

CALCULATIONS OF (γ ,n) REACTION CROSS SECTION AND GAMMA RAY INCINERATION FOR MEDIUM MASS RADIOACTIVE ISOTOPES

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Abstract

The total γ -n reaction cross section for radioactive fission products have been evaluated in the region of energy near the giant dipole resonance, from the threshold energy up to around 30 MeV. The selected fission products lay in the range of medium mass number, namely: ^{87}Rb , ^{90}Sr , ^{93}Zr , ^{106}Ru , ^{123}Sn , ^{127}Te , ^{129}I and ^{137}Cs . These radioactive isotopes were chosen as part of the radioactive waste from ^{235}U fission. Total cross section results were used to calculate the number of incinerated nuclei, and this method shows efficiency in reducing the radioactivity of these isotopes. The obtained results show that the incineration rate of radioactive nuclei increases with the flux of the incident γ -rays, whereas the required irradiation time was found to be shortened and inversely proportional to the incident γ -ray flux. These results are consistent with those given by earlier researches.

حسابات مساحة المقطع العرضي ومعدل الإحراق لتفاعل (كاما-نيوترون) لنظائر مشعة ذات عدد كتلي متوسط

الخلاصة

في هذا البحث تم حساب المساحة الكلية للمقطع العرضي لتفاعل (كاما-نيوترون) مع المخلفات المشعة الناتجة من التفاعل الانشطاري، وكان مدى الطاقات التي حُبيبت لها مساحة المقطع العرضي تلك والتي تمتد من طاقة العتبة وحتى 30 ميغا إلكترون فولت، أي في الطاقات القريبة من قمة الرنين العظمى (Giant Dipole Resonance) للنوى المدروسة. إن الدراسة الحالية ركزت على النوى ذات العدد الكتلي المتوسط، وتحديدًا هي: ^{87}Rb , ^{90}Sr , ^{93}Zr , ^{106}Ru , ^{123}Sn , ^{127}Te , ^{129}I و أخيراً ^{137}Cs . لقد تم اختيار هذه المجموعة من العناصر المشعة وذلك لأنها تكون جزءاً مهماً من المخلفات المشعة الناتجة من التفاعل الانشطاري للوقود النووي. بعد ذلك تم استخدام مساحات المقطع العرضي المحسوبة للحصول على معدل إحراق وتحويل مخلفات التفاعل الانشطاري إلى نوى أكثر استقراراً وأقل ضرراً وهي طريقة عملية لتقليل النشاط الإشعاعي للمخلفات المشعة، ولقد تبين من نتائج البحث الحالي كفاءة هذه الطريقة وفعاليتها. النتائج الحالية أظهرت بأن معدل الإحراق لمجموعة النوى المشعة المدروسة يتزايد مع شدة فيض أشعة كاما المستخدم، كما لوحظ أن زمن التشعيع المطلوب يتناقص بصورة مستمرة مع زيادة شدة ذلك الفيض.

Introduction

Energy production in nuclear power plant on the basis of fission process lead inevitably to fission products and generation new actinide

isotopes. One of the most pronounced problems with the use of nuclear power today is how safety takes care of the burned nuclear fuel, commonly called reactor waste, which is both

highly radiotoxic and which has a very long radiological half-life [1].

Different means to ensure that the waste will not interplay with the biological life on earth have been proposed through the years. The fission process, among all other various nuclear activities, is responsible for most of the radioactive waste. For example, fission products (such as Iodine, Technetium, Neodymium, Zirconium, Molybdenum, Cerium, Cesium, ... etc) may constitute as much as 2.9% of the weight of the spent fuel [2]. At present time, the main option in most countries having nuclear power production plants in operation, is deep geological disposal with or without reprocessing [see, e.g., ref.3 and references therein]. An alternative option which is now under consideration, is first to incinerate and transmute the high level radioactive wastes, in order to reduce the amount of the radio-toxicity and the half-life before disposing the remaining waste in a geological repository [2].

The feasibility of the γ -ray interaction method using photonuclear reaction (γ, n) has been examined for the incineration of the long-lived fission product isotopes. The nuclear incineration method has recently been proposed to transmute these long-lived nuclei in the high-level radioactive wastes to shorter half-life nuclei or to stable ones. The nuclear reactor method, which is the most promising method, has been well studied 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 it needs more development of proton accelerators.

In the present paper waste transmutation method using γ -rays photonuclear reaction is proposed 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 reaction cross section [1]. The importance of the present method is examined here by using the γ -ray flux as an adjustable parameter. This is made because the conversion efficiency of the electron current to the γ -ray flux is a facility-dependent during both the irradiation and cooling periods. Also, only the (γ -n) reaction is treated among several photonuclear reaction because the other reaction, such

as (γ -n α), (γ -2n), (γ -2np) etc., all have less important contribution at giant resonance energies [1]. If one adds the lack of knowledge about the required cross sections, then it will be worthy to focus on (γ -n) reactions only. (γ -p) reaction is inhibited by the Coulomb barrier, for medium and heavy nuclei.

The efficiency of γ -incineration method have been widely investigated in recent years. The investigation was to examine incineration of the long-lived radioactive isotopes because of the practical importance hoped from this technique. Specially for the case of (γ -n) reaction, investigated radioactive nuclei include the following isotopes: ^{94}Zr , ^{96}Mo , ^{124}Sn , ^{126}Te , ^{142}Ce , ^{144}Nd [4], ^{90}Sr and ^{137}Cs [5], ^{127}I [6,7], ^{138}Ba [2], $^{92,94,95,96,97}\text{Zr}$ isotopes [8,9], $^{116-118,120,124}\text{Sn}$ isotopes [10-12], $^{146,148,150,152,154}\text{Sm}$ isotopes [12,13], and many other types of reactor waste radioactive isotopes. The technique of (γ -n) reaction were also applied to many other heavy and light [1,14,15,16] isotopes.

Theory of Cross-Section Calculations of (γ -n) Reaction

For each event the specified cross section can be labeled by σ_i and the total cross-section will

be given as $\sigma_{tot} = \sum_i \sigma_i$, and this quantity is

useful in determining the total probability of γ -ray interaction with the specific target nucleus (or nuclei).

The different photon-neutron reaction cross sections that are important to the present research are listed below [1]:

i) The total photon-neutron cross section, which may be described by the following approximated relation:

$$\sigma_{\gamma, tot} = \sigma(\gamma, n) + \sigma(\gamma, pn) \quad (1)$$

ii) The integrated photon-neutron reaction cross sections, which can be found from Thomas-Reiche-Kuhn (TRK) summation relation given as:

$$\begin{aligned} \sigma &= \int_0^{\infty} \sigma(E) dE = \frac{2\pi^2 e^2 \hbar^2}{mc} \frac{NZ}{A} \\ &\cong \frac{60}{A} \frac{NZ}{A} \quad (\text{MeV}\cdot\text{mb}) \quad (2) \end{aligned}$$

where m is the mass of the nucleon, c is the speed of light, e is the unit charge and Z is the

atomic number, and A is the mass number of the target nucleus. The interaction cross section will then be given for energy range from a threshold value of energy, E_{th} , to maximum value E_{max} :

$$\sigma_{in} = \int_{E_{th}}^{E_{max}} \sigma(E) dE \quad (3)$$

iii) The first moment of the integrated cross section, σ_{-1} , also known as the "Bremsstrahlung-weighted cross section", given as [18]:

$$\sigma_{-1} = \int_0^{\infty} \frac{\sigma(E)}{E} dE = \frac{4\pi^2 e^2}{3\hbar c} \frac{NZ}{A-1} \langle r^2 \rangle \quad (4)$$

where $\langle r^2 \rangle$ is the mean-square radius of the nuclear charge distribution.

The total cross section of γ -rays interactions with nuclei can be found from the fundamental reaction cross section which is described by the Lorentz curve, [18]:

$$\sigma(E) = \frac{\sigma_m}{1 + \left[\frac{E^2 - E_m^2}{E^2 \Gamma^2} \right]^2} \quad (5)$$

where σ_m , E_m and Γ are the parameters of the Lorentz curve representing peak cross section, resonance energy and the full-width at half maximum, respectively. The giant dipole resonance of spherical nuclei consists of one such Lorentz line. This would represent photon absorption which induces neutron and proton oscillations inside the nuclear matter. The superposition of two Lorentz lines would correspond to nucleon oscillations along the longer axis in deformed nuclei. In such cases, one should sum eq.(5) to have:

$$\sigma(E) = \sum_{i=1}^2 \frac{\sigma_{mi}}{1 + \left[\frac{E^2 - E_{mi}^2}{E^2 \Gamma_i^2} \right]^2} \quad (6)$$

where the index i represents the higher ($i=2$) and lower ($i=1$) energy for each of the major axes of the deformed nucleus. Eq.(6) was based on the assumption that Γ is not being a function of energy thus the two widths would not interfere with each others, so one should put this

consideration in mind during numerical calculations. Finally the total area under Lorentz curves will be given as [1]:

$$\int_0^{\infty} \sigma(E) dE = \frac{\pi}{2} \sigma_m \Gamma \quad \left(\text{or} = \sum_{i=1}^2 \frac{\pi}{2} \sigma_{mi} \Gamma_i \right) \quad (7)$$

where one chooses either forms (with or without summation) depending on the nature of the problem under study.

γ -ray Incineration

The (γ -n) reaction is considered among several photo-nuclear reactions because it has the most important contribution among all the other possible reactions such as (γ -p), (γ -np), (γ -n α), (γ -2np)...etc. The number of nuclei, N , that will go through transmutation by γ -incineration can be obtained as follows:

Let the incineration starts at time t and ends at time $t+\Delta t$, then number of nuclei, N , produced within the time interval of $(t+\Delta t)$ is [1,5]:

$$N(t + \Delta t) = N(t) + \sum_{i=1} \left(\frac{(\Delta t)^i}{i!} \right) A^i N(t) \quad (8)$$

where the function A is given as [1]:

$$A = - \sum_j (\lambda_j + \sigma_j \phi) \quad (9)$$

and λ_j is the j^{th} transition rate, σ_j is the cross section of the j^{th} reaction, and ϕ is the γ -ray flux (particles per unit area per second). On expansion of eq.(8) using Taylor series one gets [1]:

$$N(t + \Delta t) = N(0) + \sum_i \left(\frac{(\Delta t)^i}{i!} \right) A^i N(0) \quad (10)$$

and $N(0)=1$. Eq.(10) thus simply gives:

$$N(t + \Delta t) = 1 + \sum_i \sum_j (-1)^i \left(\frac{(\Delta t)^i}{i!} \right) (\lambda_j + \sigma_j \phi)^i \quad (11)$$

In order to calculate the γ -ray incineration, the values of σ_{-1} were adopted in the present research.

Results and Discussion

In order to calculate the nuclear cross section values for all possible (γ -n) reactions, the Lorentz equations -eq.(5 and 6)- were used. These equations were employed to find reaction cross-sections of those reactions taking place with the radioactive isotopes chosen for the present research.

Mainly, the radioactive isotopes produced from fission reaction were chosen. Equations (5 and 6) were fitted [data from Ref.17] to find the parameters of the Lorentz shape so that one can calculate the reaction cross section as a function of energy, $\sigma(E)$. Cross section values were calculated for the energy range extending from threshold energy, E_{th} , to the maximum energy by steps of 0.1 MeV. Table (1) below shows the radioactive products of fission reaction for 1 kg of ^{233}U , ^{235}U and ^{239}Pu , and the neutron yield for each isotope [17].

After finding $\sigma(E)$, the interaction cross section was calculated from eq.(3). The Bremsstrahlung-weighted cross section-eq.(4)- was also calculated for different values of energies (from E_{th} to E_{max}).

The isotopes were chosen such that their lifetime is greater than 100 days and with yield greater than 0.1%. This choice made it possible to perform the incineration process without being exposed to the harmful ionizing radiation for long time.

The threshold energy used in the present research can be calculated from the relation [19]:

$$E_{th} = -Q \quad (12)$$

where Q is the energy yield value of the reaction. Since there is about ~2% variance in this equation [19], a tabulated values of the threshold energy were used in this research which gives a slightly accurate and better results. The data for the threshold energy were taken from ref.[20] and are listed in Table (2) only for (γ -n) reactions here. The maximum γ -ray energy for all isotopes listed in Table (2) is 30 MeV.

the numerical calculations for the cross section values from energy $\sim 10\text{MeV}$. The calculated values of σ_{in} and σ_{-1} for all the radioactive isotopes that are presently used in this paper are as shown in the figures Figure(1) and Figure(2) below.

The interaction and first moment cross sections of the present calculations, are increasing almost exponentially from the threshold energy to about $\sim 20\text{MeV}$, then the dependence becomes less dependent and looks like linear dependence. The dependence is of higher values for the first moment cross section at the same energy. This is seen from Figure(2), the values of σ_{-1} for ^{127}Te isotope, for example increased about four times in the energy range 15-20 MeV, while the corresponding increment in the value of the σ_{in} of same isotope was less than three times, Figure.1,. The behavior of both these values indicates a relation between the atomic mass A and probably the atomic number Z . Such interesting dependence will not be discussed here and is left for future work. In the present research we restrict ourselves to the efficiency of γ -ray incineration method.

The results of the present calculations for incineration of the selected radioactive isotopes are shown in the figures -Figure(3) to Figure(7)- where the numbers of incinerated nuclei, $N_{inc.}$, to the original number of nuclei, $N_{orig.}$, are shown as functions of time in these figures, for different γ -ray fluxes from 10^{16} to 10^{20} $\gamma/\text{cm}^2.\text{s}$.

These cases differ from each others depending on the type of the irradiated nucleus and its excitation half-life, and each case was chosen for different γ -ray flux. Such variation in the parameters of the present study shows the feasibility of the present method for treating the radioactive wastes. Although such treatment will need further effort for dealing with the reactor waste, the present results clearly show that this effort is worthy.

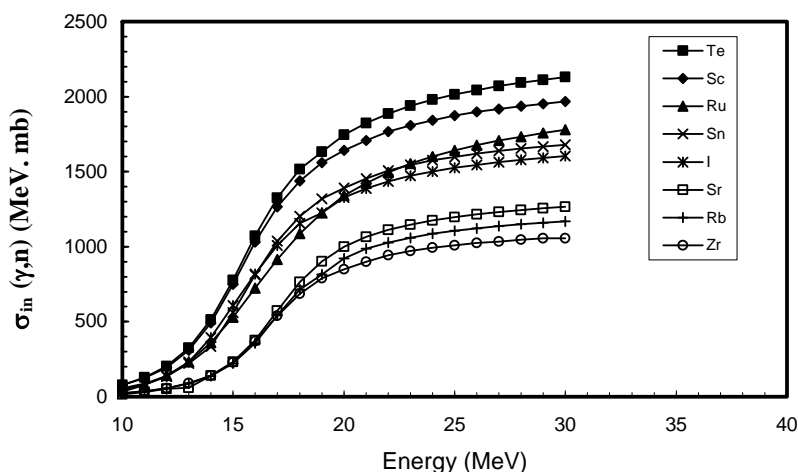
It can be seen from Table(2) that one can start

Table(1). Radioactive products of fission reaction for 1 kg of ^{233}U , ^{235}U and ^{239}Pu . [from ref. 17]

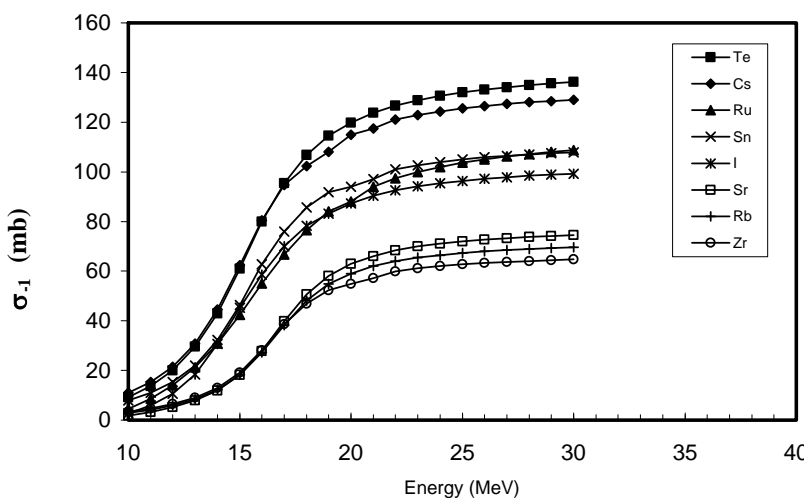
Isotope	Half life $\tau_{1/2}$	^{233}U		^{235}U		^{239}Pu		Total Activity (Ci)
		Yield %	Activity (Ci)	Yield %	Activity (Ci)	Yield %	Activity (Ci)	
^{87}Rb	5×10^{10} y	4.56	3.74×10^{-6}	2.49	2.04×10^{-6}	0.92	7.55×10^{-7}	6.54×10^{-6}
^{90}Sr	28y	6.43	9.36×10^3	5.77	8.39×10^3	2.25	3.27×10^3	21.0×10^3
^{93}Zr	1.1×10^6 y	6.98	0.2430	6.40	0.2230	4.48	0.1563	0.623200
^{106}Ru	1.01 y	0.24	8.0020	0.38	12.670	4.57	152.37	173.0450
^{123}Sn	136 d	-	-	2×10^{-2}	1.01×10^2	-	-	1.01×10^2
^{127}Te	105 d	-	-	0.35	3.42×10^4	-	-	426×10^4
^{129}I	1.7×10^7 y	-	-	0.80	1.30×10^2	-	-	1.30×10^2
^{137}Cs	30 y	6.58	5.71×10^3	6.15	5.34×10^3	6.63	5.75×10^3	16.8×10^3

Table (2). Threshold energy corresponding to some possible (γ -n) reactions for the radioactive isotopes used in the present research [20]. Maximum γ -ray energy for all reactions is 30 MeV.

Fission Product	(γ ,n) Reaction	E_{th} (MeV)
^{87}Rb	$^{87}\text{Rb}(\gamma,n)^{86}\text{Rb}$	9.93
^{90}Sr	$^{90}\text{Sr}(\gamma,n)^{89}\text{Sr}$	7.80
^{93}Zr	$^{93}\text{Zr}(\gamma,n)^{92}\text{Zr}$	6.76
^{106}Ru	$^{106}\text{Ru}(\gamma,n)^{105}\text{Ru}$	8.47
^{123}Sn	$^{123}\text{Sn}(\gamma,n)^{122}\text{Sn}$	5.59
^{127}Te	$^{127}\text{Te}(\gamma,n)^{126}\text{Te}$	6.29
^{129}I	$^{129}\text{I}(\gamma,n)^{128}\text{I}$	8.84
^{137}Cs	$^{137}\text{Cs}(\gamma,n)^{136}\text{Cs}$	8.28



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



Figure(2): The calculated values of the first-moment cross section for γ -ray interaction with the radioactive isotopes, σ_{-1} as function of γ -ray energy.

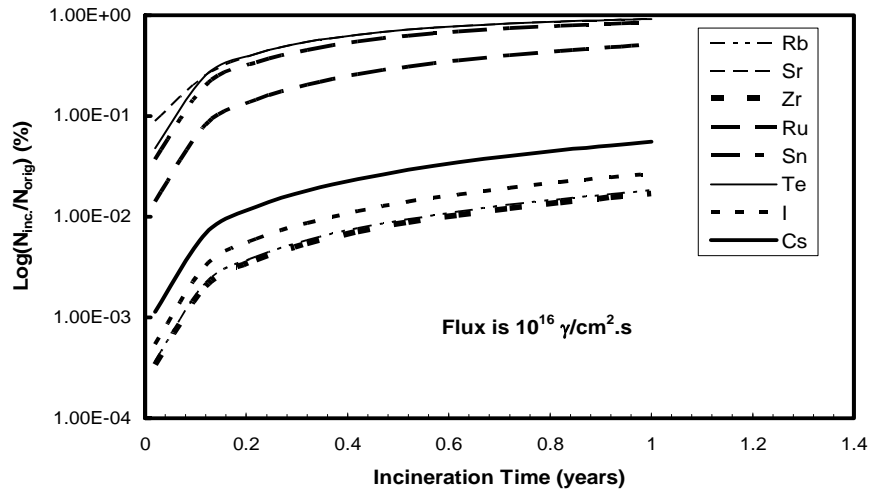


Figure (3): The ratio between logarithm $N_{inc.}/N_{orig.}$, as a function of incineration time for γ -ray flux $10^{16} \gamma/cm^2.s$

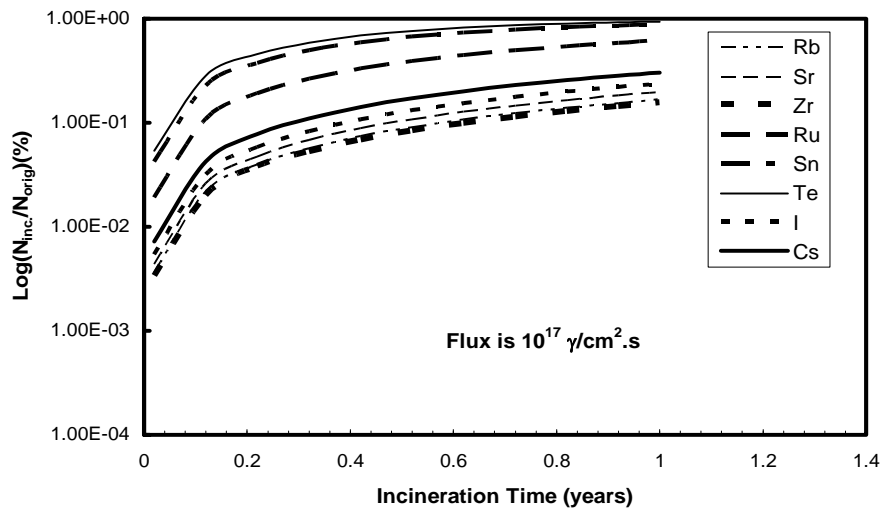


Figure (4): The ratio between logarithm $N_{inc.}/N_{orig.}$, as a function of incineration time for γ -ray flux $10^{17} \gamma/cm^2.s$

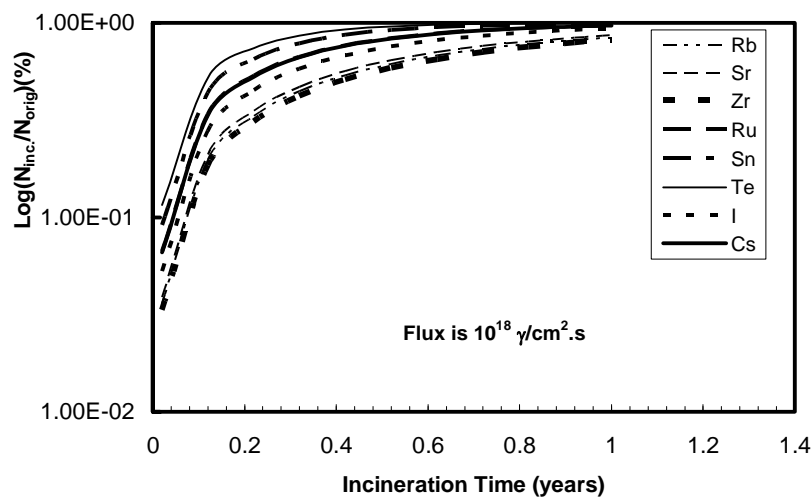


Figure (5): The ratio between logarithm $N_{inc.}/N_{orig.}$, as a function of incineration time for γ -ray flux $10^{18} \gamma/cm^2.s$

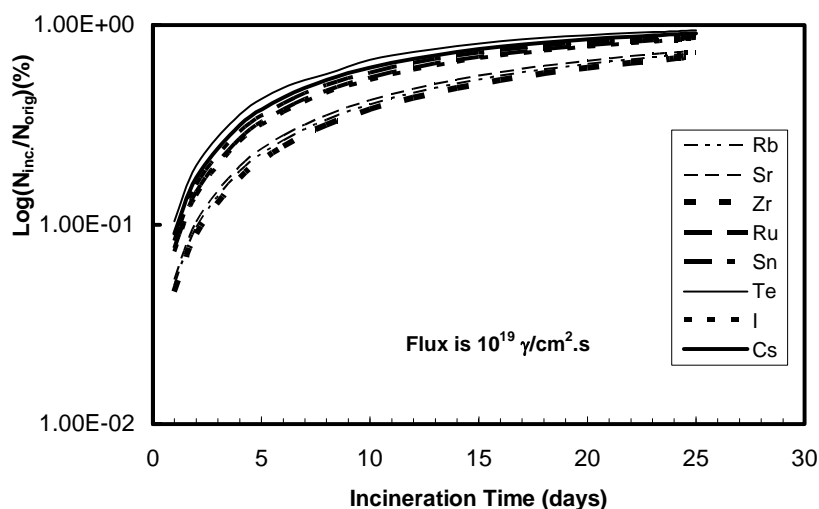


Figure (6): The ratio between logarithm $N_{inc.}/N_{orig.}$, as a function of incineration time for γ -ray flux $10^{19} \gamma/cm^2.s$

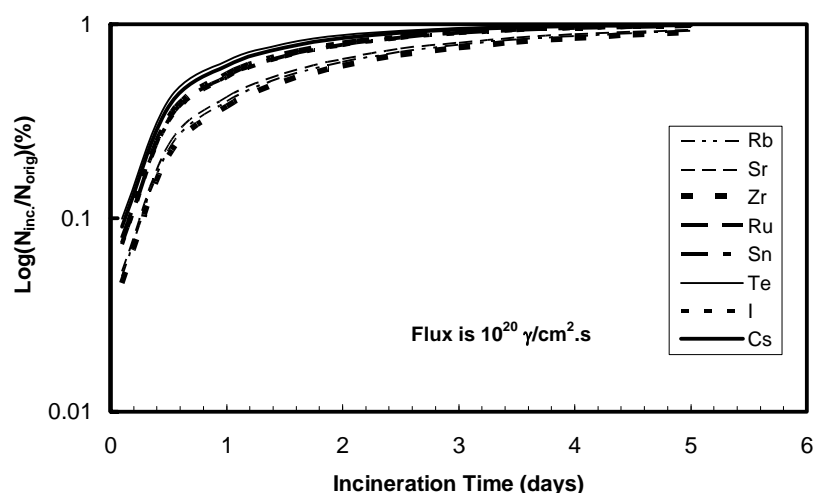


Figure (7): The ratio between logarithm $N_{inc.}/N_{orig.}$, as a function of incineration time for γ -ray flux $10^{20} \gamma/cm^2.s$

It should be mentioned here that for each curve in these figures a renormalization was made such that the number of incinerated nuclei at the beginning of irradiation is taken as unity. From Figure(3) one can see that irradiation of the selected group of radioactive waste nuclei by some fluxes is insufficient to reduce the harmful radioactivity of the samples. For example, at γ -ray flux $10^{16} \gamma/cm^2.s$, after irradiating the ^{87}Rb isotope for one year the incinerated nuclei were making a fraction less~1.83% only. This small ratio of incineration of the selected nuclei is

also shown for ^{90}Sr is 4%, ^{93}Zr is 1.7%, ^{129}I is 2.7% and ^{137}Cs is 5.5%, while for ^{127}Te , same irradiation gives about 90% of incinerated nuclei after the same time, and 50% for ^{106}Ru , and 85% for ^{123}Sn .

This shows the efficiency of incineration and transmutation of radioactive isotopes using energetic γ -ray for the nuclei with high cross-section. One can refer to Figure(1) to see how these three isotopes have higher cross-sections than other isotopes for the same energies. This indicates the feasibility of the present method

for ^{106}Ru , ^{123}Sn and ^{127}Te , even at relatively low-value fluxes.

The incineration of the other isotopes can be improved by increasing the γ -ray fluxes, as shown in Figure(4) to Figure(7) for fluxes 10^{17} to $10^{20} \gamma/\text{cm}^2.\text{s}$, respectively. For the highest flux, $10^{20} \gamma/\text{cm}^2.\text{s}$, all the selected isotopes have incineration efficiency more than 90%, even for short irradiation times (five days only). Some of the isotopes have efficiency more than 99% such as ^{127}Te and ^{137}Cs . This result is very important in the present frame of study because, referring to Table(1), the isotope ^{137}Cs has half-life of 30 years and yield more than ~6%, thus reducing its radioactivity by more than 99% in five days gives a clear example about how powerful and important the γ -ray incineration method is. This behavior was also seen when investigating other radioactive isotopes from the reactor waste [1,15]. Even at lower γ -ray fluxes the results of the present research are in general satisfactory to achieve the desired aim.

Conclusions

The present research gives a study for incineration and transmutation of six radioactive isotopes, of atomic masses ranging from 87 to 137, using γ -ray incineration. The selected isotopes were selected as to be the wastes of the nuclear fission reaction. These isotopes were subjected to different γ -ray fluxes varying from 10^{16} to $10^{20} \gamma/\text{cm}^2.\text{s}$, and the calculations were performed from threshold to maximum energy that is near the giant resonance threshold. The current results showed that incineration of the radioactive isotopes increases linearly proportional to the incident γ -ray flux. Some isotopes showed extremely good response for the present method even at flux of $10^{16} \gamma/\text{cm}^2.\text{s}$, and all of the isotopes showed reduction in their radioactivity more than 90% at the highest γ -ray flux, $10^{20} \gamma/\text{cm}^2.\text{s}$.

References

1. Saeed, M.A., **2002** "Calculation of γ -n Reaction Cross Sections For Radioactive Fission Products," M. Sc. Thesis, Department of Physics, College of Science, University of Baghdad.
2. Broeders, C. H. M., Kiefhaber, E. and Wiese, H. W., Nucl. Eng. **2000**. Des. 202,157-161,.
3. Brown, P. M., **2002** "Photo-Transmutation for Waste Management," Nuclear Solution.
4. Dietrich, S. S. and Berman, B. L., **1988**. Nuclear Data Tables 38,199-217.
5. Matsumoto, T., Nucl. **1988**. Inst. Meth. Phys. Res. A268, 234-239.
6. Rassoll, R. P. and Thompson, M. N., **1989**. Phys. Rev. C39,2201-2210.
7. Brmblett, R. L., Caldwell, J. T., B. Berman, L., Harvey, R. R. and Fultz, S. C., **1966**. Phys. Rev. 148, 1198-1203.
8. Berman, B. L., Caldwell, J. T., Harvey, R. R., Kelly, M. A., Brmblett, R. L., and Fultz, S. C., **1967**. Phys. Rev. 162, 1098-1118.
9. Lepratré, A., Beil, H., Bergere, R., Carlos, P., Reyssiars, A., and Sugawara, M., Nucl. **1971**. Phys. A175, 607-621.
10. Fuller, E. G., Petree, B., and Weiss, M., **1958**. Phys. Rev. 112, 554-562.
11. Allum, F. R., Quirk, T. W. and Spicer, B. M., Nucl. **1964**. Phys. A53, 645-655.
12. Tohei, T., Sugawara, M., Mori, S., and Kimura, M., J. **1961**. Phys. Soc. Jap. 16, 42-63.
13. Bergere, R., Beil, H., Carlos, P., and Reyssiars, A., Nucl. **1969**. Phys. A133, 417-430.
14. Muller, S., Kreschmer, A., Sonnabend, K., Zalges, A., and Galaviz, D., Cornell University online publications, arXiv:astro-ph/0512603v1 24, **Dec 2005** (" $^{187}\text{Re}(\gamma,n)$ cross section close to and above the neutron threshold"),).
15. Albayati, A. A. and Kamoon, S. S. and Saeed, M.A., , to be published **2008**. "Calculations of $(\gamma-n)$ Reaction Cross-Section in the Heavy Mass Region"
16. Nilsson, B., Adler, J. O., Andersson, B. E., J. R. M. Annand, I. Akkurt, M. J. Boland, G. I. Crawford, K. G. Fissum, K. Hansen, P. D. Harty, D. G. Ireland, L. Isaksson, M. Karlsson, M. Lundin, J. C. McGeorge, G. J. Miller, H. Ruijter, A. Sandell, B. Schroder, D. A. Sims, and D. Watts, **2007**. Phys. Rev.C75, 014007-014014.

17. Dwight Gray, E., **1972** "The American Institute Handbook of Physics," 3rd edition, McGraw Hill pub. NY.
18. Berman, B. L. and Fultz, S. C., Rev. Mod. **1975**. Phys. 47, 713-721.
19. Beckurts, K. H. and Writz, K., **1964** "*Neutron Physics*", Springer-Verlag Pub. Co., New York.
20. Wapstra, A. H. and Gave, N. B., in the "Atomic Mass Evolution, **1971**", part 2, in "*Nuclear- Reaction and Separation Energies*".303.