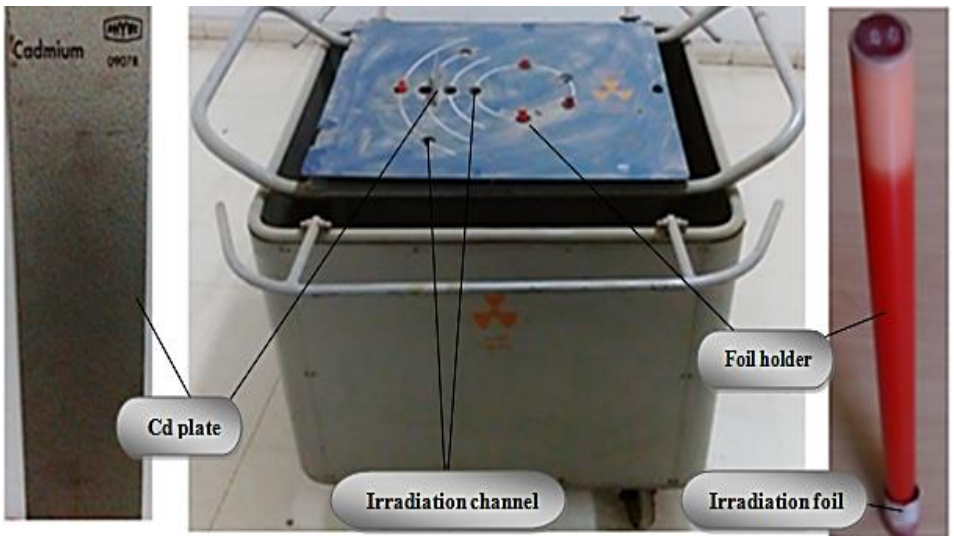

FIG. 2. **Neutron irradiator (226Ra-Be) unit (PHYWE).**

Radium-Beryllium Source: The ^{226}Ra-Be neutron source is a homogeneous mechanical mixture of an α-emitting nuclide Ra with light element Be. The mixture ratio of Be:Ra is 1:5. The weight of the mixture is 3.5 mg. It is enclosed in a two-wall cylindrical tube from nickel then steel with length of 7 cm and outer diameter of 2 cm. The two-wall cylinder is placed in a cylinder from lead with length of 7 cm and diameter of 4 cm. The flux rate of this source is up to 9.09×10^4 n/s, in view of the fact the decay of ^{226}Ra leads to the alpha-emitting progeny ^{222}Rn and ^{210}Po which produce alpha, and these particles contribute with total product neutron by ratio 6/7≈0.86. The yield of the source ^{226}Ra-Be may reach 2.0×10^7 n/s for each 1 Ci from Radium. The source ^{226}Ra-Be to be distinguished by continuous spectra of neutron with an average from 4 MeV to 5 MeV.

The relatively long-lived Radium 226, with its decay products, form a group of five α-emitting isotopes with energies ranging from 4.8 MeV to 7.7 MeV and an average energy of 5.8 MeV. These energies are enough for surmounting the

potential barrier for nuclei of beryllium which is 4.0 MeV approximately. However, alpha particles interact with the atomic electrons of beryllium, so that, they lose a part of their energy and slow down to below 4.0 MeV. Therefore, not all alpha particles can excite the nuclear interaction (α,n) in beryllium, only (1 to 1.5) × 10^4 particles can penetrate to beryllium nuclei [6].

Activation of silver

Naturally occurring silver is composed of two isotopes, ^{107}Ag, which is 51.82% abundant, and ^{109}Ag, which is 48.18% abundant. The extent to which a nucleus interacts with an incident particle may be described in terms of a capture cross section. That is, an incident particle coming within the area surrounding the nucleus will be captured. This distance is referred to as the impact parameter. The thermal-neutron capture cross section of ^{109}Ag has been measured to be 82 b (a barn is 10^{-24} square centimeters); When ^{109}Ag captures a neutron, it is converted to ^{110}Ag which decays principally to ^{111}Cd by the emission of a beta particle. These reactions are summarized below:

$$^{109}_{47}Ag + ^{1}_{0}n \rightarrow ^{110}_{47}Ag \rightarrow ^{110}_{48}Cd + ^{0}_{-1}\beta \quad (1)$$

In addition to the reactions described above, there is a small probability that isomeric states of ^{111}Ag are formed. This is the Great nuclear energy states, which are relatively stable. Maybe also decay by issuing beta particles, but the long half-lives, which can contribute very small amount by the remarkable activity [7].

The irradiation target, a high purity (99.97%) Ag foil, is illustrated in FIG. 3. TABLE 2 summarizes the characteristics of the sample used in the neutron activation experiment.

TABLE 2. **Characteristics of samples.**

Material	**Physical form**	**mass (g)**	**Diameter (mm)**	**Thickness (mm)**	**Height (mm)**	**Reaction of interest**
Ag	Pure cylinder	10.96	1.8	2	24	$^{109}Ag(n,\gamma)^{110m}Ag$

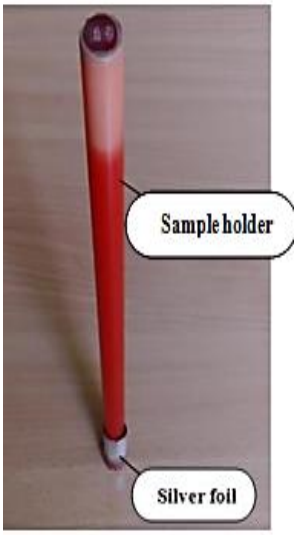

FIG. 3. **Ag target.**

FIG. 4 shows the partial decay scheme of ^{110m}Ag. The ^{nat}Ag foils were irradiated by thermal irradiator neutrons, and then adequate amounts of ^{110m}Ag nuclei were produced. The amounts of produced ^{110m}Ag nuclei were estimated by measuring the γ-rays they emitted. The yields of the γ-rays emitted from the irradiated targets were measured using a high purity Ge detector with a relative efficiency 80% [8].

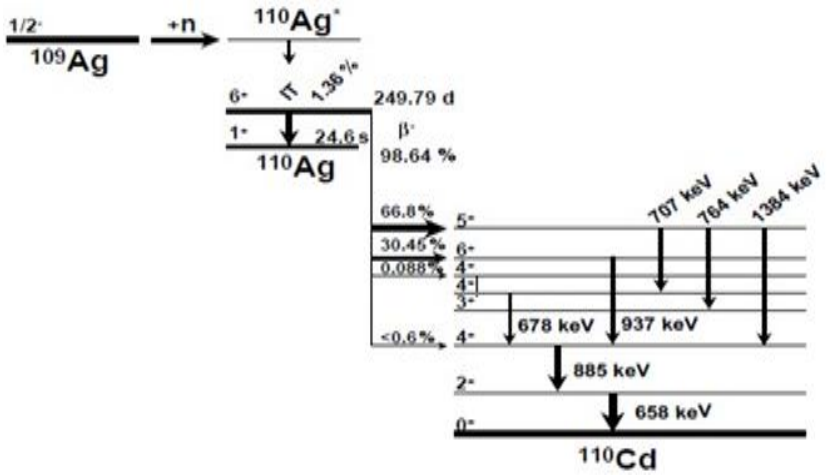

FIG. 4. **Partial decay scheme of 110 mAg.**

The ^{110m}Ag data used for the present analysis are listed in TABLE. 3.

TABLE 3. **Nuclear data used in this analysis.**

Nuclide	**Half-Life**	**Cross section(b)**	**Detected γ-rays**	
			Energy(MeV)	**Emission probability(%)**
^{110m}Ag	249.79d	82	0.658	94.0
			0.678	10.3
			0.885	72.2

Simulation of ^{226}Ra-Be unit using MCNP code

MCNP5 is a general-purpose , three dimensional general geometry, time-dependent Monte Carlo N-Particle code that can be used for neutron, photon, electron, or coupled neutron, photon, and electron transport. It is widely used around the word for many radiation protection and shielding applications [5]. The new version of the MCNP code contains the latest cross-sectional data and is able to tally the neutron flux, activation, and radiation dose based on user-defined source/moderator/reflector/shielding geometry and composition [9].

The neutron source ^{226}Ra-Be was simulated as a point source located at the center of coordinates (0,0,0). The source definition card (SDEF) was used to describe the source ^{226}Ra-Be. The neutron spectrum of ^{226}Ra-Be was used from literature

[1]. The container, around the source, was filled with paraffin except in the defined channels (FIG. 5). In addition, TABLE 4 shows the physical properties of irradiator as used in simulation by MCNP5 code.

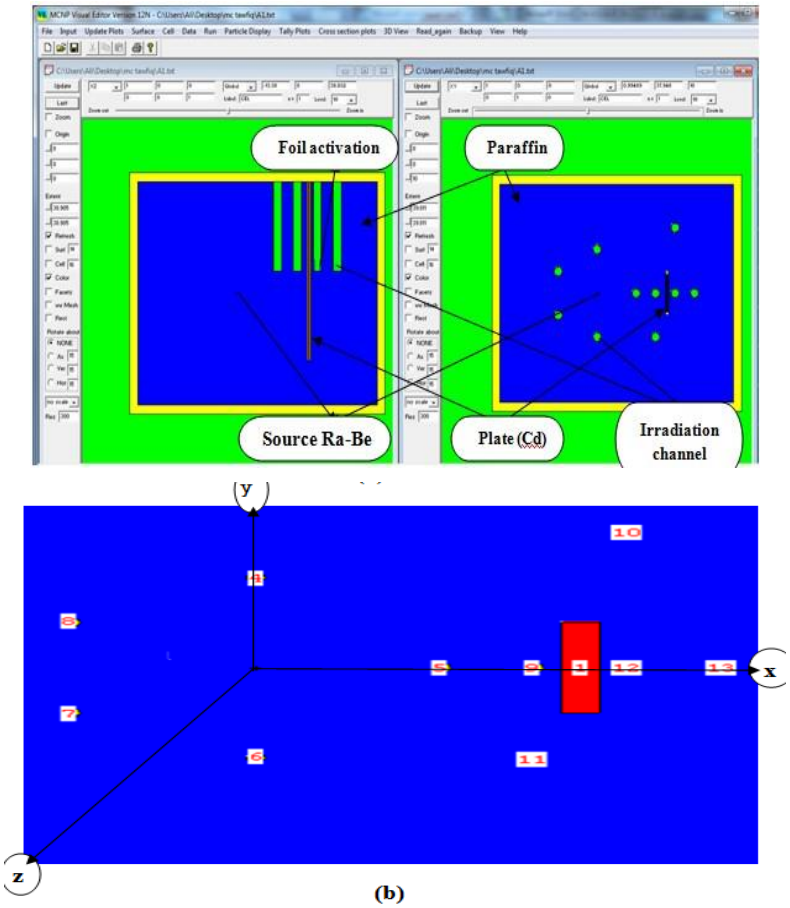

FIG. 5. **a) Diagram for the ^{226}Ra-Be irradiator unit using Vized program, b) Irradiation channels and Cadmium plate.**

TABLE 4. **Physical properties of irradiator as used in simulation by MCNP5 code.**

Name	**Shape**	**Position relative to ^{226}Ra-Be (cm)**	**Notice**
Channel number	Irradiator channels and its diameter 2 cm (according its number in simulation)		
4	Cylinder	10	On axis Oy
5	Cylinder	10	On axis Ox
6	Cylinder	-10	On axis Oy
7	Cylinder	-10	On axis xOy
8	Cylinder	-10	On axis xOy
9	Cylinder	15	On axis Ox
10	Cylinder	25	In plane xOy

11	Cylinder	20	In plane xOy
12	Cylinder	20	On axis Ox
13	Cylinder	25	On axis Ox
Materials carrier	Cylinder	radius (0.5 cm) and length (30 cm)	The length inside paraffin 20 cm
Source ^{226}Ra-Be			
^{226}Ra-BeMixture	Cylinder	The mixture is in pair-wall cylindrical tube from nickel then steel, length 7 cm, and diameter 2 cm , Ratio of ^{226}Ra to ^{9}Be: 1/5, weight: 3.5 mg.	
Cylinder of steel	Cylinder	length is 7 cm, diameter 2 cm	
Source carrier	Cylinder	Length 18 cm, diameter 4 cm	

Results and Discussions

Calculation of the neutron flux inside the irradiator channels

Using the F4card in MCNP5 (This tally allows the calculation of the flux average over a cell (particles/cm^2)), the neutron flux was calculated in different channels, whereas neutron flux is proportional to paths grand total K which have length L_k for neutrons which had energy E_j across the channels volume as illustrated in equation (2)

$$\Phi_j \propto \frac{1}{V}\sum_{k=1}^{K} L_k(E_j) \qquad (2)$$

The neutron flux Φ_j(cm^{-2}) expressed by using F4 card, as in equation (3) [9]

$$F_4 = \int_V \int_t \int_E \Phi(\vec{r},E,t)\,dE\,dt\,\frac{dV}{V} \qquad (3)$$

FIG. 6. shows the composition of the neutron flux (thermal, medium, and fast) at the channels of the irradiator unit as obtained from MCNP5 code. Obviously, the neutron flux decreases away from the source. In addition, the thermal neutron flux dominates at all channels except those which contain the Cadmium plate.

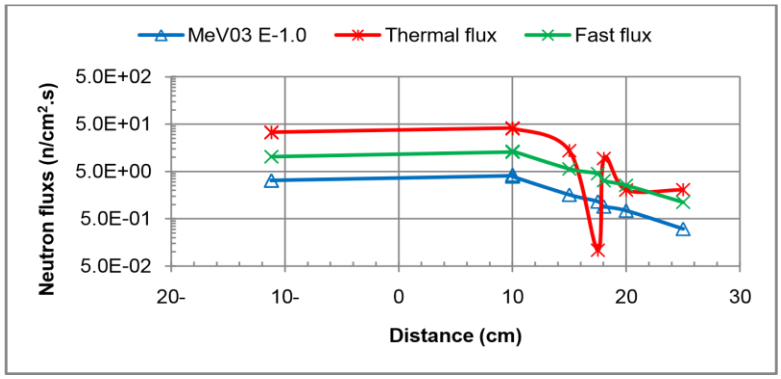

FIG. 6. **Neutron flux at irradiator channels according to their distance from the source.**

The ratio"thermal to fast":

- equals, in average, 3.17 ± 0.03 at channels 4,5,6,7, and 8.
- decreases to 0.024 ± 0.01 at Cadmium plate.
- decreases to 0.8 ± 0.02 and 1.83 ± 0.01 in channels 12 and 13, respectively, as these channels are located behind the cadmium plate.
- equals 2.41 ± 0.03, 3.01 ± 0.04, and 2.95 ± 0.02 at channels 9,10, and 11, respectively.

Calculation of the sample activity by MCNP code using FM card: This step is based on the program for the calculation of the total neutron capture reactions in the whole volume of the foil. In this program, a thermal neutron field (0.025 eV) is defined as the neutron source. The foil-source setup geometry and its construction materials are also described in material cards.

In this step, total neutron capture reactions per volume unit (in cm^{-3}) are obtained using the F4 tally together with the FM4 card (This tally allows the calculation of the reaction rate), for a bunch of neutrons (NPS card (This card refers to total histories to be run, including preceding continue-runs and the initial run)) emitted simultaneously from the neutron source. The tally output is then multiplied by the volume-value of the foil to assign the total neutron capture reactions. Tally output (M1) is commonly normalized per neutron source.

In the simulation, the cross sections are needed in the thermal energy range, using proper databases with suitable information of neutron cross sections like the ENDF Library.

Sample activity measurements by γ -spectrometry: Gamma activity of the irradiated samples was measured using a high purity Ge detector (HPGe) type EGPC 80-205-R with 80% relative efficiency and made up from EURISYS MESURES company, connected to a multi-channel pulse-height analyzer personal computer system based on the nucleus personal computer analyzer (PCA-II) card. The minimum detectable gamma ray energy of our detector configuration was 30 keV.

Radioisotopes were identified from the pulse-height spectrum by their gamma photopeak energies and half-lifes. Their activities were determined from gamma photo-peak area and detection efficiencies at the photo-peak energy. The full-energy peak efficiency for point source geometry was measured using a set of tow point source standards (Sources are ^{60}Co and ^{137}Cs, that Possess Activity1.083 µCi and 1.094 µCi respectively. Where it was calibrated on 1.09.1987).

A schematic diagram of the data acquisition system is provided in FIG. 7 [8]. The γ-ray detection efficiencies were obtained with the ^{60}Co and ^{137}Cs standardized sources. An example of theγ-ray spectrum is shown in FIG. 7. As can be seen in this figure, the γ-rays originating from ^{110m}Ag, *i.e.* 658 and 885 keVγ -rays, were clearly measured. These γ-ray yields were used to determine the activities of ^{110m}Ag produced *via* the ^{109}Ag $(n, \gamma)^{110m}$Ag reaction.

As shown in TABLE 5. the simulation results of the activity for ^{110m}Ag foil has been done well with experimental results.

TABLE. 5. **Measured and calculated activity for ^{110m}Ag foil.**

Target	Measured activity by γ spectrometry (Bq)	Calculated activity by MCNP (Bq)	Difference percentage
^{109}Ag	0.08 ± 0.02	0.063 ± 0.03	22%

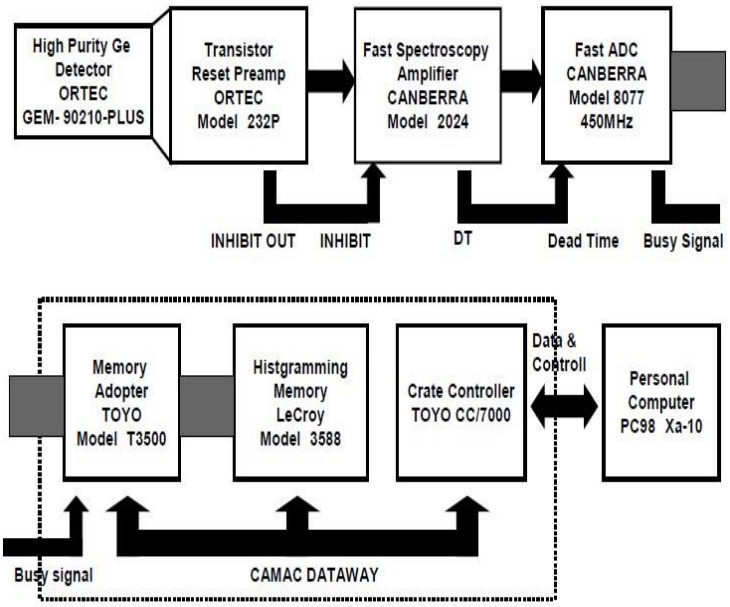

FIG. 6. **A block diagram of the electronics for the data acquisition system.**

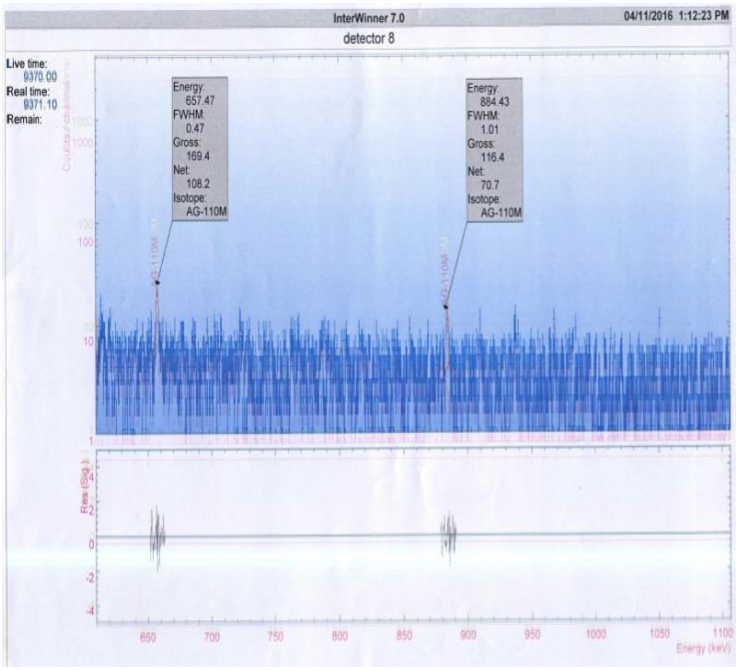

FIG. 7. **An example of the γ -ray spectra of the Ag target.**

Conclusions

The results presented in this work show that the simulation of the neutron fluxes in ^{226}Ra-Be unit is very important and useful to understand the neutron flux in each channel for neutron activation analysis. Good agreement was found between the measured and the simulated values for saturation activity per nuclei target of the ^{110m}Ag irradiated foil.

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