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Monte Carlo calculations on transmutation of trans-uranic nuclear waste isotopes using spallation neutrons: difference of lead and graphite moderators

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Abstract

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 ($\sim 32\,m^3$) and contained no fissile material, except for a small amount of fissionable waste nuclei that existed in some cases. Calculations were performed at an incident proton energy of 1.5 GeV and the 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.

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Keywords: Accelerator-driven systems; Spallation neutrons; Energy amplifier; Nuclear waste transmutation

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 (see e.g. [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

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advantage of ADS over conventional nuclear systems that operate *only* in critical conditions. A sub-critical ADS provides the opportunity to use the excess neutrons for other purposes such as nuclear waste transmutation as well as for 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 when spallation neutrons are moderated in lead [6]. The transmutations of the long-lived nuclear waste isotopes such as ^{99}Tc ($t_{1/2} = 2.1 \times 10^5$ yr) and ^{129}I ($t_{1/2} = 1.6 \times 10^7$ yr) have been studied by this method [6]. It has been also shown that ^{129}I, ^{237}Np ($t_{1/2} = 2.14 \times 10^6$ yr) and ^{239}Pu ($t_{1/2} = 2.44 \times 10^4$ yr) can be transmuted at quite acceptable rates in thermal (slow) neutron dominated neutron fields [7,8].

In earlier papers [9,10] we have reported the spallation neutron yield, energy spectrum and spatial distribution of the neutrons and energy gain in several sub-critical ADSs. In the present paper we present the results of our calculations on the transmutation of some trans-uranic nuclear waste isotopes and two long-lived fission products (^{99}Tc and ^{129}I) in two ADS with lead and graphite moderators. The breeding rates of the ^{239}Pu from ^{238}U and ^{233}U from ^{232}Th were also calculated. The trans-uranic waste isotopes that were studied are ^{237}Np ($t_{1/2} = 2.14 \times 10^6$ yr), ^{239}Pu ($t_{1/2} = 2.44 \times 10^4$ yr), ^{241}Am ($t_{1/2} = 458$ yr), ^{245}Cm ($t_{1/2} = 9.3 \times 10^3$ yr) and ^{246}Cm ($t_{1/2} = 5.5 \times 10^3$ yr).

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 shorter than that of the original waste isotope itself. 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, $\sigma_f = 19$ mb), but (n,γ) and other nuclear processes on the waste isotopes may transfer them to nuclei that have much higher fission cross-section. Fig. 1 shows some incineration channels for trans-uranic isotopes starting with ^{241}Am.

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,xn) reaction is 2–3 orders of magnitude less than those of fission and (n,γ) reactions. Therefore, although α-decay (Fig. 1) and (n,xn) 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,xn) reaction [11]. Therefore, 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 [12] along with MCNP-4B2 code [13] to calculate the spallation neutron induced transmutation yield of the long-lived radioactive waste isotopes.

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 the system does not contain a specific nuclear fuel. Fig. 2 shows XZ-cross-section of all the components of the target-moderator assemblies used in the calculations. The origin of the Cartesian coordinate system is at the centre of the assembly.

The moderator occupied a volume of $3.3 \times 3.3 \times 3\,\mathrm{m}^3$ ($\sim 32\,\mathrm{m}^3$) and the target was embedded in the moderator. In the case of the (Pb,Pb,0) the whole ~ 370 tons of lead acts as target-moderator. While in the (Pb,C,0) a cylindrical lead target of diameter 20 cm and length 1 m was placed on the Z-axis, starting from $Z = -30$ cm (Fig. 2). This target length is sufficient to produce maximum number of spallation neutrons

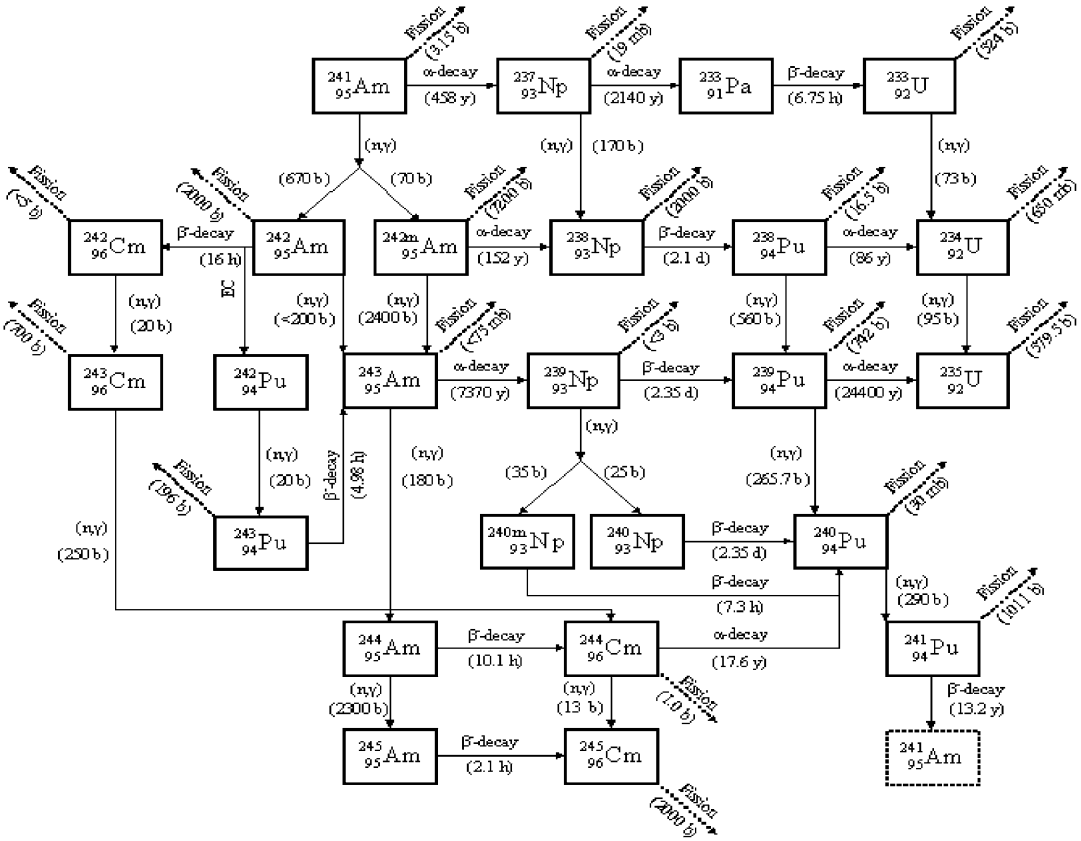

Fig. 1. Transmutation and incineration of some trans-uranic waste isotopes. The cross-section values are for thermal neutrons.

as long as the target length is concerned [9]. Twenty 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. 2). These cells were either "filled" with the nuclear waste isotope of interest or with the relevant moderator material when "not used".

The beam of protons of energy 1.5 GeV (unless otherwise stated) 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. For details of the code set-up and input file options for LAHET calculations refer to Refs. [9–11].

For every calculation, 25,000 incident proton histories ($>10^6$ neutron histories) were followed. The cross-section libraries used in calculations were those provided with MCNP-4B2 [13] for neutron transport and dosimetry calculations. The results of the transmutation calculations are given in terms of B, which is defined as the number of a given reaction in 1 g of the isotope of interest per incident proton. 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 and results in very small errors in our calculations because: (1) the flux of neutrons above 20 MeV is very small, (2) neutron capture and fission cross-sections for isotopes of interest at energies greater than 20 MeV are very low.

4. Results and discussion

Table 1 gives the characteristics of graphite and lead moderators. As it can be seen these

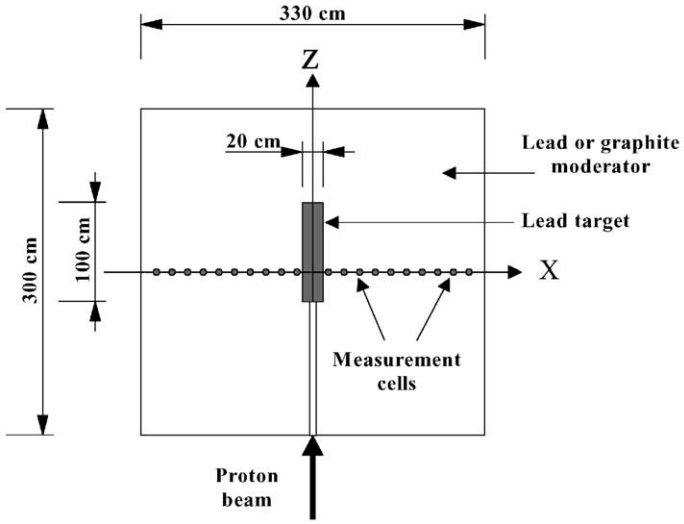

Fig. 2. *XZ*-cross-section of the target-moderator assemblies used in the LAHET Monte Carlo simulations. The origin of the Cartesian co-ordinate system corresponds to the centre of the assembly. (For details of the set-ups refer to the text.)

Table 1
Some properties of graphite and lead moderators, Σ_s and Σ_a are macroscopic scattering and absorption cross-sections, respectively

Moderator	Average logarithmic energy decrement, ξ	Slowing down power $\xi\Sigma_s$	Moderation ratio $\xi\Sigma_s/\Sigma_a$	N_1[a]	N_2[b]	Moderation time of fission neutrons to 1 eV (μs)
Graphite	0.158	0.061	170	151	298	23
Lead	0.00962	0.0035	0.623	2632	5199	400

[a] Number of neutron collisions required for crossing energy interval of 10 keV–1 eV.
[b] Number of neutron collisions required for crossing energy interval of 2 MeV–0.025 eV.

Table 2
Spallation neutron yield and neutron leakage from the systems per incident proton of $E_p = 1.5$ 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

two materials have quite different moderating properties.

Table 2 gives the spallation neutron yield per incident proton of 1.5 GeV, Y_n, and the total number of neutrons that leak out of the systems, Y_l. Although the neutron yield in lead moderating environment is 22% higher than that in the graphite system because of higher neutron leakage from the lead system, the 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.

4.1. *Transmutation of* ^{239}Pu *and* ^{238}U

If the ADS contains ^{238}U in any form, then ^{238}U(n, γ) reactions will result in the 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 $x \geqslant 15$ cm were filled with ^{239}Pu while the cells

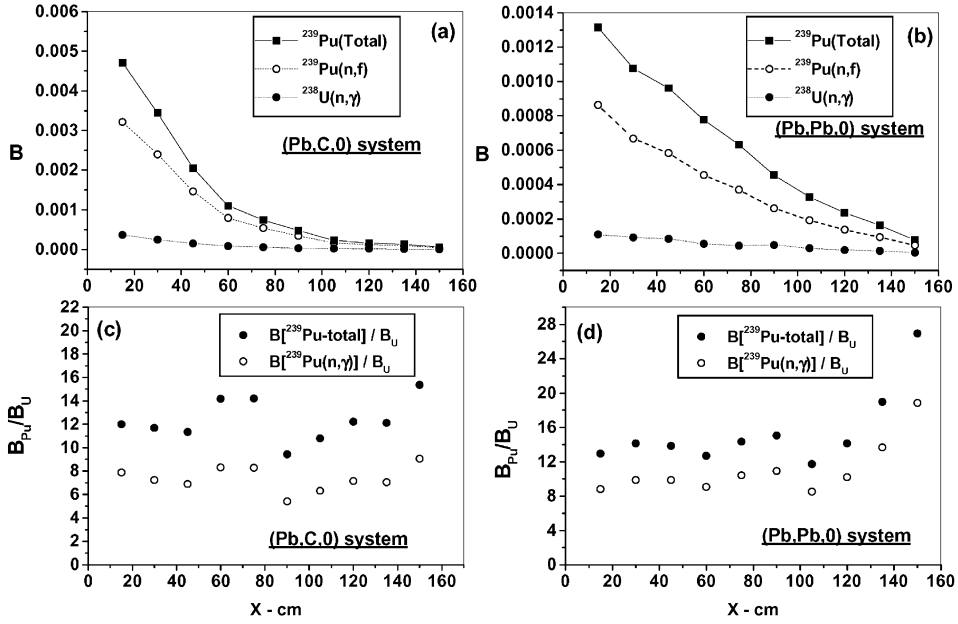

Fig. 3. (a) and (b) transmutation of ^{239}Pu by (n, f) and (n, γ) and ^{238}U via (n, γ) reactions in (Pb,Pb,0) and (Pb,C,0) at a proton energy of 1.5 GeV. (c) and (d) variation of ratio of the B-values of plutonium to uranium, with position on the X-axis. In the legends of (c) and (d) B[^{239}Pu-total] refer to B-^{239}Pu(n, f) + B-^{239}Pu(n, γ) and B_U represents ^{238}U(n, γ).

with $x \leqslant -15$ 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 $(k_{\text{eff}})_{\text{Pb}} = 0.14444 \pm 0.00059$ and $(k_{\text{eff}})_{\text{C}} = 0.17821 \pm 0.00176$ were calculated for (Pb,Pb,0) and (Pb,C,0) systems, respectively, using the MCNP-4B2 code.

In the case of ^{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. 3 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. Also shown in the Fig. 3 is the variation of ratio of the B-values of plutonium to uranium, at different locations on the X-axis. 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. $R_1 = \langle (B_{\text{Pu}})_{(\text{n,f})}/B_{\text{U}} \rangle = 7.4$ (Fig. 3c). This ratio increases to $R_2 = 12.3$ if one includes the ^{239}Pu(n, γ) reactions in the calculations as well (Fig. 3c). For (Pb,C,0) system we obtain $R_1 = 11$ and $R_2 = 15.5$ (Fig. 3d), suggesting that the destruction to production ratio in graphite system is higher than that in the lead.

To compare the transmutation capabilities of the two ADS at different x-values the ratio of B-value in (Pb,C,0), B_{C}, to that in (Pb,Pb,0), B_{Pb}, was calculated ($R_B \equiv B_{\text{C}}/B_{\text{Pb}}$). Fig. 4 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 $x \leqslant 90$ cm (see also Ref. [11]).

From these results we conclude that regardless of the presence or absence of ^{238}U, (Pb,C,0) transmutes ^{239}Pu in a significantly higher rate than (Pb,Pb,0). This is also evident from the higher k_{eff} of (Pb,C,0) as compared to that of (Pb,Pb,0).

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. 2). The second calculation involved the Cm isotopes.

Fig. 5 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. The (n,γ) reactions of these isotopes result in isotopes with high fission cross-sections (Fig. 1).

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. 5c and d R_B as a function of distance is plotted both for fission and total B-values. From Fig. 5 we note the following:

(1) For $x\leqslant 60$ cm the (Pb,C,0) transmutes ^{241}Am at a higher rate than the (Pb,Pb,0). At distances $x>60$ cm, (Pb,Pb,0) is a more effective transmuter for this isotope, Fig. 5a.
(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 $15\leqslant x\leqslant 150$ cm, Fig. 5b.

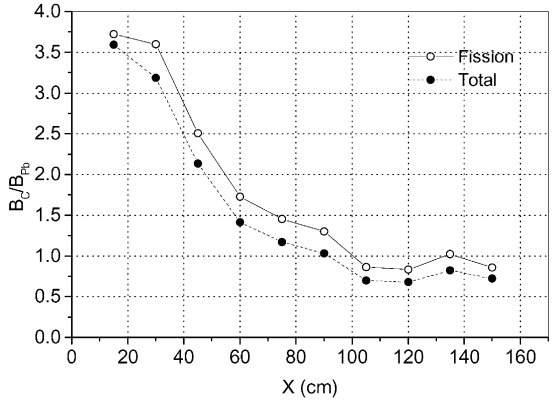

Fig. 4. Variation of the ratio of the transmutation rates of ^{239}Pu in (Pb,C,0) and (Pb,Pb,0) systems ($R_B \equiv B_C/B_{Pb}$), as a function of distance x, on the X-axis (Fig. 1). The term "total" in the legend refers to $(n,\gamma)+(n,f)$ reactions.

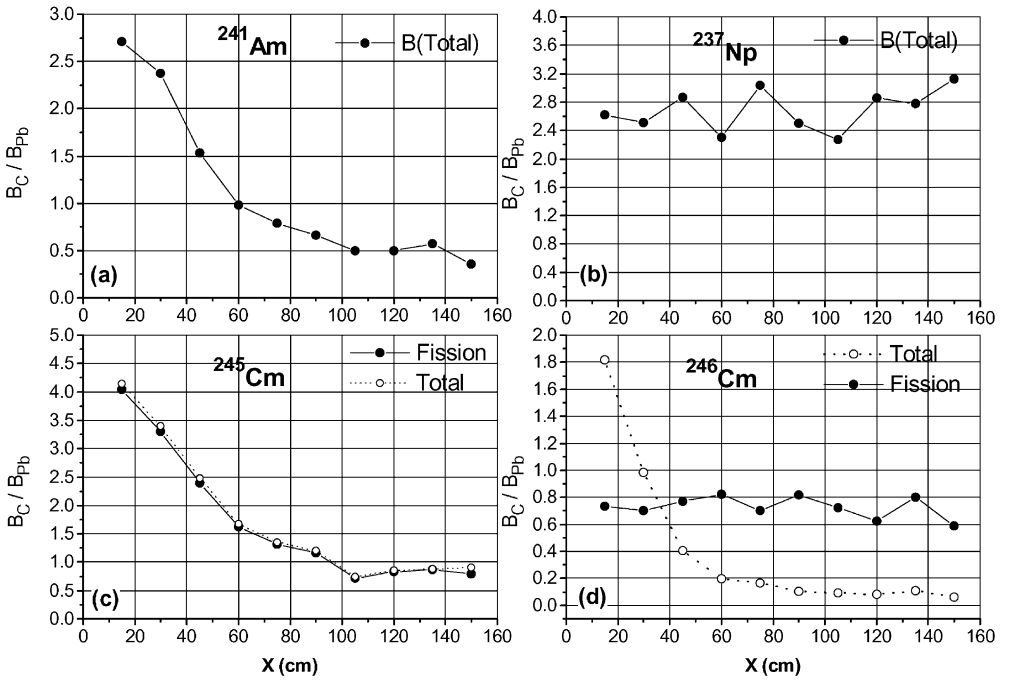

Fig. 5. Variation of the ratio of the transmutation rates of ^{241}Am, ^{237}Np, ^{245}Cm and ^{246}Cm in (Pb,C,0) and (Pb,Pb,0) systems ($R_B \equiv B_C/B_{Pb}$), as a function of distance x, on the X-axis (Fig. 1). The term "total" in the legend refers to $(n,\gamma)+(n,\text{fission})$ reactions.

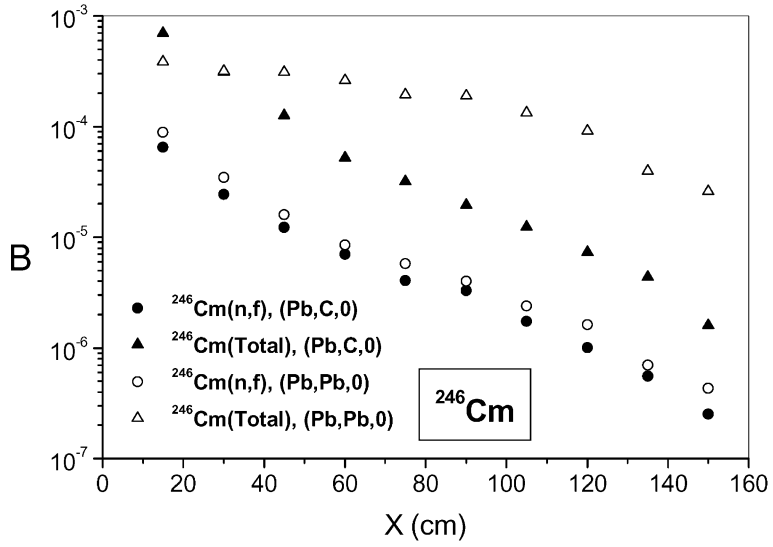

Fig. 6. Variation of the B-values of ^{246}Cm with distance on the X-axis in (Pb,Pb,0) and (Pb,C,0) assemblies for incident protons of energy 1.5 GeV. The term "total" in the legend refers to $(n,\gamma)+(n,\text{fission})$ reactions.

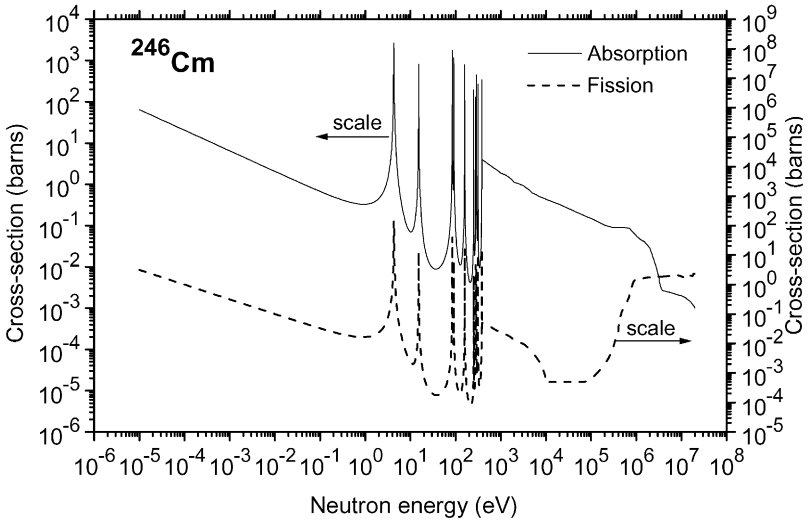

Fig. 7. Energy-dependent absorption and fission cross-section of ^{246}Cm. Data from ENDF/B-VI library.

(3) For $x \leqslant 90$ cm the (Pb,C,0) transmutes ^{245}Cm at higher rate than the (Pb,Pb,0). At distances $x > 90$ cm, the (Pb,Pb,0) is a better transmuter for this isotope, Fig. 5c.
(4) In the case of ^{246}Cm the situation is different. Transmutation by fission is higher when the moderator is lead in the entire volume of the systems studied. The total rate (fission + (n,γ)) is higher in graphite moderator only for $x < 35$ cm, Fig. 5d.

Fig. 6 shows the variation of B with x, for ^{246}Cm in the two systems studied. Fig. 7 illustrates the energy-dependent fission and absorption cross-

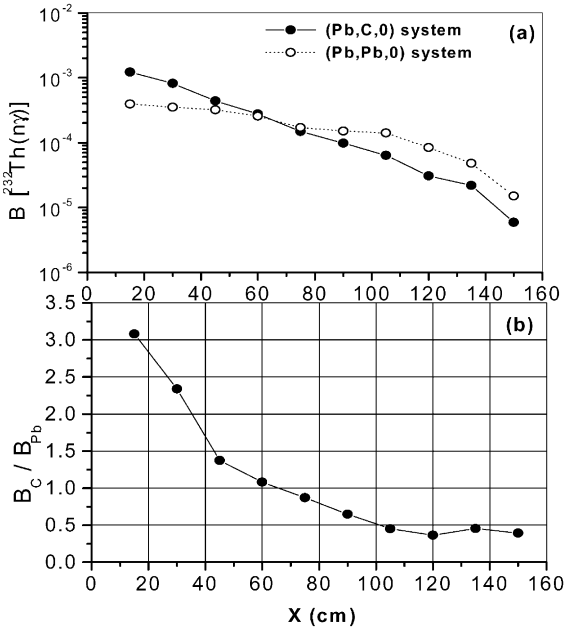

Fig. 8. Transmutation of ^{232}Th (^{233}U breeding) as a function of distance in (Pb,Pb,0) and (Pb,C,0) systems.

section of ^{246}Cm (data from ENDF/B-VI library). The results presented in Fig. 6 can be understood by examination of Fig. 7 and neutron flux and energy spectra in graphite and lead systems [10]. The slow neutron cross-sections for this isotope is relatively low and its excitation function contains several high-cross-section resonances.

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

In a conventional nuclear reactor the Pu and minor actinides (PMA) are produced by a chain of (n,γ) reactions and radioactive decays, starting with ^{238}U, 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 the (n,γ) reaction.

Fig. 8a shows the transmutation rate for ^{232}Th(n,γ) reaction (^{233}U production rate) as a function of distance x, in (Pb,Pb,0) and (Pb,C,0). In Fig. 8b, the ratio of the transmutation rates in graphite and lead systems as a function of x is shown. For $x \leqslant 60$ cm ^{233}U breeding in (Pb,C,0) is higher than that in (Pb,Pb,0). On average the 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 $x \leqslant 60$ cm, the average of R increases to 1.97.

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

Fig. 9a and b show the variations of B for ^{99}Tc(n,γ) and ^{129}I(n,γ) as a function of distance from the centre of the assemblies on the X-axis for (Pb,C,0) and (Pb, Pb,0) systems. Fig. 9c illustrates R_B as a function of x for ^{99}Tc and ^{129}I. From Fig. 9, we conclude that the 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.

1. For ^{129}I the B-values in (Pb,C,0) are higher than their corresponding values in (Pb,Pb,0) for all sample locations (by a factor of more than 14 for $x \leqslant 30$ cm).
2. In (Pb,C,0) system 65.7% of ^{99}Tc and 92.8% of ^{129}I transmutations were caused by neutrons of $E_n \leqslant 1$ eV.
3. In (Pb,Pb,0) system 96.4% of ^{99}Tc and 87.7% of ^{129}I transmutations were initiated by neutrons of $E_n > 1$ eV.

The B-values for ^{129}I and ^{99}Tc in a geometrical set-up and location, identical to that in Ref. [6] (i.e. at a distance of 45 cm from the beam axis and $Z = 7.5$ cm) were also calculated for protons of momentum 3.57 GeV/c. The samples were placed in void measurement tubes as was the case in Ref. [6]. We obtained a B-value of $(3.87 \pm 0.27) \times 10^{-4}$ for ^{129}I which is equivalent of $(2.50 \pm 0.18) \times 10^{-5}$ captures per proton of 3.57 GeV/c in an iodine sample of mass 64.7 mg, in excellent agreement with the experimental result of $(2.61 \pm 0.26) \times 10^{-5}$ [6]. In the case of the ^{99}Tc, B-values close to the experimental results were obtained only for thick and dense samples of ^{99}Tc. For spherical samples of radius 0.62 cm (1 cm^3) filled with metallic ^{99}Tc of average density of 0.216 and 11.5 g cm^{-3} B-values of $(3.13 \pm 0.25) \times 10^{-3}$ and $(1.40 \pm 0.10) \times 10^{-3}$, respectively, were obtained. The B-values reported in Ref. [6] are 2.34×10^{-3},

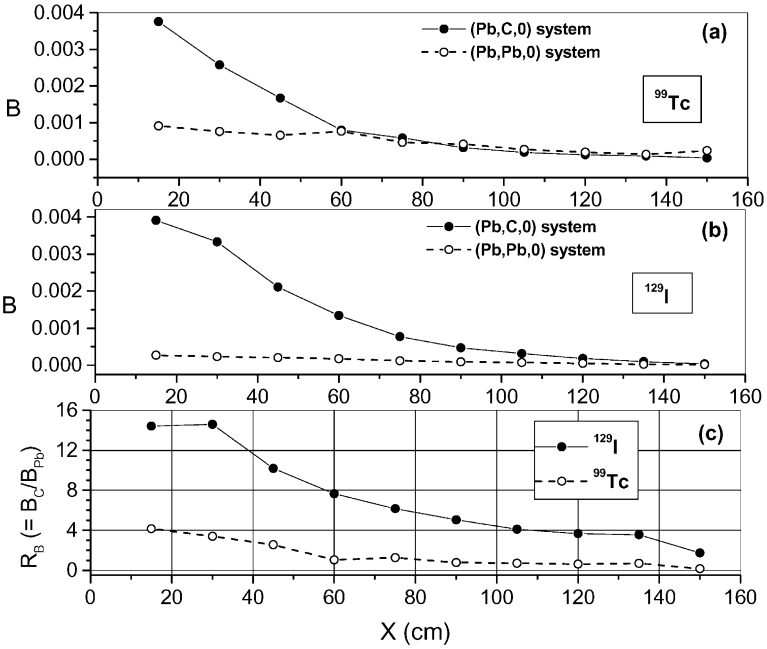

Fig. 9. Transmutation of ^{99}Tc and ^{129}I in (Pb,Pb,0) and (Pb,C,0) systems.

1.52×10^{-3} and 1.24×10^{-3} for ^{99}Tc samples of mass 14.3, 100.9 and 216.1 mg, respectively.

5. Effects of self-shielding

The transmutation rates reported in this paper so far are definitely affected by self-shielding. To reduce the 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 modelling. The resonance integral I_{Res} for ^{99}Tc and ^{129}I are 300 and 50 b, respectively [14]. 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 the amount of ^{99}Tc and ^{129}I in calculation cells, we modified their content from pure waste isotope to a mixture of waste isotope of interest and the relevant moderator. We chose a partial waste density of $0.2\,g\,cm^{-3}$ and 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. The obtained transmutation rates showed the following:

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 is quite similar to the case when the samples were much denser (Fig. 9c).

From these results we conclude 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.

The observed self-shielding effect is the result of local flux suppressions because of thermal and resonance absorption and scattering, mainly in the outer regions of the cells. Obviously, the thermal

effect will be high in graphite and resonance effect will be high in the lead moderator.

6. Further discussions and conclusions

The results reported in the present paper apply to non-multiplying accelerator-driven systems [10] and to the cases of lead and graphite neutron moderating environments. In these assemblies spallation neutrons are generated at about 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).

The neutron flux attenuates exponentially with increasing distance from the centre of the assemblies [10] and different attenuation lengths are associated with different moderators. It is shown [10] that in multiplying sub-critical ADS the neutron flux can be very high, depending on the k_{eff} of the system and intensity and energy of incident protons [10]. Addition of fissile materials into the system affects the spatial distribution of the neutrons much more than their energy spectrum [10].

In a *non-multiplying* ADS the TARC in (Pb,Pb,0) overtakes the transmutation in (Pb,C,0) only when the neutron flux in (Pb,C,0) 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) is 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 $k_{\text{eff}} = 0.95$) 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 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 multiplication is achieved by introduction of ^{233}U fissile fuel in the systems (in the 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 ($x \leqslant 60$ 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}, the (Pb,C,^{233}U) system will transmute ^{99}Tc, ^{129}I, ^{237}Np, ^{239}Pu, ^{241}Am, ^{245}Cm and ^{232}Th at much higher rates (factors of 2–10 or more) than the (Pb,Pb,^{233}U) system.

For nuclear waste isotopes with energy dependent excitation function characteristics, similar to ^{246}Cm (Fig. 7) 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 the system is designed in such a way that the flux ratio in the two systems was maintained about the same for non-multiplying cases at low x-values ($x < 35$ cm in the case of the ^{246}Cm).

The "effective resonance integral", I_{eff} decreases with increasing concentration of the absorber within the moderator. To maximize the I_{eff} and hence exploit the neutrons with energies at about "resonance energy interval", one has to have the absorber in infinitely diluted form in a homogenous ADS structure. The later provides a technical and economical challenge in design and operation of an ADS incinerator with lead moderator. In a heterogeneous system the resonance escape probability increases, due to resonance self-shielding effect in the waste lump. Resonance absorption in the outer layers of the waste lump produce local flux depressions around resonances in the neutron spectrum. Consequently, the majority of the waste isotopes are

exposed to a depressed resonance spectrum, resulting in a reduced resonance capture rate. On the other hand for transmutation in a slow neutron dominated system such as (Pb,C,0), increased resonance escape probability is an advantage rather than disadvantage.

One observes enhanced neutron capture rates over the *resonance energy interval* (1 eV–10 keV), by changing the neutron-moderating environment from a light element such as carbon to a heavy metal moderator such as lead. However, one should note that such an enhancement in resonance capture is at the 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) are the consequences of the following 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] that can engage in resonance absorption 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 [15,16]. However, it does not seem that such a system can be an efficient *nuclear waste incinerator* for the isotopes discussed in this paper.

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