E1-2001-44

MONTE CARLO CALCULATIONS ON TRANSMUTATION OF TRANSURANIC NUCLEAR WASTE ISOTOPES USING SPALLATION NEUTRONS. DIFFERENCE OF LEAD AND GRAPHITE MODERATORS

Submitted to «Nuclear Instruments and Methods A»

S.R.Hashemi-Nezhad[1]
Department of High Energy Physics, School of Physics, University of Sydney, A28 NSW 2006, Australia

R.Brandt, W.Westmeier[2]
Institut fuer Physikalische, Kern- und Makromolekulare Chemie, FB 15, Philipps-Universitaet, Marburg, Germany

V.P.Bamblevski, M.I.Krivopustov, B.A.Kulakov, A.N.Sosnin
Joint Institute for Nuclear Research, Dubna, Russia

J.-S.Wan
Northwest Institute of Nuclear Technology, 710024 Xian, China

R.Odoj
Institut fuer Sicherheitsforschung und Reaktorsicherheit, Forschungszentrum Juelich GmbH, Juelich, Germany

[1]E-mail: reza@physics.usyd.edu.au
[2]Permanent address: GmbH, 35085 Ebsdorfergrund, Germany

1. Introduction

Ion accelerators with high beam intensities (10-25 mA) make it possible to envisage Accelerator Driven Systems (ADS) for energy production and nuclear waste incineration [1-4]. In such systems the large number of neutrons that result from the interaction of high-energy ions (e.g. protons) with massive targets such as lead, can be used to operate sub-critical nuclear assemblies. The chain reaction in an ADS is sustained by continuous operation of the driving accelerator and the system remains sub-critical at all time. This sub-criticality is the key issue and is the main advantage of ADS over conventional nuclear systems that operate *only* in critical conditions. A sub-critical system is not only a much safer system than a critical one, but it provides the opportunity to use the excess neutrons in the system for other purposes such as nuclear waste transmutation and eventually nuclear waste incineration as well as breeding of fissile material from fertile isotopes such as ^{232}Th.

It is suggested that an effective method for nuclear waste transmutation is to use non- thermal neutron captures in the resonance regions of the absorption cross section of the waste isotopes [5,6]. This method is known as Transmutation by Adiabatic Resonance Crossing (TARC). A neutron spectrum suitable for TARC can be obtained if spallation neutrons are moderated in lead [6]. The transmutations of the long-lived nuclear waste isotopes such as ^{99}Tc ($t_{1/2}$ = 2.1×10^5 y) and ^{129}I($t_{1/2}$ = 2×10^7 y) have been studied by this method (6). It has been also shown that ^{129}I, ^{237}Np ($t_{1/2}$ = 2.14×10^6 y) and ^{239}Pu ($t_{1/2}$ = 2.44×10^4 y) can be transmuted at quite acceptable rates in thermal (slow) neutron dominated neutron fields [7,8].

In two earlier papers [9,10] we have reported the spallation neutron yield, neutron energy spectrum, spatial distribution of neutrons, energy gain (amplification) of ADS and transmutation of some radioactive nuclear waste isotopes in several sub-critical systems. In one of these papers [10] we have shown that the transmutation rates of ^{99}Tc and ^{129}I in an ADS with graphite moderator is significantly higher than the case when lead was used as moderator.

In the present paper we report the results of our calculations on the transmutation of some trans-uranic nuclear waste isotopes in two accelerator driven systems with lead and graphite moderators. The transmutation of ^{99}Tc and ^{129}I also has been revisited with more details. Furthermore the fissile material breeding from fertile isotopes using the above-mentioned systems was also investigated.

2. Transmutation process

The aim of the nuclear waste incineration is to transmute a given long-lived nuclear waste isotope to a nuclear species that is either stable or has a half-life that is much less than that of the original waste isotope itself. An important parameter that should be considered is that the destruction rate of the waste isotope must be

significantly higher than its possible production (by any means) in the accelerator driven system concerned.

For trans-uranic isotopes the fission process is a very effective way of incineration. However some of these isotopes have very small fission cross-section (e.g. ^{237}Np, σ_f = 19 mb). Then again the (n,γ) and other nuclear processes on the waste isotopes may transfer them to nuclei that have much higher fission cross-section. Figure 1 shows some incineration channels for trans-uranic isotopes starting with ^{241}Am. The cross-section and half-life values are taken from Refs 11 and 12 and cross-section values refer to thermal neutrons.

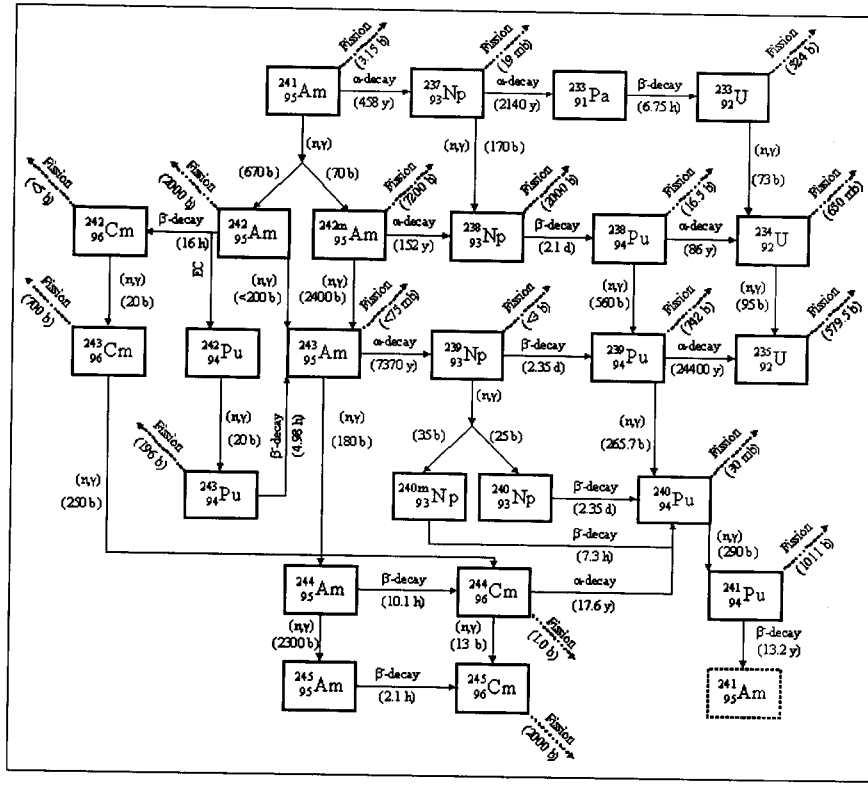

Fig. 1. Transmutation and incineration of some trans-uranic waste isotopes. The cross-section values are for thermal neutrons.

Figure 2 illustrates the variation of the absorption, fission and (n,2n) cross-section with neutron energy for ^{241}Am. The cross-section data are from ENDF/B-VI data library [13]. As it can be absorption and fission cross-sections contain

many resonances in the energy range of 0.2-120 eV. For ^{241}Am this is the energy region that is mainly exploited in TARC method.

Fig.2 also shows that ^{241}Am(n,γ) reaction, is the most important transmutation channel for this isotope and produces ^{242}Am and ^{242m}Am. The cross section for the formation of ^{242}Am is 9.6 times higher than that for ^{242m}Am (Fig.1). Both ^{242}Am and ^{242m}Am, fission in thermal neutron field with huge cross-sections of 2100 b and 7200 b respectively.

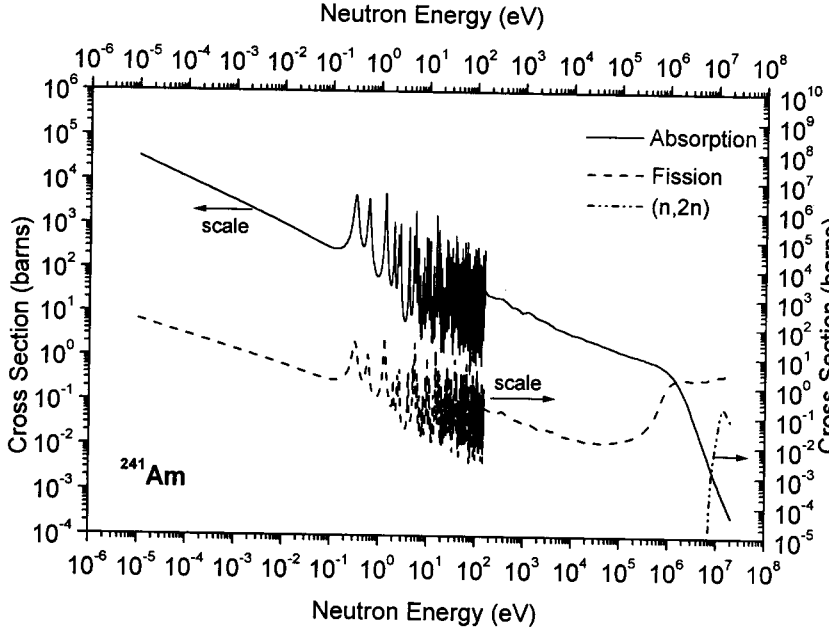

Figure. 2. Variation of the absorption, fission and (n,2n) cross-section with neutron energy for ^{241}Am. Data from ENDF/B-VI library.

In an accelerator driven incinerator, regardless of its neutron-moderating environment, fast neutrons will be present [9,10]. There exist the possibility of ^{241}Am(n, 2n)^{240}Am reactions. Such reactions will lead to the formation of ^{241}Pu which fissions with σ_f = 1011 b, Fig.3.

$$^{241}Am(n,2n)^{240}Am \xrightarrow[51h]{EC} {}^{240}Pu(n,\gamma)^{241}Pu \xrightarrow[\sigma_f=1011b]{fission} \rightarrow$$

Figure 3. Transmutation and incineration of ^{241}Am by (n,2n) reaction. The fission cross-section refers to thermal neutrons.

Calculations show that even in a lead neutron-moderating environment where energetic neutrons are more abundant than say in a graphite moderator [10], the contribution of the (n,2n) reaction is 2-3 orders of magnitude less than those of fission and (n,γ) reactions. Therefore, although α-decay and (n,2n) reactions ***can lead*** to the incineration of trans-uranic waste isotopes, contribution of these two phenomena will not be significant (due to very long half-lives of the α-decaying nuclei and because of relatively low cross section for (n,2n) reaction). We can extend more or less the same arguments to other trans-uranic isotopes (^{237}Np, ^{239}Pu, ^{245}Cm and ^{246}Cm) that we consider in this paper.

In the light of the above given discussion, for trans-uranic nuclei we calculated only the rates of (n,f) and (n,γ) reactions.

3. Calculation procedure

We used the LAHET code system [14] to calculate the spallation neutron induced transmutation yield of the long-lived radioactive waste isotopes. We used large-scale moderator systems (~30 m^3) without presence of a specific nuclear fuel in the system.

In the present paper the two target moderator systems that will be considered are: (a) Lead target and lead moderator, (Pb, Pb,0) system and (b) Lead target and graphite moderator, (Pb,C,0) system . The "0" in the bracket indicates that system does not contain a specific nuclear fuel. Possible effects of the fission of the waste isotopes in the system will be discussed. Fig.4 shows XZ-cross section of the all components of the target-moderator assemblies used in the calculations. The origin of the Cartesian co-ordinate system is at the centre of the assembly.

In all of the above-mentioned cases the moderator occupied a volume of 3.3×3.3×3 m^3. The target was embedded in the moderator as shown in Fig.4. In the case of the (Pb, Pb,0) the whole ~370 tons of lead acts as target and moderator. While in the (Pb, C,0) system a cylindrical lead target of diameter 20 cm and length of 1 m was placed on the Z-axis of the system, starting from Z = -30 cm (Fig.5). This target length is sufficient to produce maximum number spallation neutrons as long as the target length is concerned [9].

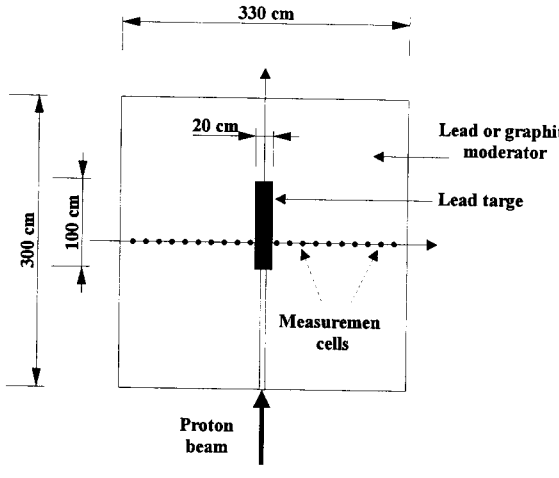

Figure 4. XZ-cross-section of the target-moderator assemblies used in the LAHET Monte Carlo simulations. The origin of the Cartesian co-ordinate system corresponds to the centre of the assembly. The two cases that were studied are; (a) lead target and moderator (b) lead target and graphite moderator. (For details of the set-ups refer to the text.)

In all target-moderator assemblies 20 spheres of diameter 1.5 cm were located on the x-axis (either side of the origin) at intervals of 15 cm, which were used as measurement (calculation) cells (Fig. 4). These cells are either "filled" with the nuclear waste isotope of interest or with the relevant moderator material when "not used".

In all calculations protons of energy 1.5 GeV were used. The proton beam was introduced into the system along the Z-axis through a 1.2 m long blind hole of diameter 6 cm. The proton beam had circular cross-section with a diameter of 1 cm.

The LAHET code system [14] is coupled with MCNP code [15]. LAHET deals with the spallation processes involving hadronic interactions and transport of the produced neutrons (in our case) down to 20 MeV. The neutrons of energy less than 20 MeV are transported with MCNP code, which in our case was version MCNP-4B2 [15].

In LAHET calculations we used the following options in the input file (ref. 14 and references therein); 1) Bertini model of intranuclear cascade, 2) Pre-equilibrium model following the intranuclear cascade, 3) Gilbert-Cameron-Cook-Ignatyuk level density model 4) Coulomb barrier on incident charged particle interactions and 5) Rutherford-Appleton Laboratory evaporation-fission model.

In this paper the term "spallation neutrons" refers to neutrons produced by high-energy proton reactions in the intranuclear cascade, evaporation and high-

energy fission processes (ref. 14 and references therein) as well as those produced in internuclear- cascade.

For every calculation 25,000 incident proton histories were followed. The cross-section libraries used in calculations were those provided with MCNP-4B2 for neutron transport and dosimetry calculations.

The results are given in terms of B, which is defined as; the number of a given reaction in one gram of the isotope of interest per incident proton of 1.5 GeV. In calculating the B-values only neutrons of energy less than 20 MeV were taken into account. This energy limit is imposed by MCNP code. This procedure imposes very small errors in our calculations, because:

1) Flux of neutrons with energy above 20 MeV is very small. For a cylindrical lead target of diameter 8 cm and length 20 cm number of neutrons with energy greater than 20 MeV that escape from the target is only ~10% of the total number of neutrons, for larger targets this will be much less.
2) Neutron capture cross-section for isotopes of interest at neutron energies greater than 20 MeV is very low.

4. Results and discussion

In this work we are mainly concerned with the comparison of the transmutation abilities of (Pb,Pb,0) and (Pb,C,0) systems. Therefore energy production from these systems is not considered and effective multiplication coefficient k_{eff}, of the systems is very low if not zero.

Table 1 gives the characteristics of graphite and lead moderators. As it can be seen these two materials have quite different moderating properties.

Table 1. Some properties of graphite and lead moderators, Σs and Σa are macroscopic scattering and absorption cross-sections respectively.

Moderator	Average logarithmic energy decrement, ξ	Slowing down power ξΣs	Moderation ratio ξΣs/Σa	N_1*	N_2**	Moderation time of fission neutrons to1eV (μs)
Graphite	0.158	0.061	170	151	298	23
Lead	0.00962	0.0035	0.623	2632	5199	400

* Number of neutron collisions required for crossing energy interval of 10 keV –1 eV.

** Number of neutron collisions required for crossing energy interval of 2 MeV–0.025 eV.

Table 2 gives the spallation neutron yield per incident 1.5 GeV proton, Y_n, and total number of the neutrons that leak out of the systems, Y_l. A bare target of D = 20 cm and L = 100 cm produces only $Y_b = 38.82 \pm 0.02$ neutrons. The difference ($Y_n - Y_b$) represents the number of neutrons that are produced within the corresponding moderator per incident proton. Although the neutron yield in lead moderating environment is 22% higher than that in the graphite system, because of higher neutron leakage from lead system the overall average neutron

density $q = (Y_n - Y_l)/V$ in two systems is very close, where V is the total volume of the target-moderator assembly (Fig.4). In other words number of neutrons that are absorbed in a given target-moderator assembly discussed in this paper ($Y_a = Y_n - Y_l$) is the same and is equal to $Y_a = 38.4$ neutrons per proton.

Table 2. Spallation neutron yield and neutron leakage from the systems per incident proton of Ep = 1.5 GeV.

ADS system	Spallation neutron yield, Y_n	Neutron leakage from the system, Y_l
(Pb,Pb,0)	57.27 ± 0.11	18.83 ± 0.06
(Pb,C,0)	44.55 ± 0.11	6.10 ± 0.02

The trans-uranic waste isotopes that were studied are; ^{237}Np ($t_{1/2} = 2.14\times10^6$ y), ^{239}Pu ($t_{1/2} = 2.44\times10^4$ y), ^{241}Am ($t_{1/2} = 458$ y), ^{245}Cm ($t_{1/2} = 9.3\times10^3$ y) and ^{246}Cm ($t_{1/2} = 5.5\times10^3$ y). The breeding rates of the ^{239}Pu from ^{238}U and ^{233}U from ^{232}Th were also calculated. Although some calculation results on the incineration of ^{99}Tc and ^{129}I are given in an earlier work [10], in this paper we report more detailed transmutation rates for these two isotopes at proton energy of 1.5 GeV.

4.1 Transmutation of ^{239}Pu in presence and absence of ^{238}U

We consider the transmutation of ^{239}Pu and ^{238}U together. If the ADS contains ^{238}U in any form, then ^{238}U(n,γ) reactions will result in production of ^{239}Pu. In such case, the incineration will make sense only if the destruction rate is significantly higher than the production rate, unless the system is deliberately designed for ^{239}Pu breeding.

In both target moderator assemblies the cells with $x \geq 15$ cm were filled with ^{239}Pu while the cells with $x \leq -15$ cm contained natural uranium. In total the assemblies contained 270.4 g of ^{239}Pu and 332 g of natural uranium. For these set-ups the k_{eff}-values of $(k_{eff})_{Pb} = 0.14444 \pm 0.00059$ and $(k_{eff})_C = 0.17821 \pm 0.00176$ were calculated for (Pb,Pb,0) and (Pb,C,0) systems respectively, using MCNP-4B2 code. Effects of such difference in the k_{eff}-values of the two systems, will be discussed later in this paper.

In the case of the of the ^{239}Pu, the transmutation rates by (n,f) and (n,γ) reactions were calculated. Successive (n,γ) reactions can lead to the formation of ^{241}Pu which fissions with a high cross-section of 1011 b (Fig.1). For ^{238}U only the B-value for (n,γ) reaction was calculated, which is the equivalent of B-value for ^{239}Pu breeding.

Fig.5 shows the transmutation rates of ^{239}Pu by (n,f) and (n,γ) and ^{238}U via (n,γ) reactions as a function of position, x in (Pb,Pb,0) and (Pb,C,0) systems. The variation of ratio of the B-values of plutonium to uranium, at different locations on the X-axis is also shown in the Fig.5. In (Pb,Pb,0) system on average ^{239}Pu-destruction via fission is higher by a factor of 7.4 than ^{239}Pu-breeding via ^{238}U

(n,γ) i.e. $R_1 = \langle (B_{Pu})_{(n,f)}/B_U \rangle = 7.4$ (Fig.5c). This ratio increases to $R_2 = 12.3$ if one includes the $^{239}Pu(n,\gamma)$ reactions in the calculations as well (Fig.5c).

For (Pb,C,0) system we obtain $R_1 = 11$ and $R_2 = 15.5$ (Fig.5d), suggesting that the destruction to production ratio in graphite system is higher than that in the lead system.

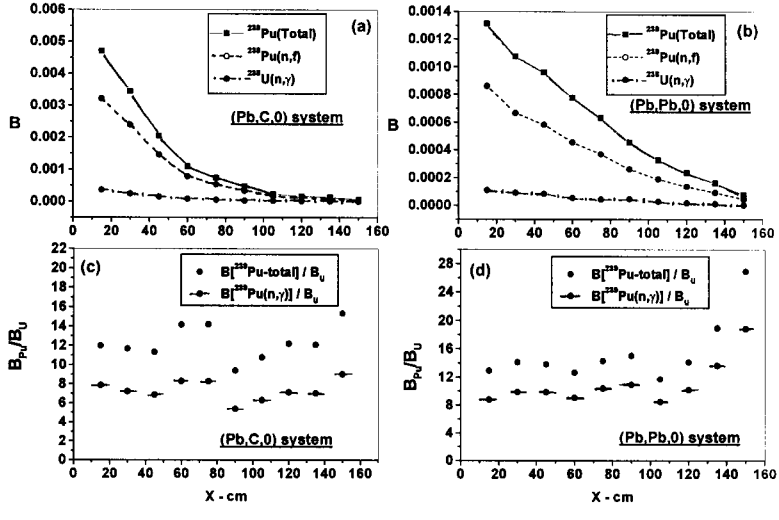

Figure 5. (a) and (b) Transmutation of ^{239}Pu by (n,f) and (n,γ) and ^{238}U via (n,γ) reactions in (Pb,Pb,0) and (Pb,C,0) systems at proton energy of 1.5 GeV. (c) and (d) variation of ratio of the B-values of plutonium to uranium, with position on the X-axis. In the legends of (c) and (d) B[^{239}Pu-total] refer to B-$^{239}Pu(n,f)$ + B- $^{239}Pu(n,\gamma)$ and B_U represents $^{238}U(n,\gamma)$.

To compare the transmutation capabilities of the two ADS at different x-values the ratio of B-value in (Pb,C,0) system, B_C, to the that in (Pb,Pb,0) system, B_{Pb}, was calculated ($R_B \equiv B_C/B_{Pb}$). Fig.6 shows the variation of the R_B with x for ^{239}Pu. It can be seen that ^{239}Pu destruction is higher in (Pb,C,0) system for all values of $x \leq 90$ cm.

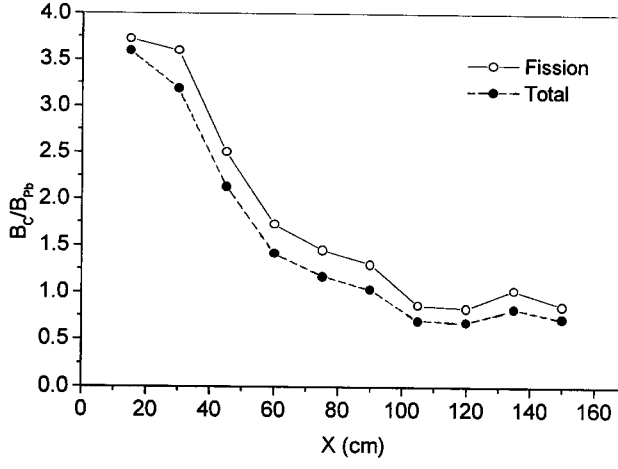

Figure. 6. Variation of the ratio of the transmutation rates in of ^{239}Pu in (Pb,C,0) and (Pb,Pb,0) systems ($R \equiv B_C/B_{Pb}$), as a function of distance x, on the X-axis (Fig.1). The term "Total" in the legend refers to (n,γ) + (n,f) reactions.

Table 3 gives the average B-value for the reactions studied. The averages were taken over B-values of the 10 cells containing the specific isotope in the systems. Such a <B> is used only for a rough comparison of the transmutations in two systems studied.

Table 3. Average B-values for transmutation and production of ^{239}Pu in (Pb,Pb,0) and (Pb,C,0) systems. The average is taken over ten calculation cells (see the text for details).

Type of reaction	(Pb,C,0) system (B_C)	(Pb,Pb,0) system (B_{Pb})	(B_C/B_{Pb})
^{239}Pu(n,f)	9.15×10^{-4}	3.67×10^{-4}	2.49
^{239}Pu(n,f)+^{239}Pu(n, γ)	1.31×10^{-3}	6.02×10^{-4}	2.18
^{238}U(n,γ)	9.64×10^{-5}	5.02×10^{-5}	1.92

From these results we conclude that regardless of the presence or absence of ^{238}U in the system, (Pb, C,0) system transmutes ^{239}Pu in a significantly higher rate than (Pb,Pb,0) system. This is more pronounced if ^{239}Pu-incineration takes place in absence of ^{238}U. Also the breeding rate of ^{239}Pu from ^{238}U is higher in the (Pb,C,0) system.

4.2 Transmutation of ^{237}Np, ^{241}Am, ^{245}Cm and ^{246}Cm isotopes

The B-value calculations for these isotopes were performed in two stages. The transmutation of ^{237}Np and ^{241}Am were calculated together, ^{237}Np occupying

the x > 0 cells and Am the x<0 cells (Fig.4). The second calculation involved the two Cm isotopes.

Figure 7 shows the variation of R_B as a function of x for ^{241}Am, ^{237}Np, ^{245}Cm and ^{246}Cm. In the case of ^{237}Np and ^{241}Am in both graphite and lead assemblies, transmutation by fission process is much less than that by (n,γ) reactions. This is expected from the big difference in cross-sections for these two reactions (Fig.1). The (n,γ) reactions of these isotopes results in isotopes with high fission cross-sections (Fig.1). In Figs 7a and 7b the R_B, represents the ratios of B-values for fission and (n,γ) combined.

In the case of ^{246}Cm the (n,γ) reaction is the dominant process while for ^{245}Cm fission is the most effective way of transmutation. For these two isotopes in Fig.7c and 7d R_B as a function of distance is plotted both for fission and total B-values.

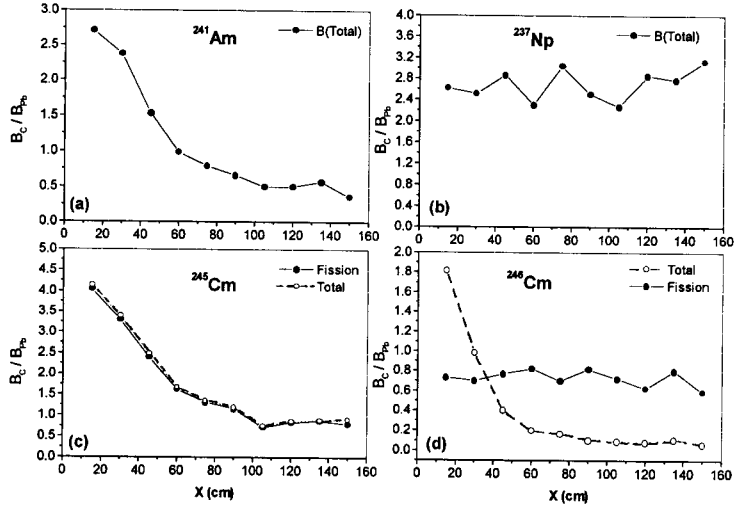

Figure 7. Variation of the ratio of the transmutation rates in of ^{241}Am, ^{237}Np, ^{245}Cm and ^{246}Cm in (Pb,C,0) and (Pb,Pb,0) systems ($R_B \equiv B_C/B_{Pb}$), as a function of distance x, on the X-axis (Fig.1). The term "Total" in the legend refers to (n,γ) + (n,fission) reactions.

From Fig.7 we note the following:

1) For x ≤ 60 cm the (Pb,C,0) system transmutes ^{241}Am at a higher rate than the (Pb,Pb,0) system. At distances x > 60 cm, (Pb,P,0) is more effective transmuter for this isotope, Fig.7a.
2) The (Pb,C,0) system transmutes ^{237}Np at higher rate (by a factor of ~2.5) than the (Pb,Pb,0) system at all values of $15 \leq x \leq 150$ cm, Fig. 7b.

3) For $x \leq 90$ cm the (Pb,C,0) system transmutes ^{245}Cm at higher rate than the (Pb,Pb,0) system. At distances $x > 90$ cm, the (Pb,Pb,0) is more effective transmuter for this isotope, Fig.7c.
4) In the case of ^{246}Cm the situation is different. Transmutation by fission is higher when the moderator is lead in entire volume of the systems studied. The total transmutation rate (fission + (n,γ)) is higher in graphite moderator only for $x < 35$ cm, Fig. 7d.

Figure 8 shows the variation of the B with x, for ^{246}Cm in the two systems studied. Fig.9 illustrates the energy dependent fission and absorption cross-section of ^{246}Cm (data from ENDF/B-VI library). The results presented in Fig.8 can be understood by examination of Fig.9. The slow neutron cross-sections for this isotope is relatively low and its excitation function contains several high cross-section resonances.

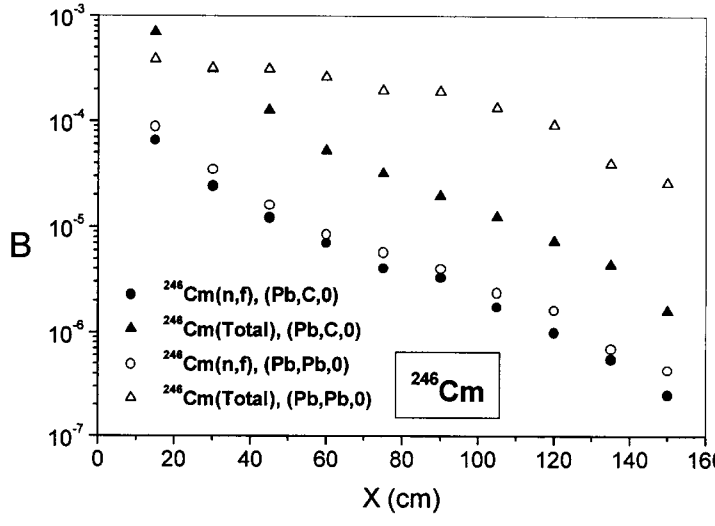

Figure 8. Variation of the B-values of ^{246}Cm with distance on the X-axis in (Pb.Pb,0) and (Pb,C,0) assemblies for incident protons of energy 1.5 GeV. The term "Total" in the legend refers to (n,γ) + (n,fission) reactions.

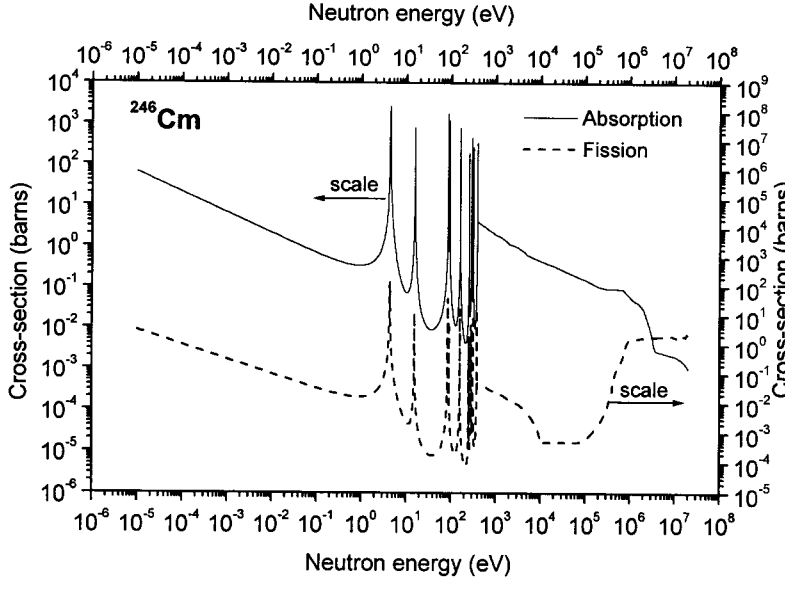

Fig.9 Energy dependent absorption and fission cross-section of ^{246}Cm. Data from ENDF/B-VI library.

4.3. Transmutation of ^{232}Th and ^{233}U breeding

In a conventional nuclear reactor the Pu and minor actinides (PMA) are produced by chain of (n,γ) reactions and radioactive decays, starting with ^{238}U(n,γ) reaction, present in this type of reactors as components of the fuel elements. To avoid the formation of PMA the logical approach is not to use ^{238}U in the fuel system. This can be achieved by breeding fissile ^{233}U from fertile ^{232}Th via (n,γ) reaction. The energy dependent activation cross-section of ^{232}Th contains large number of resonances in the energy range of 10 eV - 100 keV and it has σ_a = 7.6 b for thermal neutrons.

Fig. 10a shows the variation of the transmutation rate for the ^{232}Th(n,γ) reaction (^{233}U production rate) as a function of distance x, in the (Pb,Pb,0) and (Pb,C,0) systems. In Fig.10b the ratio of the transmutation rates in graphite and lead systems as function of x, is illustrated. For $x \leq 60$ cm ^{233}U breeding in (Pb,C,0) is higher than that in (Pb,Pb,0) system. For $x > 60$ cm the fast neutron breading overtakes the slow neutron effect. But on average transmutation rate in C-system is 1.11 times higher than that in Pb-system (for $x > 15$ cm). If the average B-value is taken for cells with $x \leq 60$ cm the average of R increases to 1.97.

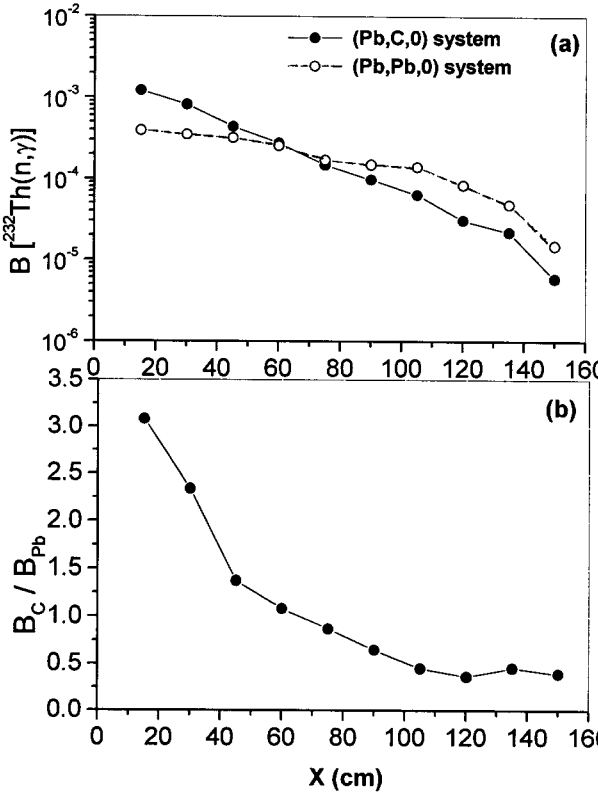

Fig.10. Transmutation of ^{232}Th (^{233}U breeding) as a function of distance in (Pb,Pb,0) and (Pb,C,0) systems

4.4. Transmutation of ^{99}Tc and ^{129}I

In an earlier work [10] we have reported the results of the LAHET calculations on the transmutation of ^{99}Tc and ^{129}I at proton energies of 2.5 GeV/c and 3.75 GeV/c in target moderator assemblies identical to those used in current work. The calculations in the case of the 3.75 GeV/c and in (Pb,Pb,0) system were in excellent agreement with experimental findings of the others [15]. In present work we report our results on transmutation of these isotopes at E_p = 1.5 GeV with more detail. Fig. 11 shows the variations of the B for these two isotopes as a function of distance from the centre of the assemblies on the X-axis, for (Pb,C ,0) and (Pb, Pb,0) systems, Figs 11a and 11b. Fig. 11c illustrates the variations of the R_B as a function of x for ^{99}Tc and ^{129}I. From Fig.11 we conclude that:

1) Transmutation rate of ^{99}Tc, in the (Pb,C,0) system is higher than that in the (Pb,Pb,0) system up to x = 60 cm (by a factor of more than 4 at x=15 cm) and at x > 60 cm they are about the same.
2) For ^{129}I the B-values in (Pb,C,0) are higher than their corresponding values in (Pb,Pb,0) for all sample locations studied (by a factor of more than 14 for x = 15 cm).

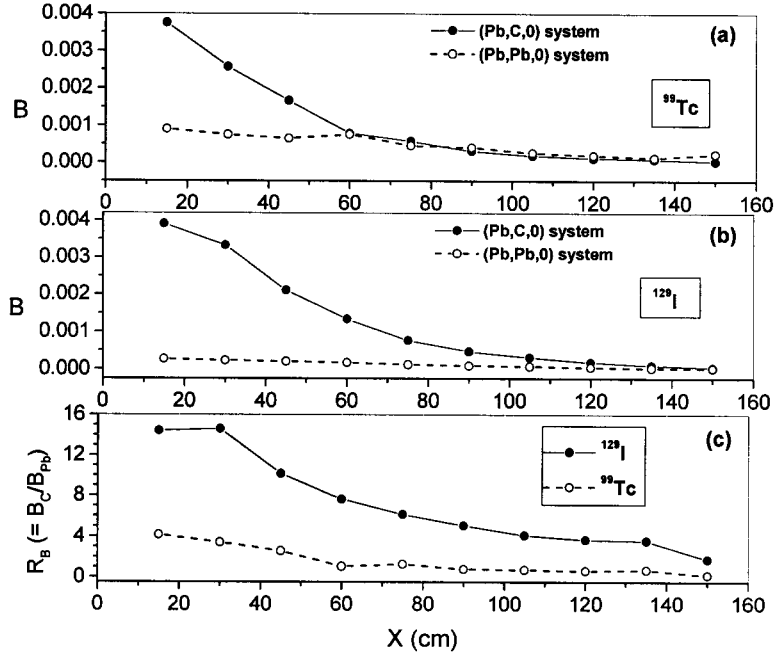

Fig. 11. Transmutation of ^{99}Tc and ^{129}I in (Pb,Pb,0) and (Pb,C,0) systems

5. Effects of self-shielding

The transmutation rates reported in this paper so far are definitely affected by self-shielding. To reduce influence of the self-shielding we need to decrease the waste to moderator atomic density ratios in the calculation cells significantly.

From the waste isotopes studied in this work we choose ^{99}Tc and ^{129}I for self-shielding effect modeling. The resonance integral I_{Res} for ^{99}Tc and ^{129}I are 300 b and 50 b respectively [17]. These two resonance integral values represent isotopes with relatively high and medium I_{Res}, thus the conclusions reached in the cases of ^{99}Tc and ^{129}I can be generalized to other waste isotopes with some confidence.

To dilute (reduce) the amount of ^{99}Tc and ^{129}I in calculation cells, we changed their content from pure waste isotope to a mixture of waste isotope of interest and the relevant moderator. We chose a waste density of 0.2 g cm^{-3} and added enough moderator material so that the overall density of the material in calculations cells remained same as the relevant moderator. In this way the density of the ^{99}Tc and ^{129}I in the cell were reduced by factors of 57.5 and 24.7 respectively compared with earlier calculations. Therefore each calculation cell contained 353 mg of ^{99}Tc or ^{129}I, distributed (uniformly in lead or graphite) in a volume of 1.77 cm^3. In other words in (Pb,Pb,0) the waste content of the cells was 17.4 mg/g while in (Pb,C,0) it was 95 mg/g. B-values for these set-ups were calculated and results are shown in Fig.12.

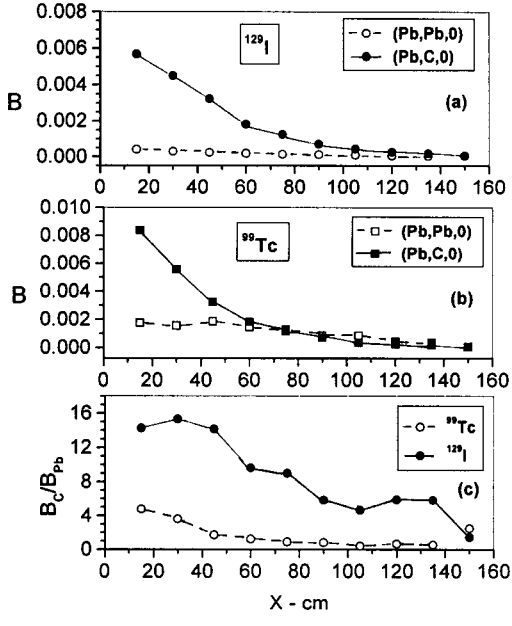

Figure 12. . Transmutation of ^{99}Tc and ^{129}I in (Pb,Pb,0) and (Pb,C,0) systems. The results shown in this figure represent the cases that the density of the ^{99}Tc and ^{129}I in calculation cells reduced by factors of 57.5 and 24.7 respectively.

From Figs. 11 and 12 we can note that:

1) As a result of reducing the densities of ^{99}Tc and ^{129}I in the calculation cells, the B-values increase in both (Pb, Pb, 0) and (Pb, C, 0) systems on average by factors of ~2 for ^{99}Tc and ~1.4 for ^{129}I.
2) For a given isotope the variation of the R_B with x (Fig. 12c) is quite similar to the case when the samples were much denser (Fig.11c). This results in

the conclusion that, the diluted waste isotopes in (Pb,C,0) system are transmuted much more efficiently than in (Pb,Pb,0) system.

Further reduction of the waste density in the cells is expected to lead to similar conclusions.

6. Further discussions and conclusions

The results reported in the present paper apply to the cases of lead and graphite neutron moderating environments that do not contain significant amount of fissile materials. We refer to these systems as none-multiplying accelerator driven systems [10]. In these assemblies spallation neutrons are generated around the centre of the system, in a relatively small region of the target-moderator. Then neutrons are distributed in the entire volume of the ADS according to the characteristics of the neutrons and moderator (i.e. energy spectrum of the source neutrons and macroscopic scattering and absorption cross-sections of the moderator).

It is already shown that neutron flux attenuates exponentially with increasing distance from the centre of the assemblies [10] and as is expected, different attenuation lengths are associated with different moderators. Also it is shown [10] that in a multiplying sub-critical ADS the neutron flux can be very high, depending on the k_{eff} of the system and intensity of the incident protons [10]. Addition of fissile materials into the system and therefore introduction of neutrons with fission spectrum, does not alter the energy spectrum of the neutrons significantly, but it totally changes the spatial distribution of the neutrons in the assembly of interest.

In non-multiplying ADSs and for the sample arrangements used in this work, the TARC in (Pb,Pb,0) over takes the transmutation in (Pb,C,0) system only when the neutron flux in (Pb,C,0) system reduces significantly with increasing distance x. For most of the isotopes studied in the present work, this happens for $x > 60$ cm, at which the neutron flux in (Pb,C,0) system is reduced to less than 19% of its value at $x = 15$ cm.

Therefore it is feasible, and in fact it is possible to design multiplying accelerator driven systems (say with $k_{eff} = 0.95$) in which the spatial distribution of the neutron flux is relatively uniform and the high neutron flux is extended almost over the entire useful volume of the ADS. The shape of the neutron flux inside the ADS can be considerably flattened by the action of appropriate reflectors. Moreover the neutron flux distribution in various zones of ADS can be made uniform by suitably arranging fuel (elements), having different fissile material content. This is a situation, similar to that one tries to achieve in conventional critical reactors.

Let us assume that neutron multiplying is achieved by introduction of ^{233}U fissile fuel in the systems (in required form). Then in (Pb,Pb, ^{233}U) system we will have high neutron flux suitable for TARC in the entire useful volume of the ADS

and in (Pb,C, ^{233}U) system the high flux of neutrons in the whole volume will be dominated by slow neutrons. It must be noted that the slow neutron flux in (Pb,Pb, ^{233}U) will be very low [10] compared to that of epithermal and fast neutrons.

Hence it is possible to maintain the high R_B-values, associated with low x-values ($x \leq 60$ cm) in non-multiplying systems, in the entire useful volume of the (Pb,C,^{233}U) system. Then for the same incident proton energy, beam intensity and k_{eff} such a system will transmute the ^{99}Tc, ^{129}I, ^{237}Np, ^{239}Pu, ^{241}Am and ^{245}Cm at much higher rates (factor of 2-10 or more) than (Pb,Pb,^{233}U) system. This discussion can be extended to the case of ^{233}U breeding from ^{232}Th (Fig.10). For this isotope we also have $R_B > 1$ for all values of $x < 60$ cm in non-multiplying systems.

For nuclear waste isotopes with energy dependent excitation function characteristics, similar to ^{246}Cm (Fig.9) where fission and absorption cross-section for slow neutrons is small and contains many high cross section resonances, the transmutation in (Pb,C, ^{233}U) will be comparable to or higher than that in the (Pb, Pb, ^{233}U), if system is designed in a way that the flux ratio in two systems maintained about the same for non-multiplying systems at low x-values ($x < 35$ cm in the case of the ^{246}Cm).

As it is shown in Sec.4.1 of this paper, for the same amount and spatial distribution of the fissile waste material in two systems, the (Pb,C,0) system has a higher k_{eff} - value. This by itself is a clear indication that, more fission is taking place in (Pb,C,0) than (Pb,Pb,0). In other words (Pb, C,0) is a better transmuter (burner) of fissile materials such as ^{239}Pu , ^{233}U and fissionable waste isotopes.

In the case of the non-fissile waste isotopes of ^{129}I and ^{99}Tc, where no fission-based neutron multiplication takes place, enhanced transmutation rates in graphite systems show clear superiority of this moderator over lead for transmutation purpose.

The transmutation rates calculated here are definitely affected by self-shielding, even to some extent in the case of the diluted samples. The "effective resonance integral" I_{eff} decreases with increasing concentration of the absorber. To maximize the I_{eff} and therefore exploit the neutrons with energies at about "resonance energy interval", one has to have the absorber in infinitely diluted form. The later provides a technical and economical challenge in design and operation of an ADS incinerator with lead moderator.

One observes enhanced neutron capture rates over the ***resonance energy interval*** (1eV to 10 keV) in activation samples, by changing the neutron-moderating environment from a light element such as carbon to a heavy metal moderator such as lead. Resonance absorption becomes possible when neutrons have well defined and discrete kinetic energies. Lead, because of its moderation characteristics provides a suitable slowing-down-environment for fast neutrons, which results in a neutron spectrum appropriate for resonance capture [10]. However one should note that such an enhancement in resonance capture is in expense of a very large reduction in the slow neutron (< 1 eV) capture rate. The

observed higher B-values, in (Pb,C,0) compared to those in (Pb,Pb,0) is the consequences of two factors:

1) The slow neutron (< 1 eV) flux in a graphite system is much higher than in a lead assembly. Most isotopes have quite high absorption and/or fission cross-sections in this energy range. The number of slow neutrons in lead moderators even with such large size used in these calculations is very small [10].
2) Although the neutron spectrum in graphite is quite different from that in lead, it contains significant number of neutrons in the energy interval of 1 eV – 10 keV [10] which can engage in resonance absorption processes, similar to the case for the lead moderators.

Lead as a moderator-coolant has quite interesting and attractive features in a fast-neutron operated ADS, designed for energy production [3,17]. However it does not seem that such a system can be an effective and efficient *nuclear waste incinerator* for the isotopes discussed in this paper.

Acknowledgements

SRH-N wishes to thank Prof. L.S. Peak and Dr. J. Ulrichs for comments and suggestions and the School of Physics, University of Sydney for support of this research project. An Institutional Grant of the Australian Research Council (ARC) supported this research program.

References

1) C.D. Bowman et al. "Nuclear energy generation and waste transmutation using an accelerator-driven intense thermal neutron source", Nulc. Instr. and Meth. A320 (1992) 336-367.
2) Ratmir G. Vassil'kov, "Free neutron production with hydrogen ion accelerators for nuclear fuel cycle needs", Muon Catalysed Fusion, 7 (1992)245-300.
3) C. Rubbia et al. "Conceptual design of a fast neutron operated high power energy amplifier", (1995), CERN/AT/95-44 (ET).
4) Charles D. Bowman, "Sustained Nuclear Energy Without Weapons or Reprocessing using Accelerator-Driven Systems", Proc. of 3rd Int. Conf. on Accelerator Driven Transmutation Technologies and Applications", June 7-11, 1999 Parha, Czech Republic, editors: M. Hron, V. Lelek, M. Mikisek, M. Sinor, J. Uher and J. Zeman.
5) Carlo Rubbia, "Resonance enhanced neutron captures for element activation and waste transmutation", (1997), CERN/LHC/97-04 (EET).
6) H. Arnould et al. "Experimental verification of neutron phenomenology in lead and transmutation by adiabatic resonance crossing in accelerator driven systems", Phys. Lett. B 458 (1999) 167-180.
7) M. Ochs et al, "Transmutation of ^{129}I and ^{237}Np using spallation neutrons produced by 1.5, 3.7 and 7.4 GeV protons", JINR preprint, E1-99-1,Dubna, Russia (2000) and Wan et al, Nucl. Instr. and Meth. A, in press.
8) J.-S. Wan et al. "First Radiochemical Studies on the Transmutation of ^{239}Pu with Spallation Neutrons", (2000), Submitted to Journal of Radioanalytical and Nuclear Chemistry.
9) S.R. Hashemi-Nezhad et al, Monte Carlo Analysis of accelerator Driven Systems; Studies on spallation neutron yield and energy gain", (2000), Kerntechnik, in press.
10) S.R. Hashemi-Nezhad et al, "Monte Carlo studies of accelerator Driven systems; energy and spatial distribution of neutrons in multiplying and non-multiplying media", (2000), submitted to Nucl. Instr. and Meth. A.
11) Handbook of Chemistry and Physics, Editor Robert C. Weast, CRC Press, (1975).
12) K.H. Beckurts and K. Wirtz, "Neutron Physics", Springer Verlag, (1964) 415-416.
13) http://hpngp01.kaeri.re.kr/CoN/endfplot.shtml.
14) Richard E. Prael and Henry Lichtenstein, "User Guide to LCS: The LAHET Code System", Los Alamos National Laboratory, report LA-UR-89-3014 (Sept. 1989).
15) Judith F. Briesmeister, editor, MCNP-4B - "A General Monte Carlo N-Particle Transport Code", report LA-12625-M (March 1997), Los Alamos National laboratory.
16) C. Rubbia et al, "Fast neutron incineration in the energy amplifier as alternative to geologic storage: The case of Spain", (1997), CERN/LHC/97-01 (EET)
17) C. Rubbia and J.A. Rubio, "A tentative programme towards a full scale energy amplifier", (1996), CERN/LHC/96-11 (EET).

Received by Publishing Department
on March 16, 2001.

Хашеми-Нежад С.Р. и др. E1-2001-44
Исследование трансмутации ядерных отходов нейтронами расщепления с использованием метода Монте-Карло. Различие свинцового и графитового замедлителей

Исследована скорость трансмутации ^{239}Pu и некоторых других актиноидов (^{237}Np, ^{241}Am, ^{245}Cm, ^{246}Cm) в двух электроядерных системах со свинцовым и графитовым замедлителями с использованием программного комплекса LAHET. Рассмотренные системы отличаются значительным объемом (~30 м^3) и не содержат делящихся материалов, за исключением небольших количеств делящихся отходов, присутствующих в некоторых случаях. Вычисления выполнены при энергии первичных протонов 1,5 ГэВ, первичная мишень — из свинца. Вычислены скорость трансмутации двух долгоживущих продуктов деления ^{99}Tc и ^{129}I в разных пространственных положениях внутри замедлителя, а также скорость накопления ^{239}Pu и ^{233}U. Показано, что электроядерные системы с графитовым замедлителем являются более эффективными для целей трансмутации, чем системы со свинцовым замедлителем.

Работа выполнена в Лаборатории высоких энергий ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2001

Hashemi-Nezhad S.R. et al. E1-2001-44
Monte Carlo Calculations on Transmutation of Transuranic Nuclear Waste Isotopes Using Spallation Neutrons. Difference of Lead and Graphite Moderators

Transmutation rates of ^{239}Pu and some minor actinides (^{237}Np, ^{241}Am, ^{245}Cm and ^{246}Cm), in two accelerator driven systems (ADS) with lead or graphite moderating environments, were calculated using the LAHET code system. The ADS that were used had a large volume (~30 m^3) and contained no fissile material, except for a small amount of fissionable waste nuclei that existed in some cases. Calculations were performed at incident proton energy of 1.5 GeV and spallation target was lead. Also breeding rates of ^{239}Pu and ^{233}U as well as the transmutation rates of two long-lived fission products ^{99}Tc and ^{129}I were calculated at different locations in the moderator. It is shown that an ADS with graphite moderator is a much more effective transmuter than that with lead moderator.

The investigation has been performed at the Laboratory of High Energies, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2001

Макет Т.Е.Попеко

Подписано в печать 11.04.2001
Формат 60 × 90/16. Офсетная печать. Уч.-изд. листов 2,07
Тираж 390. Заказ 52588. Цена 2 р. 50 к.

Издательский отдел Объединенного института ядерных исследований
Дубна Московской области