ISSN: 0067-2904

Activity Treatment of Some Long-Lived Radioactive Nuclides Using Thermal Neutron Incineration

Hassanain H. Alkazzaz [1]*, Asia H. Al-Mashhadani [1], Kamal H. Lateef [2]
[1]*Department of physics, College of science, University of Baghdad, Baghdad, Iraq*
[2]*Chairman of Iraqi Radioactive Sources Regulatory Authority, Baghdad, Iraq*

Received: 8/3/2022 Accepted: 18/9/2022 Published: 30/6/2023

Abstract

In the present work, the possibility of treating many types of radioactive sources was examined practically. Six types of sealed radioactive sources were selected: ^{137}Cs, ^{133}Ba, ^{90}Sr, ^{152}Eu, ^{226}Ra, and ^{241}Am. The sources were exposed to a neutron flux emitted from ^{241}Am/Be source for 33 days. The results showed a measurable reduction of activity for ^{226}Ra, ^{241}Am, and ^{152}Eu, while the other radionuclides, ^{137}Cs, ^{133}Ba, and ^{90}Sr, showed less response to neutron incineration.

Keywords: radioactive waste treatment, neutron capture, ^{137}Cs, ^{133}Ba, ^{152}Eu, ^{90}Sr, ^{226}Ra, ^{241}Am, radioactive sources incineration.

معالجة بعض النويدات المشعة ذات الاعمار النصفية الطويلة باستخدام الحرق بالنيترونات الحرارية

حسنين حكمت محمد [1]*, آسيا حميد المشهداني [1], كمال حسين لطيف [2]
[1] قسم الفيزياء, كلية العلوم, جامعة بغداد, بغداد, العراق.
[2] رئيس الهيئة العراقية للسيطرة على المصادر المشعة, بغداد, العراق.

الخلاصة

في هذا البحث، تم فحص إمكانية معالجة العديد من أنواع المصادر المشعة عمليا، حيث تم اختيار ستة أنواع من المصادر المشعة المختومة وهي ^{137}Cs، ^{133}Ba، ^{90}Sr، ^{152}Eu، ^{226}Ra و ^{241}Am. تم تعريض هذه المصادر لفيض من النيوترونات المنبعثة من المصدر المشع ^{241}Am / Be لمدة 33 يومًا ، أظهرت النتائج انخفاضًا محسوساً في النشاط الاشعاعي للنظير ^{226}Ra، ثم النظيرين ^{241}Am و ^{152}Eu، بينما أظهرت النويدات المشعة الأخرى ^{137}Cs، ^{133}Ba ، ^{90}Sr استجابة أقل للحرق النيوتروني.

1. Introduction

Most countries worldwide suffer from the problem of the accumulation of useless radioactive sealed sources of medium to long half-lives; these radioactive sources are mainly used in medical therapy devices and industrial activities. Some old technologies use radioactive sources such as in gas and oil well-logging, nuclear gauges for measuring density and porosity, lightning preventers that are still in use in some facilities, and a lot of industrial applications [1]. As a result, the accumulation of these radioactive sources causes an explicit threat to the environment and citizens, in addition to the high cost of traditional treatment and long-term

*Email: hassanainalkazzaz@yahoo.com

waste management. Thus, there is a need for ways of treatment based on destroying the long-lived radioactive nuclides of the waste.

Many methods can be used to remediate the radioactive waste produced as fission fragments from nuclear power plant activities or the accumulation of useless radioactive sources; one of these methods is burning by thermal neutrons [2].

The use of neutrons to reduce radioactivity or radiotoxicity of long-lived radioactive elements is a very promising method and gives good results for the near future. The cost of producing thermal neutrons with a high neutron flux is relatively low compared with the cost of producing protons and gamma photons [3]. Nuclear reactors that generate electrical energy can be used for the purpose of nuclear waste transmutation. Neutron generators can be used as a source of high flux neutrons; it is considered as a clean source of neutrons in terms of the generating mechanism; the generated neutrons lunch only when supplying an electric current to accelerate deuteron ions to about 300 keV to collide with a tritium target to produce a nearly-isotropic and mono-kinetic 14 MeV neutron output via the T(d,n)α fusion reaction [4]. So, it is preferred as a clean source of neutrons alternative to nuclear reactors that generate a significant amount of radioactive waste.

Neutrons are nuclear particles with a neutral charge. They interact with different nuclei easily and with relatively low energy if compared with nuclear-charged particles of equal mass and energy. Usually, most charged particles need to be accelerated to a relatively high energy to overcome the target nucleus barrier and conduct the reaction, but in neutron bombardment, the beam should be moderated to an epithermal level to get a giant increment in reaction cross-section.

This method of using neutrons in incineration and/or transmutation of long-lived radionuclides has first been reported by Steinberg et al. [5] as a possible method for the disposal of radioactive wastes. Taube et al. [6] studied the possibilities for the transmutation of ^{137}Cs and ^{90}Sr in a high-flux fast reactor; they found it can decrease the half-lives of cesium from 30 years to about 4 – 5 years, and 2 – 3 years for strontium. Fioni et al. [7] conducted an experimental study of incinerating ^{241}Am radionuclide using a high-intensity neutron flux reactor; they performed about a 46% reduction of the initial amount of ^{241}Am by neutron capture and fission.

2. Sources of neutrons

Generally, neutrons can be generated by many methods: binary reaction such as (α, n) and (γ, n), spontaneous fission such as ^{252}Cf, fission reactor by (n, xn'), and accelerator like T(d, n) and D(d, n) fusion [8].

In a laboratory-scale experiment, it is recommended to use alpha emitter sources to generate neutron flux such as ^{241}Am/^{9}Be, ^{238}Pu/^{9}Be, ^{239}Pu/^{9}Be, ^{210}Po/^{9}Be and ^{226}Ra/^{9}Be. The yield of neutrons and mean energy differ depending on the energy of the emitted alpha and the reaction mechanism. Concerning the ^{241}Am/^{9}Be source, reaction equations are described as follows[9]:

$$^{241}_{95}\mathrm{Am} \rightarrow {}^{237}_{93}\mathrm{Np} + {}^{4}_{2}\alpha^{++}(5486\ \mathrm{keV}) + \gamma(59.5\ \mathrm{keV})$$

$$^{9}_{4}\mathrm{Be} + {}^{4}_{2}\alpha^{++} \rightarrow {}^{12}_{6}\mathrm{C} + {}^{1}_{0}\boldsymbol{n} + \gamma$$

The half-life of the alpha source (^{241}Am) is about 432.2 years, the yield of neutrons is distributed with a continuous energy spectrum starting from slow neutrons up to 11 MeV maximum energy, with an average neutron energy of 4.2MeV. The yield neutron spectrum from

(Am/Be) source was measured by Vijaya and Kumar [10] using data on angular distribution; they introduced a theoretical model of relative neutron intensity for 1 Ci activity of Americium. Marsh et al. [11] performed high-resolution measurements of neutron energy spectra of Am/Be and Am/B sources in the energy range 100 keV to 15 MeV. The measured spectrum showed multi-peaks at 2.25, 3.2, 5.0, 6.5, 7.7 and 9.9 MeV. Figure (1) shows the measured spectrum by Vijayaand Kumar [10] and Marsh et al. [11].

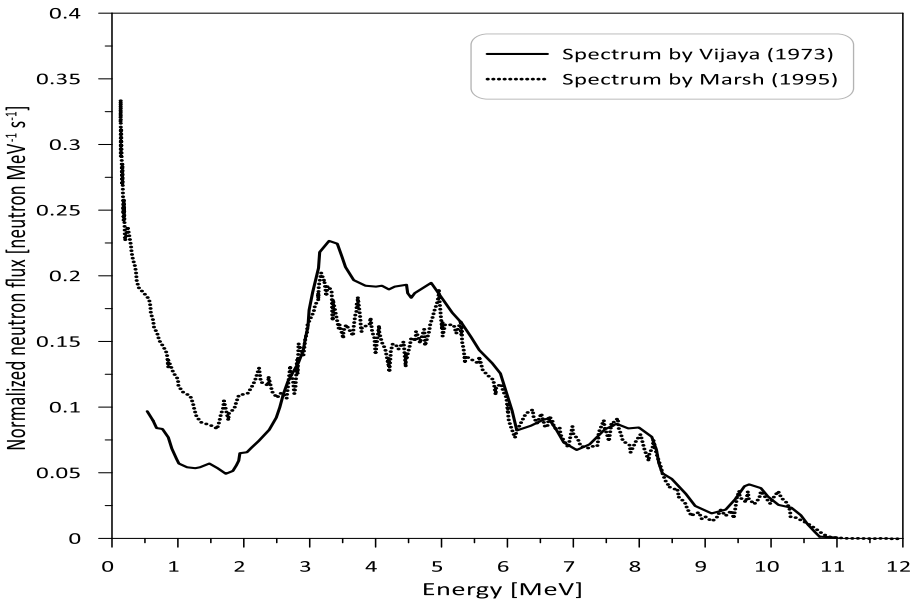


Figure 1: Neutron spectrum emitted by ^{241}Am/^{9}Be source of 370 GBq activity normalized to unity, differential flux data are taken from Vijaya and Kumar [10] and Marsh et al. [11].

3. Transmutation and incineration:

In this paper, six radionuclides were considered for this study, ^{137}Cs, ^{133}Ba, ^{90}Sr, ^{152}Eu, ^{226}Ra, and ^{241}Am. These radionuclides are widely used in medical and industrial applications worldwide; however, ^{60}Co is also used in widespread application but it is of shorter half life (about 5.27 years). Table (1) illustrates the general information of these radionuclides.

Through the use of neutron incineration, the transmutation rate depends on the integration of multiplication of neutron flux density by the absorption cross-sections of neutrons over the energy spectrum. During irradiation interval, the change rate of the number of nuclides $\boldsymbol{i^{th}}$ that is exposed to the external neutron of flux density $\boldsymbol{\phi}$ is given by a linear system of differential equation as follow [13]:

Table 1: Information about the radionuclides under study

Radionuclide	Half-life [Years]	Decay mode	Specific activity [Bq/g]	Significant Application [12]
^{137}Cs	30.08 year	β^- 100% prompt γ : 661.9 keV	3.21x10^{12}	Irradiators: sterilization and food preservation, Teletherapy, density gauges.
^{133}Ba	10.551 years	EC 100%	9.43x10^{12}	Calibration, X-ray production
^{90}Sr	28.79 years	β^- 100%	5.11x10^{12}	Radioisotopic Thermoelectric generators (RTGs)
^{152}Eu	13.517 years	EC, β^+ 72.08% β^- 27.92%	6.44x10^{12}	Lightning preventers

^{226}Ra	1600 years	α - 100%	3.66x10^{10}	Lightning preventers, emergency signs
^{241}Am	432.6 years	α - 100%	1.27x10^{11}	Well logging, porosity gauges

$$\frac{dN_i}{dt} = \textbf{production rate} - \textbf{destruction rate} - \textbf{decay rate}, \left(i = 1,2,\ldots n\right) \quad (1)$$

so that

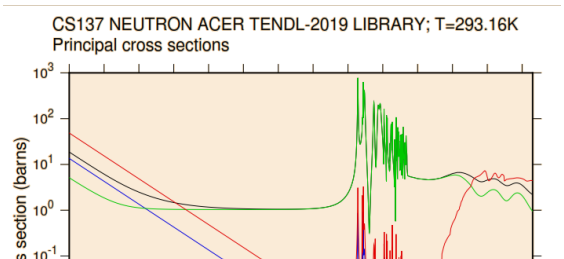
$$\frac{dN_i}{dt} = \sum_{i \neq j}\left[\left(\gamma_{j\to i}\sigma_{f,j}\phi + b_{j\to i}\lambda_j + \sigma_{j\to i}\phi\right)N_j\right] - \left(\lambda_i + \sum_{i\neq j}\sigma_{i\to j}\cdot\phi\right)N_i \quad (2)$$

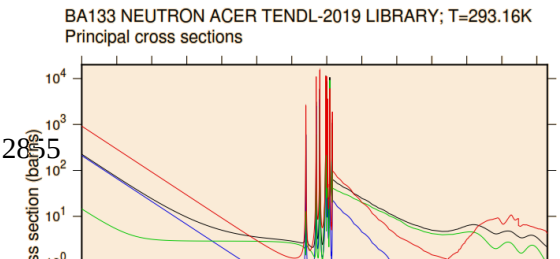
Where:
N_j is the atomic density of the nuclide i [cm^{-3}];
N_j is the atomic density of the nuclide j [cm^{-3}];
$\sigma_{f,j}$ is the spectrum-integrated microscopic fission cross-section of nuclide j [cm^2];
$\sigma_{j\to i}$ is the spectrum-integrated microscopic of transmutation cross-section of reaction $j \to i$ [cm^2];
λ_i is the decay constant of isotope i [s^{-1}];
λ_j is the decay constant of isotope j [s^{-1}];
$b_{j\to i}$ is the branching ratio for a specific decay of nuclide i from nuclide j ;
$\gamma_{j\to i}$ is the fractional fission product yield of nuclide i from the fission of nuclide j ;
ϕ is the spectrum-integrated neutron flux [cm^{-2} s^{-1}].

The product ($\sigma_{i\to j} . \phi$) represents the reaction rate of i^{th} nuclide producing j^{th} nuclide, since both neutron flux and reaction cross-section are energy-dependent. The cross-section of the reaction varies rapidly across neutron spread energy, so that the average cross-section $\langle\sigma_{i\to j}\rangle$ could be taken from the equation [14]:

(3)

The effectiveness of the incineration system depends mainly on the neutron spectrum. Usually, reaction cross-sections of most nuclides are very large near epithermal neutron energy in which the giant resonance of absorption is located. Reaction cross-section decreases roughly as the energy of neutron increases. Figure (2) shows the neutron elastic, inelastic and total cross-section for the radionuclides listed in Table (1), noting that total cross-section is the sum of elastic and inelastic (absorption) cross section [15].





(a) (b)

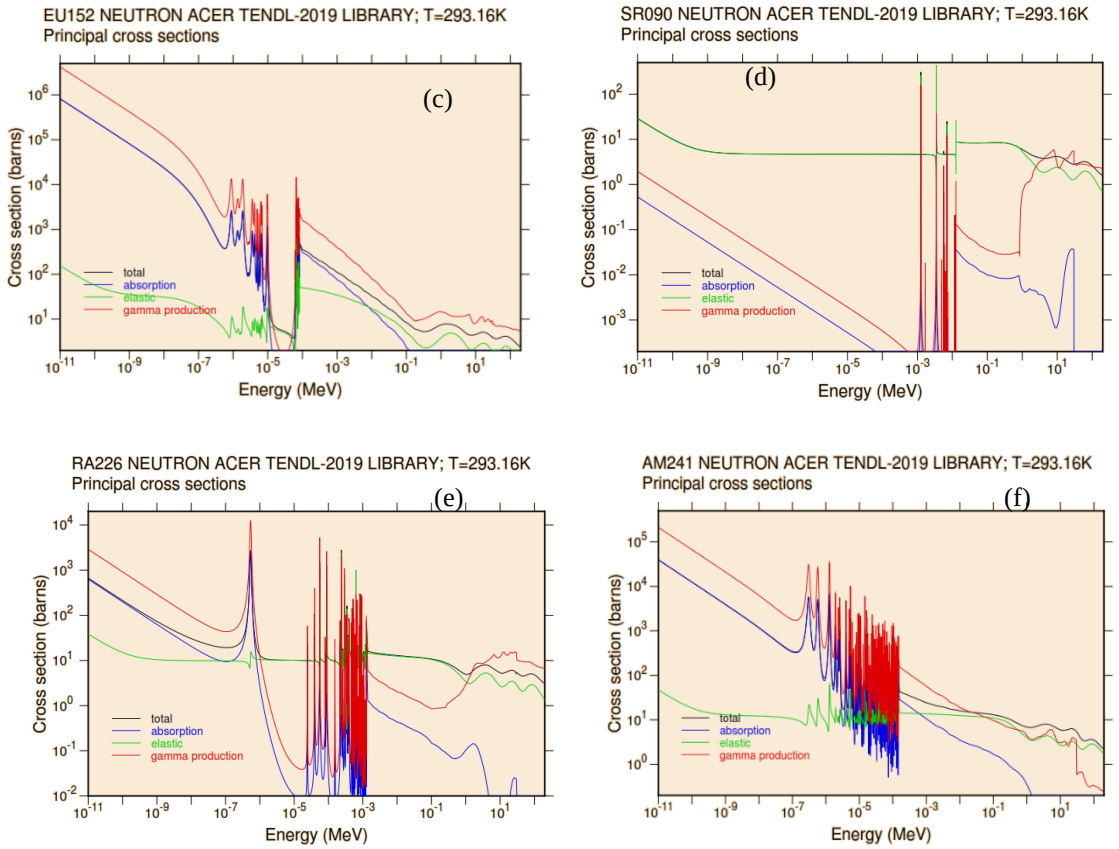


Figure 2: Neutron cross section of (a) ^{137}Cs, (b) ^{133}Ba, (c) ^{90}Sr, (d) ^{152}Eu, (e) ^{226}Ra, and (f) ^{241}Am radionuclides, figures taken from TENDL-nuclear data library [15].

4. Experimental method:

Six sealed radioactive sources (^{137}Cs, ^{133}Ba, ^{90}Sr, ^{152}Eu, ^{226}Ra, and ^{241}Am) of low activity (1 μCi – 5 μCi) were selected for this study, as shown in Figure (3). The activity of the radioactive sources was characterized experimentally using a high purity germanium detector. The system of (Falcon 5000 – 20) by CANBERRA was used, (Falcon 5000 – 20) assembly contains high purity germanium detector (type: BE2830 Φ60x30 mm) with a self-cooling system using pulse tube cooler technology; it cools down the germanium crystal from room temperature to –170◦ C in about 3 hours, no liquid nitrogen is required for cooling.

The (Falcon 5000-20) system was configured in a suitable arrangement by constructing lead shied blocks as a cavity to reduce background radiation noise as shown in Figure (4). The radioactive sources were loaded using a detector-face mask holder, and the distance from source to detector face was 115 mm, time of acquisition was 300 second. Figure (5) shows the setup

of the high purity germanium system with lead shield blocks enclosure for background radiation isolation and the sample radionuclide under measurement.

Figure 3: Sealed radioactive sources under study.

Figure 4: Full setup of Falcon5000-20 unit with lead shield cavity.

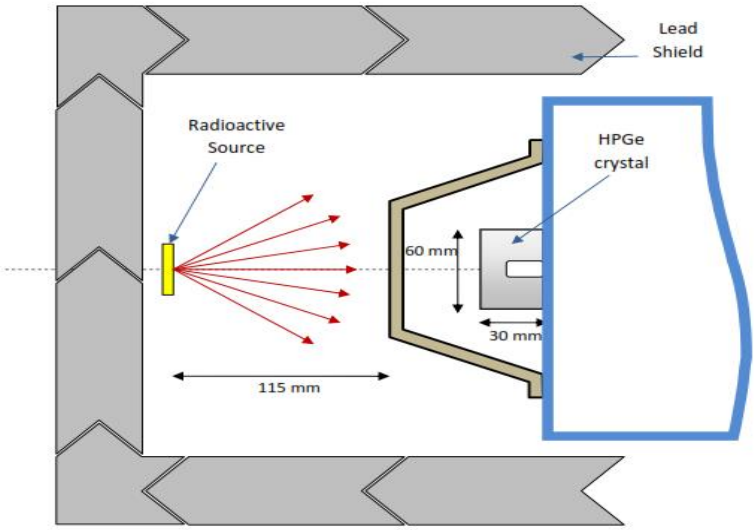


Figure 5: The geometry configuration of sample radionuclide and HPGe detector with lead wall.

A full gamma spectrum was registed to each source before and after irradiation by neutrons through spectrometry measurements. A set of energy peaks and the area under each peak were measured using Genie2000 and FitzPeaks V3.71 software; also, the software provides values of uncertainty through internal calculation algorithms.

Falcon (5000-20) unit has factory calibration sets of efficiency using four mathematical models: dual, linear, empirical, and interpolation fit; the first three models use modified polynomials of the fifth-order, the last model uses cubic spline interpolation and satisfies all sample points. These models were extracted and embedded in MATLAB script to calculate the actual activity using the following efficiency equation [16]:

$$A = \frac{N_{net}}{\varepsilon_{abs} \cdot I_{\gamma} \cdot t} \quad (4)$$

Where:
A is the activity of source,
N_{net} is the net area of total counts under the photo-peak by subtracting background counts,
I_γ is the branching ratio of particular gamma emission energy taken from the decay scheme of the radionuclide.
t is the acquisition time.
ε_{abs} is the absolute efficiency of detector given by the following equation:

$$\varepsilon_{abs} = \varepsilon_{int} \cdot G = \varepsilon_{int} \cdot \frac{\Omega}{4\pi} = \varepsilon_{int} \cdot \frac{r^2}{4d^2} \quad (5)$$

Where:
ε_{int} is the intrinsic efficiency of the detector,
G is the geometry factor of the source with the detector,
r is the diameter of the detector crystal facing the source, and
d is the distance from source to detector.

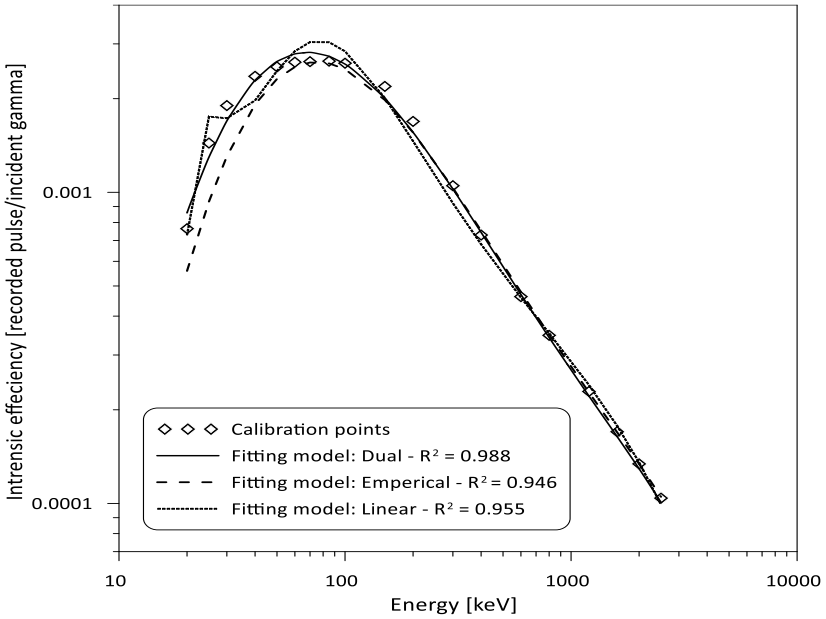


Figure 6: Intrinsic efficiency calibration curve for Falcon 5000 – HPGe detector.

The calibration data provided by factory was employed by Jasim et al. [17] to characterize radioactive waste drum, by applying the regression analysis ($\mathbf{R}^2$) on fitting models. It is clear that the highest regression value was for the dual model fitting; so it was used for estimating activity driven through the photo-peak area. Figure (6) shows the efficiency curve with the three fitting models applied by the factory.

The samples (radioactive sources) were exposed to the neutron source type $^{241}Am/^{9}Be$ of 12Ci (444 GBq) activity, as shown in Figure (7), for a period of 33 days. Typically, this type of source provides a yield of $2.2x10^6$ neutrons per second per 1 Ci [8], so the expected total neutron flux of the source is about $2.64x10^7$ neutron per second distributed isotropically. The sample sources were put in six containers of 50 ml capacity which were arranged in radial form around

a cylindrical wall of 5 cm thickness made of paraffin. The irradiative source was dropped down in the central cavity of the paraffin cylinder wall, as shown in Figure (8).

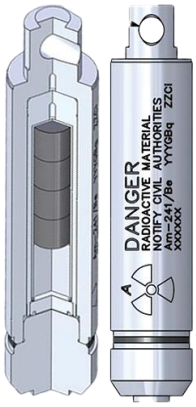


Figure 7: The $^{241}Am/^{9}Be$ source of activity 12 Ci which used as a neutron source.

Figure 8: The arrangement of samples in radial form around the neutron moderator shield.

5. Calculation method:

Estimating the real-time of irradiation is very important to ensure the precision of incineration rate calculation. the moment of pre-spectrometry measurement before irradiation is denoted as (t_0), the moment of starting incineration by neutron is (t_1), the moment of finishing incineration is (t_2) and the final spectrometry measurement moment is (t_3).

For non-fissionable nuclides, the differential Equation (2) can be re-written as [10]

$$\frac{dN_i}{dt} = -\left(\lambda_i + \sum_{i \neq j} \sigma_{i \to j} \cdot \phi\right) N_i \tag{6}$$

$$\frac{dN_i}{N_i} = -\left(\lambda_i + \sum_{i \neq j} \sigma_{i \to j} \cdot \phi\right) dt \tag{7}$$

Three intervals can be considered, the first interval ($t_0 \rightarrow t_1$) is a natural decay time only, the second interval ($t_1 \rightarrow t_2$) is a reaction with a neutron in addition to natural decay, and the last interval ($t_2 \rightarrow t_3$) is the cool down interval after irradiation. By integrating Equation (7) over time:

$$\int_{N_0}^{N_3} \frac{dN_i}{N_i} = -\int_{t_0}^{t_1} \lambda_i \, dt - \int_{t_1}^{t_2} \left(\lambda_i + \sigma_{absorption} \cdot \phi\right) dt - \int_{t_2}^{t_3} \lambda_i \, dt \tag{8}$$

Where $\sigma_{absorption}$ is the sum of individual channels cross-sections, integrating Equation (8):

$$N_3 = N_0 \cdot e^{-\lambda_i (t_3 - t_0)} \cdot e^{-\sigma_{absorption} \cdot \phi \, (t_2 - t_1)} \tag{9}$$

Multiplying Equation (9) by decay constant λ_i of nuclide i, it becomes:

$$A_3 = A_0 \cdot e^{-\lambda_i (t_1 - t_0)} \cdot e^{-\sigma_{absorption} \cdot \phi \, (t_f - t_i)} \tag{10}$$

Where: A_0, A_3 is the exclusive activity of the ith nuclide measured at time t_0 and t_3, respectively, the coefficient ($\sigma_{absorption} \cdot \phi$) in the second term represents the effective reaction rate [sec^{-1}] of neutrons with the radioactive substance (reaction probability per unit time per

nuclide). Through the calculation of activity for each photo-peak using Equations (4) & (5), the average activity of multi-peak radionuclide can be calculated by weighting each acceptable peak activity by its respective branching ratio as follows [18]:

$$A_{avg} = \frac{\sum_{i=1}^{n} A_{Ei} \cdot I_{\gamma i}}{\sum_{i=1}^{n} I_{\gamma i}} \tag{11}$$

Where: A_{Ei} is the activity of nuclide peak i at energy E_i, $I_{\gamma i}$ is branching ratio of the peak at energy E_i or number of gamma emmision of energy E per 100 disintegration, and n is the number of peaks included in the activity.

The total activity A_{total} is equal to the sum of averge activityies $A_{avg(i)}$ of all nuclides, and is given by:

$$A_{total} = \sum_{i=1}^{all\ nuclides} A_{avg(i)} \tag{12}$$

6. Results and discussion:

Activities of the radioactive sources were measured precisely using photo-peaks of the radionuclides understudy. A complete area under the peak from spectrum analysis report was used after excluding the background noise for all significant weighted peaks using Equations (4,5, and 11). Figures (9a & 9b) show the average value of activity for multi-energy nuclide ^{133}Ba and ^{152}Eu. The resulting activity may then be used to calculate the reaction rate parameter of each nuclide using Equation (10) as follows:

From equation (10), the reaction rate per second per nuclide can be calculated as:

$$\text{Reaction Rate} = \sigma_{absorption}\phi_{neutron} = \frac{ln(A_0) - ln(A_3) - \lambda(t_3 - t_0)}{t_2 - t_1} \tag{13}$$

Where: A_0, A_3 are the exclusive activity of the i [th] nuclide measured at time t_0 and t_3 respectively, t_1, t_2 are the time of starting and ending the irradiation measured to a reference date. Table (2) shows the experimental measurement of reaction rate using Equation (13).

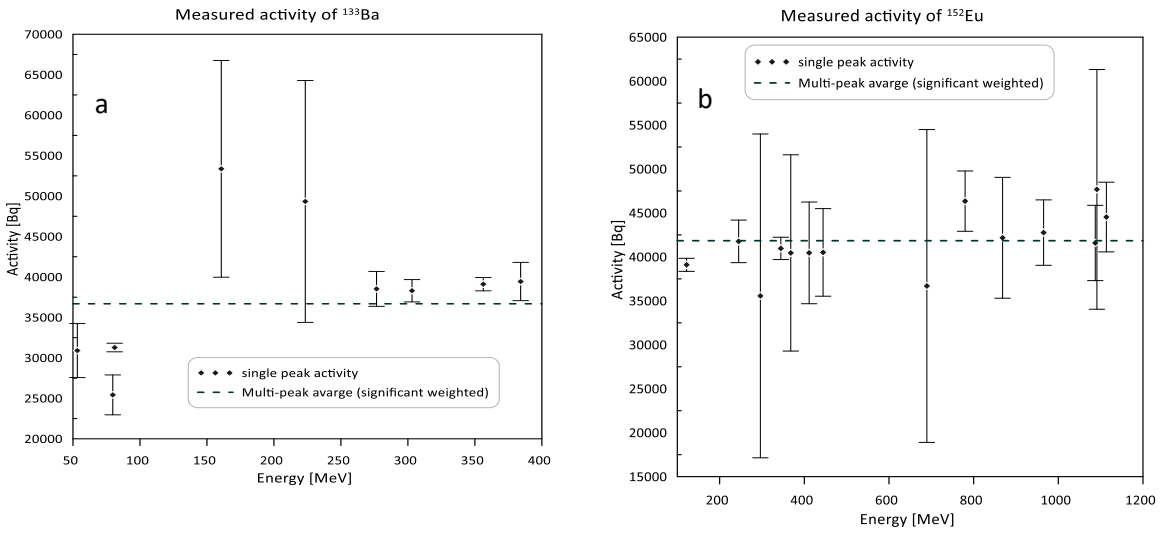
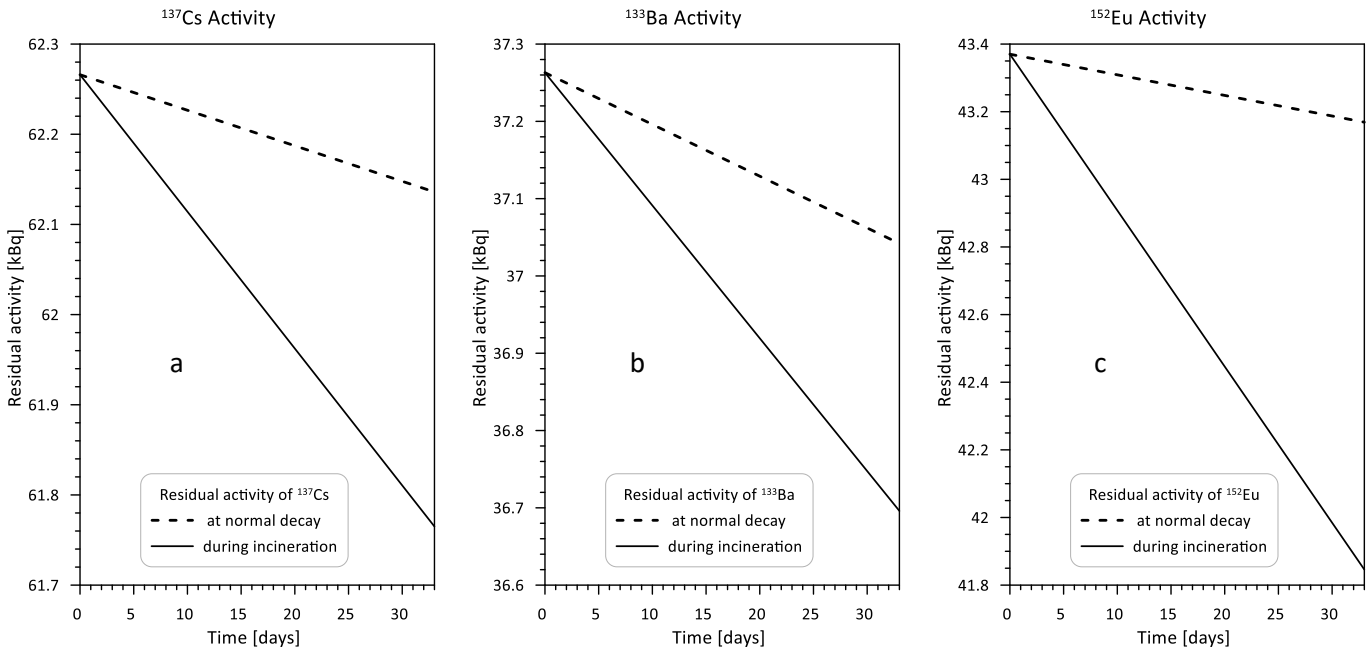


Figure 9: Average activity for multi-peaks radionuclide (a) ^{133}Ba, and (b) ^{152}Eu.

Table 2: The experimental measurement of activities and calculated reaction rate in s^{-1} , decay constant of natural decay in s^{-1} also listed table.

Nuclide	Activity at starting time [kBq]	Activity at ending time [kBq]	Decay constant [sec^{-1}]	Reaction rate measured [sec^{-1}]	Reaction rate to decay constant ratio	Thermal neutron cross-section [barn] [19]	Resonance integral cross section [barn]
^{137}Cs	62.266	61.765	$7.307x10^{-10}$	$2.1036x10^{-9}$	2.8789	0.27	0.35
^{133}Ba	37.263	36.696	$2.0933x10^{-9}$	$3.2889x10^{-9}$	1.5712	4.2	110
^{152}Eu	43.370	41.846	$1.6281x10^{-9}$	$1.0913x10^{-8}$	6.7031	12800	1580
^{226}Ra	371.315	348.106	$1.3737x10^{-11}$	$2.2623x10^{-8}$	1646.8	12.8	280
^{241}Am	195.722	187.139	$5.0855x10^{-11}$	$1.5676x10^{-8}$	308	587	1425

From Table (2), it is clear that there was a significant drop in the activity measured before and that measured after exposure to neutrons. Figure (10) show the effect of neutrons incineration on samples activity. The calculated reaction-rate [s^{-1}] and natural decay constant [s^{-1}] are shown in a histogram in Figure (11). The total reduction of the original radionuclides was at maximum for ^{226}Ra, and at minimum for ^{137}Cs. The ratio of reaction rate to decay constant listed in Table (2) show that Radium 226 has the best results (more than 1600 time), while Barium 133 gave a ratio of about 1.5 times. Note that ^{90}Sr radionuclide has been removed from Table (2) because it is a pure beta emitter (no gamma peaks can be seen in gamma spectroscopy), so the measurement of a beta particle depends on the total count of the continuous spectrum of emitted electron/positron that considers as un-accurate manner because of interference with the buildup daughter ^{90}Y nuclide which also a beta emitter.



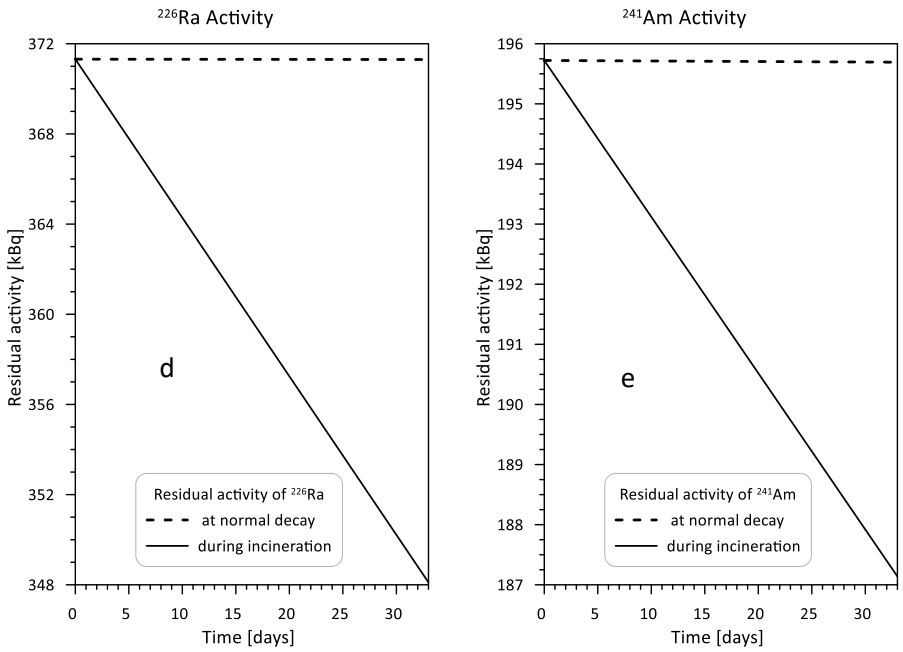


Figure 10:. Activity drop of (a) ^{137}Cs, (b) ^{133}Ba, (c) ^{152}Eu, (d) ^{226}Ra, and (e) ^{241}Am radionuclides during incineration period of (33 days), while dash-line represent the activity drop by natural decay only.

Table 3: The reaction threshold [keV] of the binary channels of neutron – gamma, double neutron, proton, deuteron, tritium, and alpha particles with reaction cross-section [mbarn] of a thermal neutron (inside brackets)

Nuclide	(n,γ) Threshold (Cross-section)	(n,2n) Threshold (Cross-section)	(n,p) Threshold (Cross-section)	(n,d) Threshold (Cross-section)	(n,t) Threshold (Cross-section)	(n,α) Threshold (Cross-section)
^{137}Cs	0 (270)	8339	3404.8	5219	7063	0
^{133}Ba	0 (4200)	7244.5	0	5506.8	6422	0
^{90}Sr	0 (10.4)	7900	5868	9407	10335	0
^{152}Eu	0 (6E7)	6348.60	0 (41.1)	3398.8	2733.6	0 (25.8)
^{226}Ra	0 (12800)	6425	3084	5240	4981	0 (3.2E-8)
^{241}Am*	0 (7.9E5)	6675	0	2264.83	2543.0	0 (2.4E-4)

* Americium is fissionable nuclide with a fission cross-section (3200 mb).

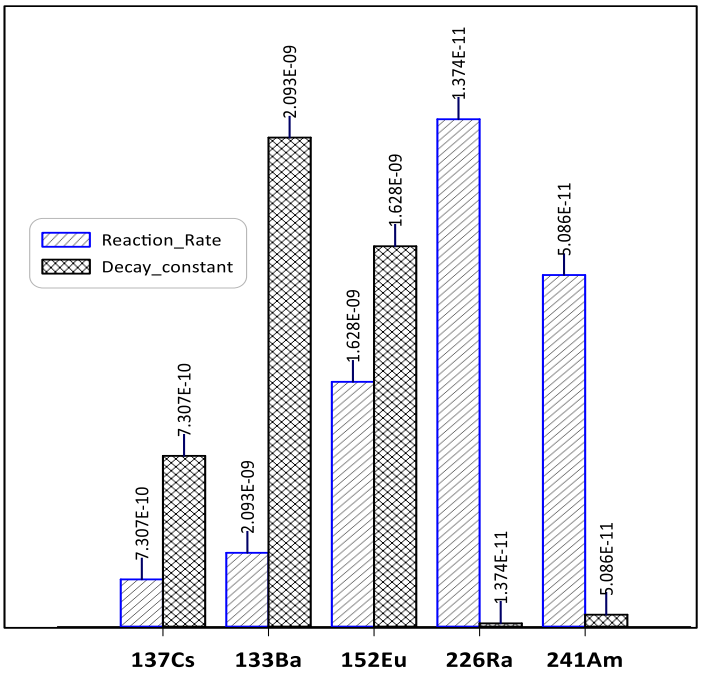


Figure 11: Histogram representation of reaction rate [s^{-1}] and decay constant [s^{-1}] of ^{137}Cs, ^{133}Ba, ^{152}Eu, ^{226}Ra, and ^{241}Am radionuclides.

As shown in Table (3), all neutron-gamma reactions are possible in the thermal region of neutron energy (0.0253 eV equivalent to neutron speed 2200m/s). The capture cross-sections are relatively high in the thermal region but less than the giant resonance band of nuclides that lay in the epithermal region of the neutron spectrum (0.5 eV – 50 keV). As a result of long term exposure to neutrons, the radionuclides are transmuted to short-lived radioactive nuclides that can naturally decay to stable nuclides, as shown in the following sets of equations:

$$
\begin{aligned}
&{}^{137}_{55}\boldsymbol{Cs}(30.08\text{y}) + \boldsymbol{n} \rightarrow {}^{138}_{55}\boldsymbol{Cs}(33.4\text{m}) + \gamma \rightarrow {}^{138}_{56}\boldsymbol{Ba}(\text{Stable}) + \beta^{-} \\
&{}^{133}_{56}\boldsymbol{Ba}(10.55\text{y}) + \boldsymbol{n} \rightarrow {}^{134}_{56}\boldsymbol{Ba}(\text{Stable}) + \gamma \\
&{}^{90}_{38}\boldsymbol{Sr}(28.79\text{y}) + \boldsymbol{n} \rightarrow {}^{91}_{38}\boldsymbol{Sr}(9.6\text{h}) + \gamma \rightarrow {}^{91}_{39}\boldsymbol{Y}(58.5\text{d}) + \beta^{-} \rightarrow {}^{91}_{40}\boldsymbol{Zr}(\text{Stable}) + \beta^{-} \\
&{}^{152}_{63}\boldsymbol{Eu}(13.5\text{y}) + \boldsymbol{n} \rightarrow {}^{153}_{38}\boldsymbol{Eu}(\text{Stable}) + \gamma \\
&{}^{226}_{88}\boldsymbol{Ra}(1.6\text{ky}) + \boldsymbol{n} \rightarrow {}^{227}_{88}\boldsymbol{Ra}(42.2\text{m}) + \gamma \rightarrow {}^{227}_{89}\boldsymbol{Ac}(21.7\text{y}) + \beta^{-} \rightarrow\rightarrow {}^{207}_{82}\boldsymbol{Pb}(\text{Stable}) \\
&{}^{241}_{95}\boldsymbol{Am}(432.6\text{y}) + \boldsymbol{n} \rightarrow {}^{242}_{95}\boldsymbol{Am}(16\text{h}) + \gamma \rightarrow\rightarrow {}^{234}_{92}\boldsymbol{U} \rightarrow\rightarrow {}^{206}_{82}\boldsymbol{Pb}(\text{Stable})
\end{aligned}
\tag{14}
$$

From Equations (14), caesium captures one neutron to be transmitted to ^{138}Cs which has a short half-life of about 33 minutes. ^{138}Cs decays shortly to a stable nuclide ^{138}Ba by emitting beta particle. This type of neutron capture can be performed in all energies of neutrons but of a narrow cross-section not exceeding 350 mbarn at resonance band. Other types of reactions are conducted with fast neutrons because of the high energy needed, except (n, alpha) reaction that can be conducted in the thermal region but rarely happens.

Barium – 133 is the second radionuclide that captures one neutron to be directly converted to a stable isotope ^{134}Ba with a wider cross-section than caesium of about 110 barns at resonance

region (epithermal neutron energy). Reactions (n,p) and (n,alpha) are probable in low energy but also quite rare.

Strontium – 90 radionuclide is widely used as a source of pure beta emitter; however, it cannot be measured by gamma spectroscopy. By using neutron activitation, it can be transmitted to ^{91}Sr isotope that rapidly decays to another radionuclide, ^{91}Y, which is also a pure beta-minus emitter that requires a few months of cooling time to decay to a stable nuclide, ^{91}Zn. Unfortunately, the cross-section of neutron capture for strontium – 90 is very small
.

Europium – 152 is a multi-gamma emitter radionuclide that can cause radiation injuries when exposed to it. ^{152}Eu has a very high neutron capture ability with a cross-section of 12800 barns at the thermal band of neutron energy. It is directly transmuted to a stable isotope of ^{153}Eu.

Radium – 226 is a non-fissionable heavy long-lived radionuclide that naturally occurs in the ground shell; it is part of the ^{238}U decay series, its half-life is 1600 years. It emits alpha particles to generate radon – 222 nuclide (a heavy inert gas) with a half-life of 3.8 days which also decays by emitting alpha particles to continue the progeny carrying mass number of the order (4n+2) and ends up with ^{206}Pb stable nuclide.

Radium was widely used as a radio-luminance substance in military applications and industry. ^{226}Ra has a wide cross-section of capturing thermal neutrons to generate ^{227}Ra of about (42.2 min) half-life. ^{227}Ra is a part of the actinium series (of the order 4n+3) that ends up with ^{207}Pb stable nuclide. ^{227}Ra decay to ^{227}Ac (of the half-life of 21.77 years) by beta particle emission, which is still a long-lived radionuclide. With the continuous activation by thermal neutrons, ^{227}Ac is transferred to ^{228}Ac (6.15 hours half-life) that belongs to the thorium – 232 series (of order 4n), where ^{228}Ac rapidly decays to ^{208}Pb stable nuclide.

Finally, ^{241}Am radionuclide is a heavy fissionable nuclide that belongs to the actinoids group in the periodic table; americium is an artificial nuclide produced by neutron activation of uranium in a nuclear reactor. Fortunately, ^{241}Am has a wide cross-section of capturing neutrons of about $7.9x10^2$ barn to generate ^{242}Am with a half-life of (16 hours); also, it has a fission cross-section of 3.2 barns to generate lighter products.

7. Conclusions:

From the results of neutron irradiation of the six radionuclides, the following can be concluded:

1- The use of neutron incineration is an easy way in the nuclear transmutation process because of the low cost of generating neutron flux either by radioactive sources that emit neutrons or by using T(d, n) and D(d, n) accelerators. Neutron incineration is more effective than photon incineration because of the high cross-section of neutron activation and the low threshold energy of reaction, especially for the (n, γ) reaction, so that low flux of neutron is equivalent to high flux of photons.

2- The results show a high rate of transmutations for the heavy nuclei such as ^{226}Ra and ^{241}Am through the measurement of the reaction rate per nucleus; while results show a low rate of transmutation for the light nuclei such as ^{133}Ba and ^{137}Cs. The ratio of reaction rate to decay constant show highest rate for ^{226}Ra by (1646.8) times, then for ^{241}Am by (308), ^{152}Eu by (6.7), ^{137}Cs by (2.87), and ^{133}Ba by (1.57). This method is more effective for the longest half-life nuclei and for the neutron-rich nuclei that lie on the right side of the stability line in the nuclei chart.

3- From Equations (14), the nuclei ^{133}Ba and ^{152}Eu are directly transmuted to stable isotope by capturing one neutron and emitting gamma photon; other nuclei need short cooling time to decay to stable nuclei after transmutation to radioactive isotopes but in short half-lives like ^{137}Cs and ^{90}Sr, but for heavy nuclei like ^{226}Ra and ^{241}Am the strategy differs, that is these nuclei are a part of long decay series that often ends in one isotope of Pb. The total half-life of the series is very long, but transmuting an element from one isotope to another isotope of a different mass number means the transition from one decay series to another one that might be shorter of half-lives of the rest elements down to the end of the decay series.
4- This method is not recommended to be used for fissionable heavy nuclei because of the variety of fission products that might be generated, that might be with a longer half-life than the original nucleus.

Acknowledgment:

I would like to extend my gratitude to the *Iraqi Radioactive Sources Regulatory Authority* for the help in using the HPGe unit in measurements, and a special gratitude to the staff of the physics deparetment in the college of education – Ibn Alhaithem to allow me to use the neutron-irradiation unit system in the advance nuclear laboratory.

References:

[1] IAEA-TecDoc-1817, "Selection of Technical Solutions for the Management of Radioactive Waste," Vienna, 2017.
[2] M. G. Sowerby, "Nuclear incineration," *Interdisciplinary Science Reviews*, vol. 20, no. 3, pp. 241–255, 1995.
[3] S. Chwaszczewski, and B. Słowinski, "Transmutation of radioactive waste," *Applied Energy*, vol.75, no.1–2, pp. 87–96, 2003.
[4] M. Ripani, "Neutron sources and transmutation of nuclear waste," *Annals of Nuclear Energy*, vol. 62, pp. 590–595, 2013.
[5] M. Steinberg, G. Wotzak, and B. Manowitz, "Neutron burning of long-lived fission products for waste disposal," BNL-8558. Brookhaven National Laboratory, Upton, New York, 1958.
[6] M. Taube, J. Ligou, and K. H. Bucher, "The transmutation of fission products (Cs-137, Sr-90) in a liquid fuelled fast fission reactor with thermal column," EIR-Bericht Nr. 270, 1975.
[7] G. Fioni, M. Cribier, F. Marie, M. Aubert, T. Bolognese, J. -M. Cavedon, F. Chartier, S. Ayrault, O. Deruelle, F. Doneddu, H. Faust, A. Gaudry, F. Gunsing, Ph. Leconte, F. Lelievre, J. Martino, R. Oliver, A. Pluquet, C. Veyssiere, S. Röttger, M. Spiro, "Incineration of ^{241}Am induced by thermal neutrons", *Nuclear Physics*, vol. A 693, pp. 546-564, 2001.
[8] B. Shleien, L. A. Slaback, and B. K. Birky, "*Handbook of health physics and radiological health", Baltimore: Williams & Wilkins*, 1998.
[9] T. Bjørnstad, "Modern Industry: Neutron Basic Interactions, Sources and Detectors for Materials Testing and Inspection," *Encyclopedia of Nuclear Energy, Elsevier Inc.*, vol. 4, pp. 372-391, 2021.
[10] A. D. Vijaya, and A. Kumar, "The neutron spectrum of Am–Be neutron sources," *Nuclear instruments and methods*, vol. 111, no. 3, pp. 435 – 440, 1973.
[11] J. W. Marsh, D. J. Thomas, and M. Burke, "High resolution measurements of neutron energy spectra from AmBe and AmB neutron sources," *Nuclear Instruments and Methods in Physics Research Section*, vol. A 366, pp. 340 – 348, 1995.
[12] IAEA TecDoc-1344, "Categorization of Radioactive Sources," Vienna, 2004.
[13] J. Cetnar, P. Stanisz, and M. Oettingen, "Linear chain method for numerical modelling of burnup systems," *Energies*, vol. 14, no. 6, pp. 1-19, 2021.
[14] M. Benedict, T.H. Pigford, H.W. Levi, "*Nuclear Chemical Engineering," McGraw-Hill, New York*, 1985.
[15] A.J. Koning, D. Rochman, J. Sublet, N. Dzysiuk, M. Fleming and S. van der Marck, "TENDL: Complete Nuclear Data Library for Innovative Nuclear Science and Technology," *Nuclear Data Sheets*, vol. 155, pp. 1 – 55, 2019.

[16] R. S. Peterson, "Experimental gamma ray spectroscopy and investigations of environmental radioactivity," *published by spectrum techniques*, 1996.
[17] M. H. Jasim, S. S. Dawood, "The Characterization of Radioactive Waste Drums Using Nondestructive Scanning Gamma Ray System," *Iraqi Journal of Science*, vol. 58, no. 3A, pp: 1198 – 1206, 2017.
[18] ORTEC, "GammaVision Maestro-PRO" (A66-BW) Software User's Manual (revision M), 2020.
[19] S. F. Mughabghab, Atlas of Neutron Resonances, National Nuclear Data Center, Brookhaven National Laboratory, 2006.