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Neutron Yield From Gamma Ray Incineration of Radioactive Fission Products

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Abstract

The neutron flux in this paper, which is generated as a result of γ incineration of the radioactive fisssion products isotopes has been evaluated .It is obvious from this paper that the neutron flux value depends on the number of incineration nuclei and the nuclear cross-section of the incinerated isotopes, and the neutron flux is directly dependent on γ-ray flux. The neutron flux increases from $10^{10}$to 10^{17}n/s.gm as the irradiation flux increases from $10^{16}$to 10^{20} γ/cm^2.s. It is concluded that the γ-incineration technique can be used to produce a switchable neutron source of high flux.

Keywords
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الحصيلة النيوترونية الناتجة من أحراق نواتج الأنشطار المشعة بأشعة كاما

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الخلاصة

في هذا البحث تم حساب فيض النيوترونات الناتجة من احراق المخلفات المشعة الناتجة من التفاعل الانشطاري.
اوضحت النتائج بان فيض النيوترونات يعتمد على عدد النوى المحترقة وعلى مساحة المقطع العرضي للتفاعل كذلك تبين ان فيض النيوترونات الناتجة بسبب الحرق يتناسب طرديا مع فيض كاما المستخدم لتشعيع النظائر حيث يزداد فيض النيوترونات من $١٠^{١٠}$الى $١٠^{١٧}$نيوترون/ثانية ٠غرام عندما يزداد اشعة كاما من $١٠^{١٦}$الى $١٠^{٢٠}$كاما/سم^{٢}.ثانية وعليه يمكن اعتبار هذه التقنية كمصدر نيوتروني ذو فيض عالي يمكن التحكم والسيطرة عليه.

Introduction

Energy production in nuclear power plant on basis of fission process lead inevitably to fission products and to generation of 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 long radiological half-life[1] .

Different means to ensure that the waste will not interplay with biological life which has 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, 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. An alternative option which is now under consideration, is first to incinerate and transmute the high level radioactive waste 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 isotope. The nuclear incineration method has recently been proposed to transmute these long-lived nuclei in the high-level radioactive wastes to shorter or 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[2]. On the other hand, the spallation method using a high energy proton beam has also been proposed[2], but need more development of proton accelerators. The efficiency of γ-incineration method has 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[3] , ^{90}Sr and ^{137}Cs[4] ,^{127}I[5,6] ^{138}Ba[2], $^{92,94-97}$Zr isotopes[7,8], $^{116-118,120,124}$Sn isotopes[9,11], ^{146,148,150,152,154}Sm isotopes[11,12] , and many other types of reactor waste radioactive isotopes. The technique of (γ-n) reaction were also applied to many other heavy and light isotopes[1,13,14,15].

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 [1]:

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

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

ii) The integrated photon-neutron reaction cross sections, which can be found from Thomas-Reciche-Kuhn (TRK) summation relation given as [17]:

$$\sigma = \int_0^{\infty} \sigma(E)\, dE = \frac{2\pi^2 e^2 \hbar^2}{mc} \frac{N\,Z}{A}$$

$$\cong \frac{60\ N\,Z}{A} \ (MeV.mb) \quad \text{.............}(2)$$

Where *m* is the mass of the nucleon, *c* is the speed of light, *e* is the unit charge , *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}[17]:

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

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

$$\sigma_{-1} = \int_0^{\infty} \frac{\sigma(E)}{E} dE = \frac{4\pi^2 e^2}{3\hbar c} \frac{N\,Z}{A-1} < r^2 > \quad \text{............}(4)$$

Where $<r^2>$ 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[17].

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

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:

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

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 Lorenz curves will be given as:[18]

$$\int_{0}^{\infty}\sigma(E)\,dE=\frac{\pi}{2}\sigma_{m}\,\Gamma\;\left(or=\sum_{i=1}^{2}\frac{\pi}{2}\sigma_{mi}\,\Gamma_{i}\right)\text{..... (7)}$$

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-nα), (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*+Δ*t*, then number of nuclei, *N*, produced within the time interval of (*t*+Δ*t*) is:[1,4]

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

where the function *A* is given as[18]

$$A=-\sum_{j}\left(\lambda_{j}+\sigma_{j}\,\phi\right)\quad\text{........................ (9)}$$

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 [18]

$$N(t+\Delta t)=N(0)+\sum_{i}\left(\frac{(\Delta t)^{i}}{i!}\right)A^{i}\,N(0)$$

and *N*(0)=1. Eq.(10) thus simply gives:[1]

$$N(t+\Delta t)=1+\sum_{i}\sum_{j}(-1)^{i}\left(\frac{(\Delta t)^{i}}{i!}\right)\left(\lambda_{j}+\sigma_{j}\,\phi\right)^{i}\quad\text{...(11)}$$

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

Total photo neutron cross section

$\sigma(\gamma,\text{total})=\sigma[(\gamma,n)+((\gamma,pn)+(\gamma,2n)+(\gamma,p2n)+(\gamma,3n+\ldots\ldots]$...................... (12)

Below it, where appropriate, is the single photo-neutron cross section;

$\sigma(\gamma,1n)=\sigma[(\gamma,n)+\sigma(\gamma,pn)]$ (13)

The double photo-neutron cross section is $\sigma[(\gamma,2n)+(\gamma,p2n)]$ and the triplet photo-neutron is $\sigma(\gamma,3n)$. Most of the photo neutron data are taken for medium and heavy nuclei (A>50). [18]

Cross section and yield

Below is a brief review of the theory of interest to the present research. Consider γ-ray flux interacting with a target of mass number *A* and atomic number *Z*. the γ-rays is specified by its energy *E* and flux ϕ. The yield of emitted particle of type *b* from the interaction of flux of particles of type *a* with *N* nuclei per unit volume is given as:

$$n_{b}=\phi_{a}\,\sigma_{a,b}\,N\quad\text{......................... (14)}$$

Where ϕ_a being the flux of the incident particles of type *a*, and $\sigma_{a,b}$ is the interaction cross-section of the interaction of the two types of particles; and $N=\rho N_A/A$, where N_A is Avogadro's number.[1].

Results and Discussion

The results are shown numerically in Tables (1 to 5). Each table contains the calculated number of incinerated nuclei as well as the neutron flux generated from the reaction. Also, plots of these values are shown in Figures (1) and (2), as a function of mass number of the incinerated nucleus.

Selected isotopes were chosen mainly as examples of the nuclear reactor wastes. These examples have a mass range that spans from mass number A=87 to A=155, which gives a good explanation of the present treatment since this ranges well covers the medium mass number products. Treatment of these products to re-produce more stable elements will be of great importance in the reduction of harmfulness of reactor waste, thus gives a useful and practical application.

It is more convenient to compare the results from Figure (1). As shown in this figure, the maximum neutron flux was produced from treating the ^{144}Ce, with the highest gamma-flux of 10^{20} gammas/cm^2.sec., reaching to a peak 8.125x10^{16} (n/sec.gm). A notable behavior is seen also in this gamma-flux that neutron fluxes change more smoothly from one nucleus to another, and this smooth behavior continues until the use of the 10^{17} gamma fluxes, then the results start to change more rapidly. The lowest gamma flux (10^{16} gamma/cm^2.sec) gives the most deviated results.

Another remark is seen that the neutron fluxes from ^{129}I and ^{144}Ce isotopes are always larger than other fluxes, and at all used gamma-ray fluxes. This indicates that treatment of these two isotopes as neutron sources will be

The number of incinerated nuclei, shown in Figure (2), indicates that as the gamma fluxes increase the number of transmutation nuclei also increase. An interesting remark that can be seen from this figure is that this process is more efficient at lower mass numbers, except at isotopes ^{106}Ru and ^{123}Sn, where incineration rate is almost the same at higher fluxes. This makes a useful remark when using gamma-ray incineration method of practical use in disposal of nuclear reactor wastes.

Conclusions

The nuclear reactor waste contains many instable isotopes which can be used to produce neutron sources of high fluxes using gamma-ray incineration method. The results of the present paper showed that produced fluxes change smoothly with mass number at high gamma-fluxes (10^{20} gamma/cm^2.sec), and start to deviate rapidly at fluxes as low as (10^{17} gamma/cm^2.sec). Also it was concluded that this method can be of practical importance in disposal of harmful waste of nuclear reactors.

Table (1). Neutron flux as a function of incineration nuclei (N) for γ-flux=10^{20} γ/cm^2.s

Nuclide	**Incinerate nuclei atoms/g×10^{21}**	**σ-١_(γ,sn)cm^2 ×10^{-26}**	**Flux=Nσφ (n/sec.g) ×10^{16}**
^{90}Sr	**6.24**	**7.3**	**4.550**
^{93} Zr	**5.862**	**7.2**	**4.220**
^{123} Sn	**4.807**	**11.6**	**5.576**
^{106} Ru	**5.559**	**9.2**	**5.114**
^{129} I	**4.563**	**10.0**	**4.563**
^{137}Cs	**4.357**	**13.2**	**5.750**
^{144}Ce	**4.167**	**19.5**	**8.125**
^{151}Sm	**3.980**	**18.0**	**7.164**
^{155}Eu	**3.860**	**15.8**	**6.098**
^{127}Te	**4.713**	**13.8**	**6.053**
^{87}Rb	**6.370**	**6.5**	**4.140**
^{144}Nd	**4.158**	**13.4**	**5.570**

Table (2). Neutron flux as a function of incineration nuclei (N) for γ-flux=10^{19} γ/cm^2.s

Nuclide	**Incinerate nuclei atoms/g×10^{21}**	**σ-١_(γ,sn)cm^2 ×10^{-26}**	**Flux=Nσφ (n/sec.g) ×10^{15}**
^{90}Sr	**4.970**	**7.3**	**3.620**
^{93} Zr	**4.488**	**7.2**	**2.230**
^{123} Sn	**4.325**	**11.6**	**5.017**
^{106} Ru	**4.880**	**9.2**	**4.489**
^{129} I	**3.960**	**10.0**	**3.960**
^{137}Cs	**3.980**	**13.2**	**5.250**
^{144}Ce	**3.980**	**19.5**	**7.761**
^{151}Sm	**3.795**	**18.0**	**6.830**
^{155}Eu	**3.674**	**15.8**	**5.804**
^{127}Te	**4.439**	**13.8**	**6.120**
^{87}Rb	**4.985**	**6.5**	**3.240**
^{144}Nd	**3.870**	**13.4**	**5.185**

Table (3). Neutron flux as a function of incineration nuclei (N) for γ-flux=10^{18} γ/cm^2.s

Nuclide	**Incinerate nuclei atoms/g×10^{21}**	**σ-١_(γ,sn)cm^2 ×10^{-26}**	**Flux=Nσφ (n/sec.g) ×10^{14}**
^{90}Sr	**5.766**	**7.3**	**4.209**
^{93} Zr	**5.290**	**7.2**	**3.809**
^{123} Sn	**4.848**	**11.6**	**5.620**
^{106} Ru	**5.499**	**9.2**	**5.059**
^{129} I	**4.360**	**10.0**	**4.360**
^{137}Cs	**4.252**	**13.2**	**5.610**
^{144}Ce	**4.158**	**19.5**	**8.100**
^{151}Sm	**3.938**	**18.0**	**7.088**
^{155}Eu	**3.800**	**15.8**	**6.004**
^{127}Te	**4.729**	**13.8**	**6.526**
^{87}Rb	**5.815**	**6.5**	**3.779**
^{144}Nd	**4.080**	**13.4**	**5.467**

Table (4). Neutron flux as a function of incineration nuclei (N) for γ-flux=10^{17} γ/cm^2.s

Nuclide	Incinerate nuclei atoms/g×10^{21}	σ -١_(γ,sn)cm ^{2} ×10^{-26}	Flux=Nσφ (n/sec.g) ×10^{13}
^{90}Sr	1.323	7.3	0.97
^{93} Zr	1.013	7.2	0.73
^{123} Sn	4.322	11.6	5.01
^{106} Ru	3.510	9.2	1.11
^{129} I	1.111	10.0	4.360
^{137}Cs	1.336	13.2	1.76
^{144}Ce	3.080	19.5	6.01
^{151}Sm	1.428	18.0	2.57
^{155}Eu	2.140	15.8	3.38
^{127}Te	4.446	13.8	6.14
^{87}Rb	1.159	6.5	0.75
^{144}Nd	1.319	13.4	1.76

Table (5). Neutron flux as a function of incineration nuclei (N) for γ-flux=10^{16} γ/cm^2.s

Nuclide	Incinerate nuclei atoms/g×10^{21}	σ-١_(γ,sn)cm ^{2} ×10^{-26}	Flux=Nσφ (n/sec.g) ×10^{11}
^{90}Sr	0.290	7.3	2.11
^{93} Zr	0.109	7.2	0.78
^{123} Sn	4.152	11.6	48.00
^{106} Ru	2.890	9.2	26.50
^{129} I	0.125	10.0	1.25
^{137}Cs	0.243	13.2	3.21
^{144}Ce	2.550	19.5	49.70
^{151}Sm	0.202	18.0	3.64
^{155}Eu	1.342	15.8	21.20
^{127}Te	4.320	13.8	59.60
^{87}Rb	0.125	6.5	0.81
^{144}Nd	0.155	13.4	2.08

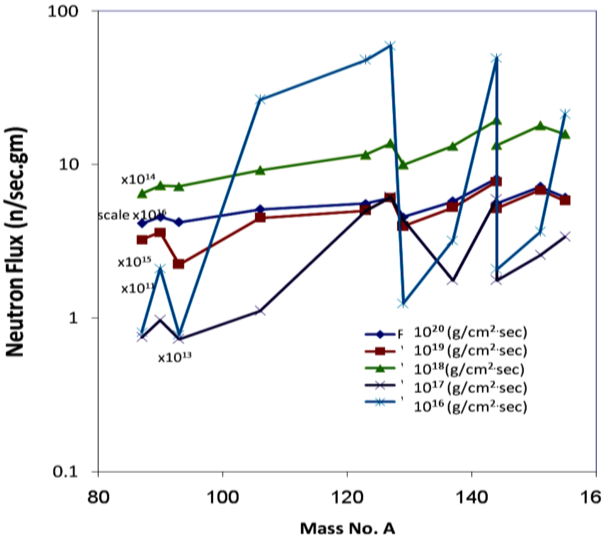

Fig. (1). Neutron flux as a function of mass number for different incident gamma-ray fluxes. Notice the different scales of each curve, as indicated next to them.

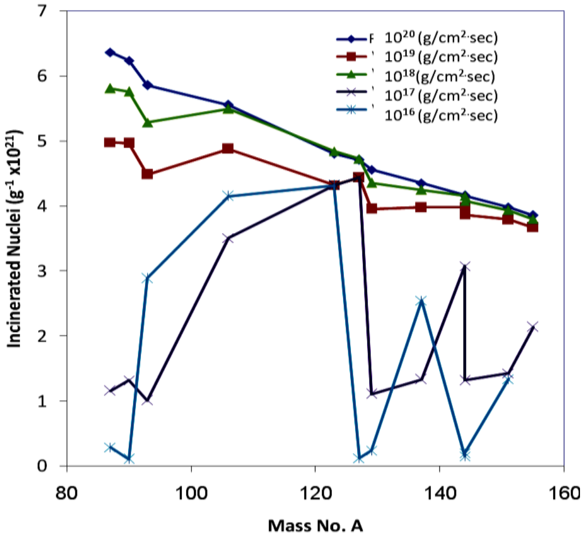

Fig. (2). Incineration of Nucleide as a function of mass number for different incident gamma-ray fluxes.

References

[1] M. A. Saeed, "*Calculation of γ-n Reaction Cross Sections For Radioactive Fission Products,*" M. Sc. Thesis, Department of Physics, College of Science, University of Baghdad, **(2002)**.

[2] C. H. M. Broeders, E. Kiefhaber and H. W. Wiese, *Nucl. Eng. Des.* 202, **(2000),** 157.

[3] P. M. Brown, *"Photo-Transmutation for Waste*" **(2003).**

[4] T. Matsumoto, *Nucl. Inst. Meth. Phys. Res.* A268,**(1988),** 234

[5] R. P. Rassoll and M. N. Thompson, *Phys. Rev.* C39, **(1989)**, 2201.

[6] R. L. Brmblett, J. T. Caldwell, B. L. Berman, R. R. Harvey and S. C. Fultz, *Phys. Rev*. 148, **(1966)**, 1198.

[7] B. L. Berman, J. T. Caldwell, R. R. Harvey, M. A. Kelly, R. L. Brmblett, and S. C. Fultz, *Phys. Rev*. 162, **(1967)**, 1098.

[8] A. Lepratre, H. Beil, R. Bergere, P. Carlos, A. Reyssiars, and M. Sugawara, *Nucl. Phys*. A175, **(1971)**, 607.

[9] E. G. Fuller, B. Petree, and M. Weiss, *Phys. Rev.* 112, **(1958)**, 554.

[10] F. R. Allum, T. W. Quirk and B. M. Spicer, *Nucl. Phys*. A53, **(1964),** 645.

[11] T. Tohei, M. Sugawara, S. Mori and M. Kimura, *J. Phys. Soc. Jap*. 16**, (1961),** 42.

[12] R. Bergere, H. Beil, P. Carlos, A. Reyssiars, *Nucl. Phys*. A133, **(1969)**, 417.

[13] S. Muller, A. Kreschmer, K. Sonnabend, A. Zalges, and D. Galaviz, Cornell University online publications, *arXiv:astro-ph/0512603v1* 24 Dec (**2005)**, "*^{187}Re(γ,n) cross section close to and above the neutron threshold*".

[14] M. A. Saeed, "*Calculations of(γ-n) Reaction Cross-Section in the Heavy Mass Region*", to be published.

[15] B. Nilsson et al., *Phys. Rev*.C75, **(2007)**, 014007.

[16] E. Dwight Gray, "The American Institute Handbook of Physics," 3rd edition, McGraw Hill pub. NY **(1972)**.

[17] B. L. Berman and S. C. Fultz, *Rev. Mod. Phys*. 47, **(1975)**, 713.

[18] Al-Bayati,A.A and Kamoon ,S.S and Saeed M.A.,Iraqi Journal of Science,Vol.49, **(2008)**, No.2 , p.p101.