CALCULATIONS OF (γ-n) REACTION CROSS-SECTION AND GAMMA RAY INCINERATION OF ^{144}Ce, ^{144}Nd, ^{151}Sm AND ^{155}Eu

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Abstract

The evaluation of the (γ,n) cross section for radioactive fission products has been done in the present paper. The current research deals with heavy isotopes resulting from fission reactor, namely, ^{144}Ce, ^{144}Nd, ^{151}Sm and ^{155}Eu. The range of energy lays in the region of energy near the giant dipole resonance, from the threshold energy up to around 30 MeV. The present radioactive isotopes were chosen as part of the radioactive waste from ^{235}U fission. Total cross section results then were found and calculated, and used to calculate the number of incinerated nuclei. By varying the gamma-ray fluxes and repeating the calculations, the present method shows efficiency in reducing the radioactivity of these isotopes. The results given in the present paper showed direct proportion between incinerated nuclei and the strength of the incident gamma-ray flux, which are consistent with earlier work.

حساباتُ مساحةِ المقطعِ العرضّيِ لِتفاعُلِ (كاما – نِيوترونْ) ومعدل الإحراق بواسطة أشعة كاما للنوى ^{144}Ce، ^{144}Nd، ^{151}Sm و ^{155}Eu

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الخلاصة

تمَ حساب المساحةِ الكُليةِ للمقطعِ العرضيْ لتفاعلِ أشعة كاما معَ المخلفاتِ المشعةِ ذات الأوزان الثقيلة الناتجة من التفاعل الانشّطاري في البحث الحالي. إن الدراسةَ الحاليةَ تركزُ على النوى ذات العدد الكتلي العالي، وذلك لما لهذِهِ النوى من أهمية في نتائج المفاعل الانشطاري وتشمل:^{144}Ce, ^{144}Nd, ^{151}Sm وأخيرا ^{155}Eu. إن مدى الطاقات التي حُسِبتْ لها مساحة المقطع العرضي تلك تبدأ منْ طاقةِ العتبة المحددة بـ30 ميغا إلكترون فولت وتنتهي بطاقاتٍ قريبةٍ منْ قمة الرنين العظمى للثنائي المغناطيسي (Giant Dipole Resonance)، ولكافة النوى المدروسة. بعد ذلك تمَ إيجادْ وحساب مساحات المقطع العرضي الكلي، وتم استخدام هذه النتائج لحساب العدد الكلي للنوى المتحولة مما يسهم بتقليل مخاطر مخلفات التفاعل الانشطاري وتحويلها إلى نوى أكثر استقراراً. من تغيير فيض أشعة كاما وإعادة الحسابات، بينتْ نتائجُ البحث الحالي كفاءةَ هذه الطريقةِ وفاعليتها. النتائج المعطاة في البحث الحالي أظهرتْ بأن هنالك تناسباً خطيا بينَ شدةِ فيضِ أشعةِ كاما المُستخدم ومعدلِ الإحراق لمجموعةِ النوى المشِعة المدروسة، وهي نتيجة تتفق تماماً مع البحوث السابقة.

Introduction

The environmental safety has become a matter of priority in all of the developed countries in the present days. This interest specially rises with all the development in the various power production methods. Utilization and management of radioactive wastes from nuclear power facilities have received an important priority among the environmental protection studies. Beside that, nations that posses nuclear weapons have become much concerned with disposal or peaceful uses of surplus weapon made of plutonium or of highly-enriched uranium. Many important attempts, that are being developed by using many approaches, are currently under study in Europe, North America, Japan, and Russia [1, 2].

This interest will be an essential step for the further development of high power molten targets in the 20-50 MW range required for Accelerator Driven Systems (ADS) such as high intensity spallation neutron sources, hybrid reactors, neutrino factories, etc. It is clear for this type of power production, that new power generating systems require an adequate disposal mechanism. Techniques for reduction the harmfulness of radioactive wastes therefore have to be developed. The outstanding performances obtained for new fission chambers with a very intense neutron flux [3] made it possible to be employed in order to measure the incineration rate of minor actinides using high neutron fluxes. Some new methods such as the so called "*Developed Innovative Microscopic Fission Chambers*", (μFCs) flux have confirmed a new method to determine the transmutation rate of minor actinides, as shown in Figure.(1) where the ability of ^{235}U fission activity is reduced by a linear relation with irradiation time.

Recently, many studies used various bombarding fluxes to reduce the radioactivity of fission wastes. The used flux may be γ -ray, neutrons or protons and making use of various cross-section studies. For example Titarenko et al.[4] have measured 114 nuclide-production cross sections for isotopes-enriched ^{208}Pb target that was bombarded with intensively energetic 1.0 GeV proton beam.

Two possible treatments are there: possible transmutation of nuclear wastes, and the "*Spallation Neutron Source*" (SNS). The interest shown in both of these techniques encourage to anticipate the accumulation and analysis of more nuclear data. This is possible for both ADS and SNS applications and have the same growth in academic interest and practical commitments in the foreseen future. Therefore experimental data on the cross sections of proton-induced reaction products as applied to the ADS and SNS main target and structure materials are of great interest and importance and should be emphasized that the charge distributions in the isobaric decay chains are important as well. The information thus obtained would make it possible, first, to raise the information content of the comparisons between experimental and simulated data and, second, to reduce the uncertainties in experimental determination of the cumulative and measured cross sections. The possibility of the γ -ray interaction using (γ,n) photonuclear reaction has been examined for the incineration of the long–lived fission product isotope [5-9]. The nuclear incineration method was used to transmute these long-lived nuclei in the high-level radioactive wastes to shorter half-life nuclei [5, 6]. The nuclear reactor method, which is the most promising one so far, has been well studied [3, 4, 7] and its feasibility has been theoretically proven for the trans-uranium actinides. On the other hand, the spallation method using a high energy proton beam has also been proposed, but need more development of proton accelerators [10].

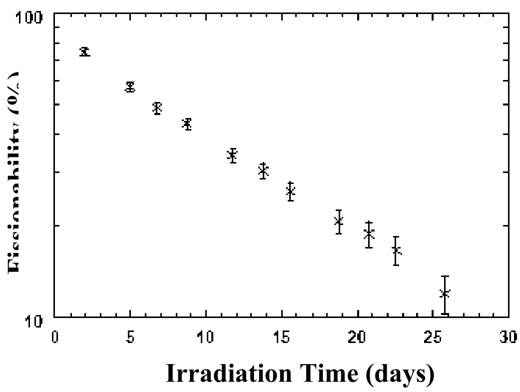

Figure 1: Experimentally determined incineration curve using the μFCs (with ^{235}U as adeposit) irradiated at the lowest position of V4 experimental tube at ILL Grenoble [3].

In the present paper, waste transmutation method using γ -rays photonuclear reaction is made through numeric calculations for the incineration of the high level radioactive wastes. The used γ -rays are of high energy (10-30 MeV), which can be effectively produced by an electron linear accelerator. This energetic γ flux should interact with most nuclei through

the giant resonance of the photonuclear cross section.
In section II a brief outline of the theory is given. Section III lists the results of the present research and a full discussion is given there for the selected isotopes, while in section IV a conclusion of this paper is given.

Theory and Calculation Methods

The total cross-section is then given as [5]:

$$\sigma_{tot} = \sum_{i}^{M} \sigma_i \qquad (1)$$

Where M is the maximum number of events. It is of practical importance to consider only those important events that contribute to σ_{tot} appreciably. This quantity is useful in determining the total probability of γ -ray interaction with the specific target nucleus. The interaction cross section will then be [5]:

$$\sigma_{in} = \int_{E_{th}}^{E_{max}} \sigma(E)\, dE \qquad (2)$$

Finally, to calculate the number of nuclei, N, that will go through transmutation by γ -incineration, the shape of Lorentz line is considered as in Refs.[5,6] and is obtained from the relation:

$$N(t+\Delta t) = 1 + (-1)^j \left(\frac{(\Delta t)^j}{j!} \right) \left(\lambda_j + \sigma_j \phi \right)^j \qquad (3)$$

and λ_i is the j^{th} transition rate, σ_j is the cross section of the j^{th} reaction, and $Ø$ is the γ -ray flux (particles per unit area per second). In order to calculate the γ -ray incineration, the values of σ_{-1} were adopted in the present research. The activity and percentage yield of each of the selected radioactive isotopes are given in Table (1) below. This table shows the activity and yield for three types of nuclear fuel, and the quantities there will be useful during the discussion of the results.
To find the total cross section of γ -rays with nuclei one can use the Lorentz curve, [11], given in general form by [5]:

$$\sigma(E) = \sum_{i=1}^{2} \frac{\sigma_{mi}}{1 + \left[\frac{E^2 - E_{mi}^2}{E^2 \Gamma^2} \right]^2} \qquad (4)$$

Where σ_m, E_m and Γ are the parameters of the Lorentz curve for peak cross section, resonance energy and the full-width at half maximum, respectively; and i corresponds to the lower and upper energy lines. From Table (2) one can see that it is safe to start the numerical calculations for the cross section values from energy~10MeV. The calculated values of σ_{mi} and σ_{-1} for all the radioactive isotopes that are presently used in this paper are shown in Figure (2) and Figure (3), respectively, in the next paragraph.

Table 1: Radioactive products of fission reaction for 1 kg of ^{233}U, ^{235}U and ^{239}Pu, and the neutron yield for each of the presently used isotopes, [taken from ref. 12].

Isotope	Half life $\tau_{½}$	^{233}U		^{235}U		^{239}Pu		Total Activity (Ci)
		Yield %	Activity (Ci)	Yield %	Activity (Ci)	Yield %	Activity (Ci)	
^{144}Ce	280 d	4.50	1.45×10^{5}	5.62	1.82×10^{5}	3.93	1.27×10^{5}	4.54×10^{5}
^{144}Nd	5×10^{15} y	4.61	2.28×10^{-11}	3.98	1.9×10^{-11}	3.13	1.5×10^{-11}	5.8×10^{-11}
^{151}Sm	80 y	0.33	98.96	0.443	129.97	0.80	236.32	465.26
^{155}Eu	1.81 y	–	–	0.033	42.051	–	–	42.051

Table 2: Threshold energy corresponding to some possible (γ-n) reactions for the radioactive isotopes used in the present research [13]. Maximum γ -ray energy for all reactions is 30 MeV.

Fission Product	(γ,n) Reaction	E_{th} (MeV)
$^{144}_{58}Ce_{86}$	^{144}Ce(γ,n)^{143}Ce	4.78
$^{144}_{60}Nd_{84}$	^{144}Nd(γ,n)^{143}Nd	7.82
$^{151}_{62}Sm_{89}$	^{151}Sm(γ,n)^{150}Sm	5.60
$^{155}_{63}Eu_{92}$	^{155}Eu(γ,n)^{154}Eu	8.20

Results and Discussion

In order to calculate the nuclear cross section values for all possible (γ -n) reactions, eq.(3) and eq.(4) were used. These equations were employed to find the different cross sections for (γ -n) reactions taking place with the radioactive isotopes chosen for the present research. In Figure (2) and Figure (3), the calculated cross sections, σ_{int} and σ_{-1} for γ -interactions with each of the present radioactive nuclei are shown. Similar to previous results [5, 6], the interaction and first moment cross sections of this calculation both increased exponentially from the threshold energy to about ~20MeV, after that the dependence reached a saturation and became almost a constant. This dependence is higher for the first moment cross section at the same energy, compared with the integrated cross section.

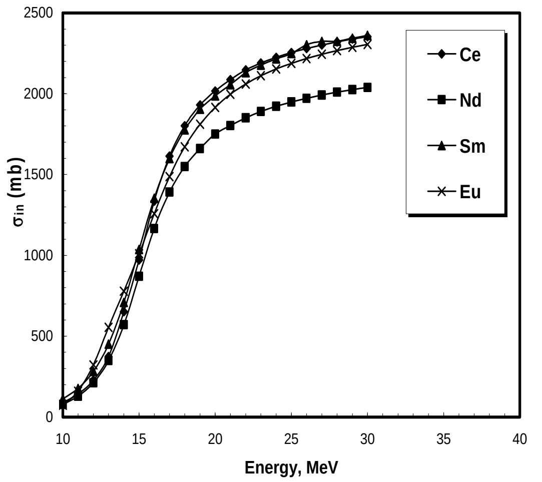

Figure 2: The calculated values of the interaction cross section for γ - ray interaction with the radioactive isotopes, σ_{in} as function of γ - ray energy.

The results of incineration of the selected group of radioactive nuclei are shown in the following figures, Figure (4) to Figure (8). Due to the high difference in the results of these isotopes, the figures were made between logarithm of the ratio of incinerated nuclei, $N_{inc.}$ to the original number of nuclei, N_{orig} against the energy *E* of γ -ray. By this one insures that the maximum value of all curves will be a unity (corresponding to 100% incineration); thus making comparison more obvious. In all the following figures, the same calculations were repeated by changing only the flux, Ø, of the incident γ -ray, as indicated in each figure, starting from $Ø=10^{16}$ γ /cm^2.s - for Figure(4)- to maximum flux of value $Ø=10^{20}$ γ /cm^2.s,Figure(8).

From Figure (4) one can see clearly that the incineration results of ^{144}Ce isotope were the highest among the others. Although the expected next curve would be ^{144}Nd, because of the close atomic numbers and similar mass numbers, however the results of ^{144}Nd are the lowest. This result, although seems unexpected, but actually it is consistent with the results of the measured cross sections, Figure (1) and Figure (2). Incineration is a process that highly depends on the interaction cross section, as already mentioned in the introduction, therefore, the higher γ-nucleus cross section means higher incineration rate, thus producing higher ratio $N_{inc}./N_{orig}$. This dependence is limited, however, to the strength of the γ -flux in this case, as it will be seen when noting the incineration at γ - ray fluxes higher than 10^{19} γ /cm^2.s.

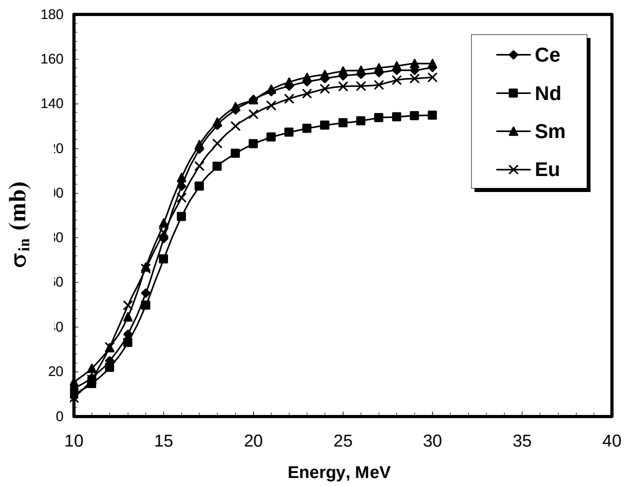

Figure 3: The calculated values of the first-momentum cross section for γ - ray interaction with the radioactive isotopes, σ_{-1} as function of γ - ray energy.

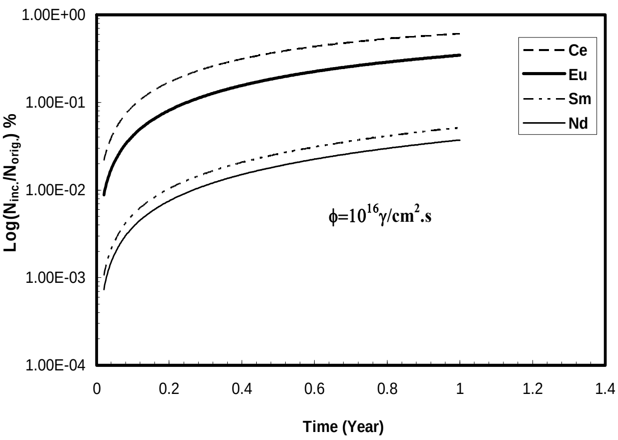

Figure 4: The relation between logarithm the ratio $N_{inc}./N_{orig}.$, and incineration time, for γ-ray flux 10^{16} γ /cm^2.s

Increasing the flux from 10^{16} γ /cm^2.s to 10^{17} γ /cm^2.s, the results shown in Figure (5) indicate that the value of $N_{inc}./N_{orig.}$ become more closer to unity in one hand and closer to each other in the other hand. Comparing the results of Figure (5) with those of Figure (4) clearly shows how much improvement obtained for all the present radioactive isotopes. This clarifies that, when increasing the flux of γ - rays, the differences in incineration results becomes less. This is seen more clearly in Figure (6), where the flux was increased to 10^{18} γ /cm^2.s.

In the following figures, Figure(6) to Figure(8), the curves are more difficult to be distinguished from each others where the differences become much less than in the first figure with γ -ray flux 10^{16} γ /cm^2.s. From these figures one can notice how less the differences in the reaction cross sections contribute to incinerating these radioactive nuclei. Such results indicate the powerfulness of incineration and transmutation of radioactive isotopes using γ -rays.

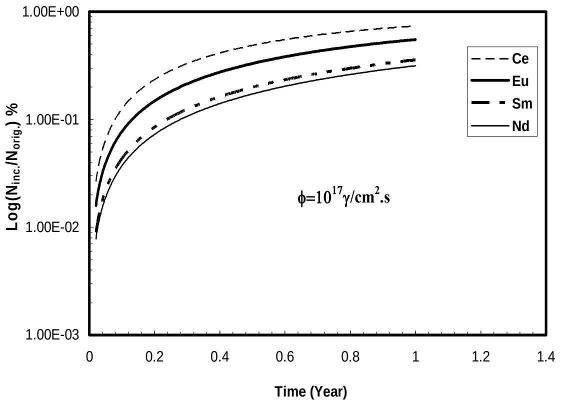

Figure 5: The relation between logarithm the ratio $N_{inc.}/N_{orig.}$, and incineration time, for γ-ray flux 10^{17} γ /cm2.s

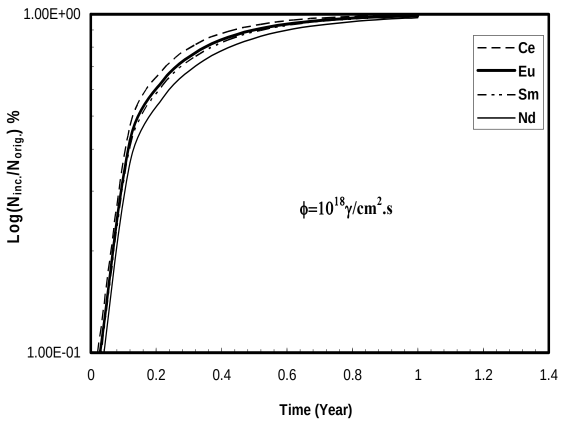

Figure 6: The relation between logarithm the ratio $N_{inc.}/N_{orig.}$, and incineration time, for γ-ray flux 10^{18} γ/cm^2.s

For γ -ray fluxes 10^{19} γ /cm^2.s and 10^{20} γ /cm^2.s, the results are shown in Figure (7) and Figure (8), respectively. One can see that at such high γ -ray fluxes the incineration rate was very efficient in such a way that in few days irradiation the incineration was almost 100%. From Figure(7) with the γ -ray flux 10^{19} γ /cm^2.s irradiation for 25 days resulting incineration rate better than 95% for all the radioactive isotopes. When increasing the γ -ray flux to 10^{20} γ /cm^2.s, the results were even more improved as shown in Figure.(8) where irradiation of the samples for 5 days only gave incineration rate about 99% for all the nuclei.

The most important result is that for ^{144}Ce, which has a relatively high percentage yield from the ^{235}U-see Table (1)-where it was successfully incinerated almost 100% at such high γ -ray flux. This indicates the importance of γ -ray incineration method to reduce the harmfulness of radioactive isotopes resulting from nuclear fission reaction. Due to its importance, this method is preferred to be applied to more radioactive isotopes to include all the nuclear reactor waste.

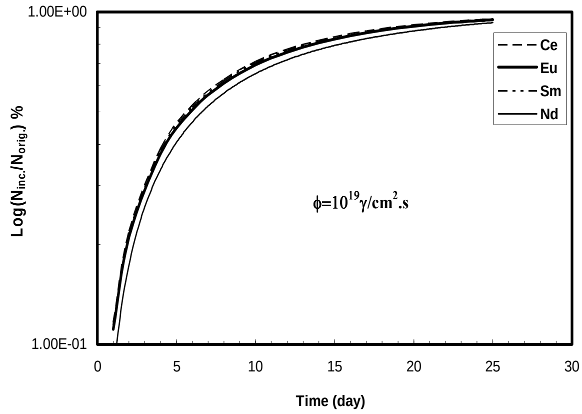

Figure 7: The relation between logarithm the ratio $N_{inc.}/N_{orig.}$, and incineration time, for γ-ray flux 10^{19} γ /cm^2.s

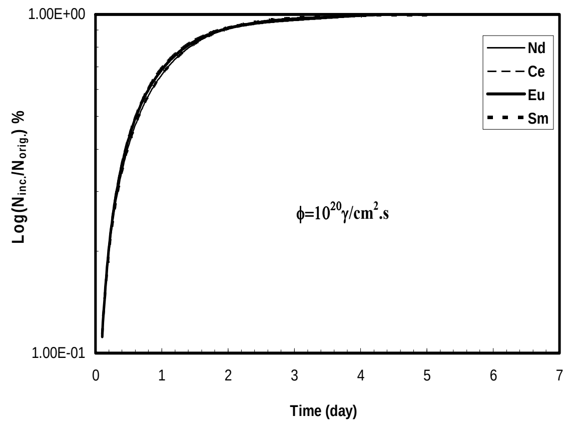

Figure 8: The relation between logarithm the ratio Ninc./Norig., and incineration time, for γ-ray flux 10^{20} γ/cm^2.s

From all the above figures -Figure (4) to Figure (8) - an obvious relation is seen that there is a proportional relation between γ-ray flux and incineration rate for all the selected nuclei. As the used flux increased, incineration increases.

This clarify the importance of using the highest possible γ-ray fluxes available at hand to treat the radioactive reactor waste in as short time as possible. However, the results of relatively low γ -ray fluxes-Figure(4)-also shows that some radioactive isotopes such as ^{144}Ce and ^{155}Eu, still properly respond to the present method, but at the price of the long irradiation time, to achieve the desired results.

Conclusions

The reduction of the radioactivity for four radioactive isotopes that result from reactor waste was investigated in the present research by using γ -ray incineration method. Different γ -ray fluxes were used to examine the incineration rate, from 10^{16} to 10^{20} γ /cm^2.s. As the flux of γ -ray increased, incineration rate improved with good efficiency. At the maximum γ -ray flux used, almost all the selected isotopes were incinerated reaching a ratio better than 99% after few days of irradiation, indicating the useful and powerfulness of this method in treating radioactive waste from nuclear fission reactor.

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