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MONTE CARLO STUDIES OF ACCELERATOR DRIVEN SYSTEMS. ENERGY AND SPATIAL DISTRIBUTION OF NEUTRONS IN MULTIPLYING AND NON-MULTIPLYING MEDIA

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

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

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

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

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

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

1. Introduction

Neutrons produced in the interaction of relativistic projectiles with heavy targets (e.g. protons on lead) can be used for energy production and nuclear waste transmutation, in Sub-critical Nuclear Assemblies (SNA), [1-3]. These systems are also known as Accelerator Driven systems (ADS).

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. This method is known as Transmutation by Adiabatic Resonance Crossing (TARC), when a Pb moderating environment is used [4], otherwise it is the standard resonance capture process well known to neutron physicists. The transmutations of the long-lived nuclear waste isotopes such as ^{99}Tc and ^{129}I have been studied by this method [5]. It is also shown that ^{129}I and ^{237}Np can be transmuted at acceptable rates in thermal (slow) neutron dominated neutron fields [6]. The experimental results obtained by means of TARC are in good agreement with Monte Carlo calculation results based on FLUKA [7] and associated codes [see ref. 5 and references therein].

As the effectiveness of TARC is highly dependent on the fluence and energy distribution of the neutrons at the waste sample location, it is important to investigate how neutron energy distribution and fluence varies with distance from the centre of the cascade.

In a realistic ADS, there will be fissile materials in the system [2,8,9]. Such fissile materials are present either as fuel *only* (eg ^{233}U, ^{235}U) or as combined fuel-waste, such as Pu isotopes and other actinides such as Am and Cm. The presence of such nuclei within the system enhance the neutron fluence and also can alter the energy distribution of neutrons in the system, because neutrons with a fission energy spectrum are additional to those resulting from spallation processes. Therefore we studied ADSs that contained a fissile material in the system.

To compare systems using a light element as moderator with those using a lead moderator, we studied the systems with graphite as moderator. Finally the transmutation rates of the ^{129}I and ^{99}Tc were studied in assemblies with graphite and lead moderators.

2. Calculation procedure

The behaviour of the spallation neutrons produced in the interaction of relativistic protons with thick lead target embedded within a large moderating environment was studied. The spatial distribution of the neutron fluence and the neutron energy distribution in large-scale moderator systems (~ 30 m^3) with and without the presence of a neutron-multiplying medium, was calculated using the LAHET code system [10]. Here the neutron-multiplying region refers to the one that contains fissile materials. In the present paper three different cases that will be considered are (see Fig.1);

(a) Lead target and lead moderator without presence of neutron multiplying medium, (Pb, Pb,0) system.

(b) Lead target, lead moderator plus a ^{235}U neutron multiplying region, (Pb, Pb,^{235}U) system.
(c) Lead target and graphite moderator without a neutron multiplying region, (Pb,C,0) system.

Fig.1 shows the XZ-cross section of the all components of the all target-moderator assemblies used in the calculations. The origin of the Cartesian co-ordinate system is at the centre of the assembly.

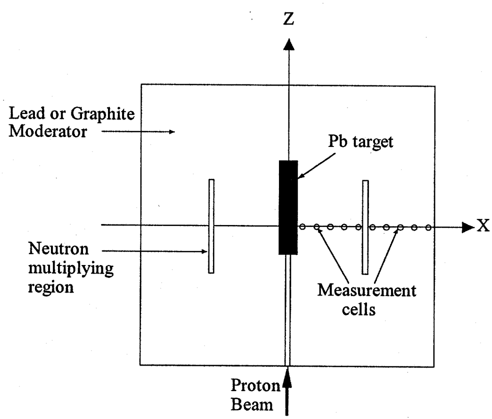

Figure 1. XZ-cross-section of the target-moderator-multiplier assemblies used in the LAHET Monte Carlo simulations. The origin of the Cartesian co-ordinate system corresponds to the centre of the assembly. Note that not all of the components shown in figure were present in every simulation. The three cases that were studied are; (a) lead target and moderator without a neutron multiplying region (b) lead target and moderator and a neutron multiplier region and (c) lead target and graphite moderator without a neutron multiplying region. (For dimensions and details of the set-ups refer to the text.)

In all of the above-mentioned cases the moderator occupied a volume of 3.3×3.3×3 m^3. The target and multiplying region (if present) were embedded in the moderator as shown in Fig.1. In the case of the (Pb, Pb,0) and (Pb, Pb,^{235}U) systems 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. 1).

To model the effect of a neutron multiplying medium on the neutron fluence and energy distribution of the neutrons, a cylindrical stainless steel shell container, filled with UO_2 as fuel, was placed within the lead assembly. The axis of fuel container and beam axis are concentric and its centre coincide with the centre of the assembly (Fig. 1). The wall thickness of the container was 2 mm and it had inner and outer diameters of 79.8 and 85.2 cm respectively. The length of the fuel container was 100 cm. It was assumed that 20% of the U-atoms in UO_2 are ^{235}U. In the design of the target-moderator-multiplier systems shown in Fig. 1, technical considerations are not taken into account. In particular, the chemical composition of the fissile material in the multiplying region is chosen arbitrarily. Obviously this is a very simplified form of an ADS but nevertheless it can provide useful information on the behaviour of the neutrons in ADSs that use a large amount of lead or graphite for the moderation and storage of the neutrons.

In all target-moderator assemblies ten spheres of radius 3 cm filled with the corresponding moderator medium were located on the x-axis starting from x = 15 cm at intervals of 15 cm which were used as measurement (calculation) cells (Fig. 1). In the case of the (Pb, Pb,0) system seven extra spherical cells of diameter 1 cm were located at x < 15 cm, with intervals of 2 cm, starting from x = 0 cm (these cells are not shown in Fig.1).

In most of the calculations protons of momentum 2.5 GeV/c (kinetic energy E_p=1732 MeV) 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 [10], which is coupled with the MCNP code [11] deals with spallation processes involving hadronic interactions and transport of the produced neutrons (in our case) down to 20 MeV. Neutrons of energy less than 20 MeV are transported with MCNP code, which in our case was version MCNP-4B2. In all calculation involving MCNP 4B2 the continuous energy mode was used.

In LAHET calculations we used the following options in the input file [ref. 10 and refs. therein];

- The Bertini model of intranuclear cascade
- A Pre-equilibrium model following the intranuclear cascade
- The Gilbert-Cameron-Cook-Ignatyuk level density model
- Coulomb barriers on incident charged particle interactions
- The 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. 10 and references therein] as well as those produced in internuclear- cascade.

For every calculation 25,000-50,000 incident proton histories were followed and in most cases the results were normalized to 10^9 incident protons (beam current of I = 0.16 nA).

3. Results and discussion

3.1 Energy spectrum of spallation nucleons

Figure 2 shows the energy distribution of the nucleons resulting from the spallation process when protons of energy 1.73 GeV impinge on Pb-targets of (Pb,Pb,0) and (Pb,C,0) systems. The difference between the spallation neutron energy distributions in the two systems may be summarised as follows;

- The energy distribution of the spallation neutrons in two target-moderator assemblies of (Pb,C,0) and (Pb,Pb,0) are rather similar.
- The peaks of the neutron distributions appear at energy interval of 2.7-3.7 MeV in both systems.
- The energy distribution of the spallation protons in the two systems are quite different and the distribution in (Pb,C,0) extends to lower energy than (Pb,Pb,0) system.
- The peak of the proton distributions appear at energies of 8.5 MeV and 10 MeV for (Pb,C,0) and (Pb,Pb,0) systems respectively.

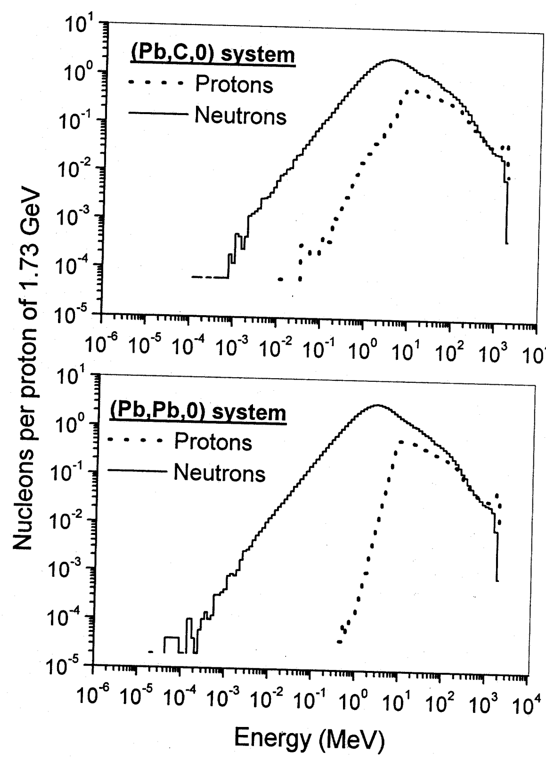

Figure 2. Energy distribution of the nucleons resulting from the spallation process when protons of energy 1.73 GeV impinge on Pb-targets of (Pb,Pb,0) and (Pb,C,0) systems.

Table 1 gives the total number of spallation neutrons produced in each of the accelerator driven systems studied, Y_n. Also given is the number of neutrons that leak out of each system per incident proton of E_p = 1.73 GeV. the last column of the Table 1 gives the fraction of the total number of neutrons that leak out of the system, Y_l. In the case of the (Pb, Pb, ^{235}U) system, leakage neutrons include the neutrons that are produced in the neutron-multiplying region via fission. The cylindrical neutron multiplying shell in (Pb, Pb, ^{235}U) contains 2.77 tons of UO_2 from which 488.3 kg is ^{235}U. For this system an effective neutron multiplication coefficient of k_{eff} = 0.8891 ± 0.0005 was calculated using MCNP-4B2 code [11]. In (Pb, Pb, ^{235}U) the Y_n = 66.11 spallation neutrons multiply to N_t = 596 neutrons. Neutron leakage from the (Pb, Pb, ^{235}U) system is ~50% of that from the (Pb, Pb,0) system. This is due to the fact that in (Pb, Pb, ^{235}U) the majority of the neutrons have a fission origin and so have different energy distribution compared to that of the spallation neutrons. The difference $Y_a = N_t - Y_l$ is the number of the neutrons per incident proton, that are absorbed in each system (for non-multiplying systems $N_t = Y_n$).

Table 1. Spallation neutron yield and neutron leakage from the system per incident proton of E_p = 1.73 GeV.

Type of ADS	Spallation neutron yield Y_n	Neutron leakage from system	
		Leakage Y_l	Percentage of total neutrons in system
(Pb, Pb,0)	66.23 ± 0.07	21.71 ± 0.03	32.8 ± 0.06
(Pb, C,0)	52.38 ± 0.16	7.20 ± 0.02	13.7 ± 0.06
(Pb, Pb,^{235}U)	66.11 ± 0.07	90.29 ± 0.69	15.0 ± 1.1

3.1 Energy spectrum of neutrons in the moderator

3.2.1 (Pb, Pb,0) system

Figure 3 shows the neutron fluence (E×dF/dE) as a function of energy for the (Pb,Pb,0) system, at different positions on the X-axis. From Fig.3 it can be seen that;

- In a large lead moderator and in the absence of a neutron-multiplying medium, even in the vicinity of the beam line the neutron fluence at given x-value does not vary substantially with increasing energy in the energy range of 1 eV to 1 MeV. Within this energy range there is a factor of <70 increase in the neutron fluence for a 6 orders of magnitude increase in the energy. For neutrons in the energy interval of 1 eV - 1 keV this factor is only ~ 4
- The maximum of the energy distributions appears at E ≈ 520 keV independent of the location in the moderator. Due to slowing down of neutrons in the system, the width of this peak increases with increasing x and becomes less pronounced, to the extent that neutron fluence becomes almost constant in the energy interval of 1 eV to 100 keV. The energy distribution of the neutrons that leak out of the system quite clearly shows the existence of such a peak in the above-mentioned energy.

- Slow neutrons ($E_n \leq 1$ eV) make a very small fraction of the total neutron fluence at a given location, suggesting that accelerator driven system of the form (Pb, Pb,0) cannot be very efficient for nuclear waste transmutation via thermal neutron capture process.
- It is quite interesting to note that the variation of the neutron fluence with energy at x = 0 (i.e a location within the target and on the beam line) and x =15 cm in the energy interval 1 eV to ~ 100 keV are almost the same.

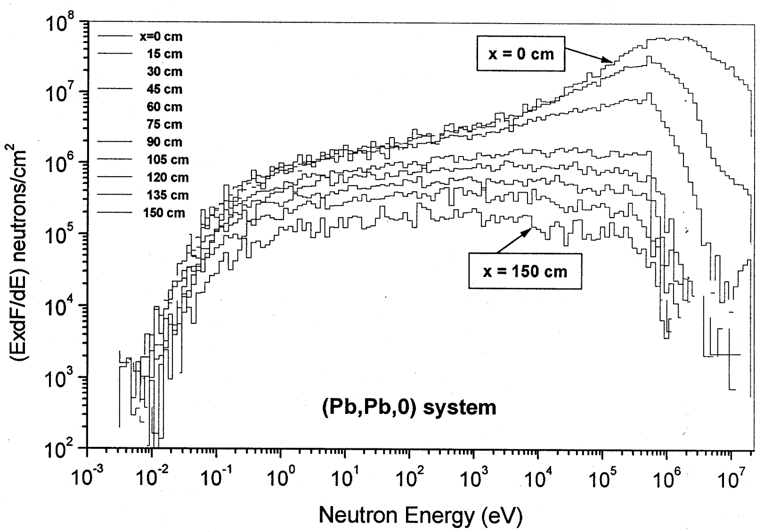

Figure 3. Neutron fluence E×dF/dE as a function of energy for the case of (Pb,Pb,0); lead target and moderator in absence of a neutron multiplier region (see the text for details).

3.2.2 (Pb,Pb,^{235}U) system

Figure 4 illustrates the neutron fluence (E×dF/dE) as a function of energy for (Pb,Pb,^{235}U) system. For clarity of the figure only energy distribution in three cells at x = 15, 60 and 120 cm are shown.

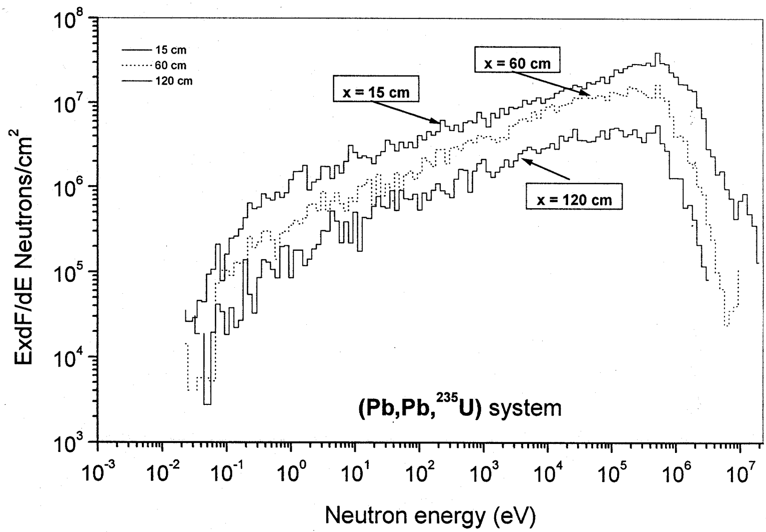

Figure 4. Neutron fluence ExdF/dE as a function of energy for the case of lead target-moderator in the presence of a neutron multiplier region (Pb,Pb,^{235}U) system. For clarity only the energy distribution in three locations are shown (see the text for details).

A comparison of the Figs 3 and 4 shows that, qualitatively, the presence of a multiplying medium and addition of fission neutrons to the system has not altered the overall shape of the energy distributions of the neutrons significantly, in the vicinity of the centre of the cascade. However in the regions close to the multiplying region e.g. x = 75-90 cm and also in areas far away from this region e.g. x ≥ 120 cm the energy distributions of the neutrons are significantly different from those in the (Pb,Pb,0) system at the corresponding locations. For example at x=120 cm for (Pb,Pb,0) system the neutron fluence in the energy range of 1 eV to 100 keV remains *almost* constant, while for the case of (Pb,Pb,^{235}U) there is more than an order of magnitude fluence increase for the same energy interval. In spite of the addition of fission neutrons to spallation neutrons, the peak of the energy distribution appears at ~ 520 keV, the same as for the (Pb, Pb,0) system.

3.2.3 (Pb,C,0) system

Figure 5 illustrates the variation of the neutron fluence as a function of energy for (Pb,C,0) system in the neighbourhood of the target, at three different locations on the x-axis. Due to very rapid thermalisation of the neutrons in graphite moderator, the fluence of the high energy neutrons drops dramatically and statistical error in high energy bins becomes increasingly larger with increasing distance from the centre of the assembly. The shape of the energy distributions for (Pb,C,0) are very different compared to the case for lead moderator. As expected the slow neutrons make quite a large fraction of the total neutron fluence F(x), in a given location in the moderator. Fig. 6 shows the variation of the R = $F(x)_{(E_n \leq 1eV)} / F(x)_{(E_n \leq 20\ MeV)}$, the ratio of the slow neutron fluence ($E_n \leq 1$ eV) to total neutron fluence ($E_n \leq 20$ MeV), as a function of distance x, for the (Pb, C,0) and (Pb, Pb,0) systems. For lead moderator, at most 8.4% of the total neutrons are slow ($E_n \leq 1$ eV). This occurs at cell 10 (x = 150 cm) where the overall neutron fluence is quite low. In the case of the graphite moderator for x ≥ 60 cm almost all the neutrons are slow. For (Pb,C,0) system at higher energies (1 eV to 1 MeV) the variation of the neutron fluence with energy is not substantial (Fig.5). This is especially true for x < 50 cm.

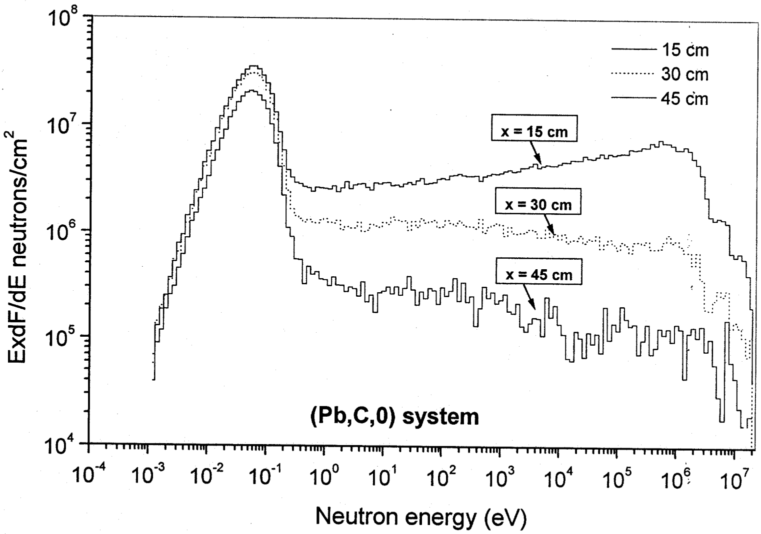

Figure 5. Neutron fluence E×dF/dE as a function of energy for the case of lead target and graphite moderator in absence of a neutron multiplier region (Pb,C,0) system (see the text for details). For clarity the energy distribution in only three locations are shown.

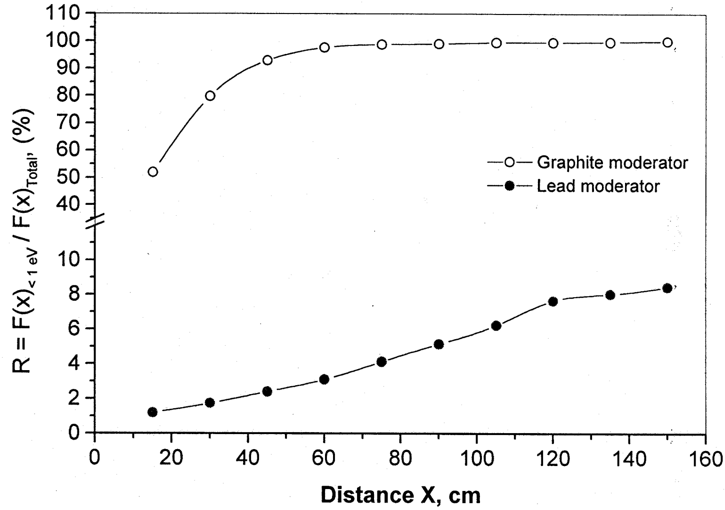

Figure 6. Variation of the $R = F(x)_{(E_n \leq 1eV)} / F(x))_{(E_n \leq 20 MeV)}$, ratio of slow neutron fluence ($E_n \leq 1$ eV) to total neutron fluence ($E_n \leq 20$ MeV) as a function of distance x, for the (Pb, C,0) and (Pb, Pb,0) systems.

4. Spatial distribution of neutrons

Fig. 7 shows the variation of the fluence F(x), for neutrons of $E_n \leq 20$ MeV with the distance on the x-axis ($x \geq 15$ cm), for all three target-moderator systems studied. For target-moderator assemblies in the *absence* of a multiplying medium, neutron fluence F(x) as a function of distance from the centre x, can be described very well with an exponential attenuation curve of the form;

$$F(x) = y_0 + Ae^{-(x-x_0)/t} \qquad x \geq 15cm \qquad (1)$$

The values of the fitting parameters are given in Table 2. The "attenuation length" t, of the neutrons in two systems of the (Pb, Pb,0) and (Pb, C,0) are different and neutron number in the (Pb, C,0) system attenuate faster. For these two cases (without multiplying media) from Eq.1 a half-thickness ($L_{1/2}$) can be calculated. Where $L_{1/2}$ is defined as the increase in moderator thickness x, that results in reduction of the overall neutron fluence by a factor of two. For neutrons of $E_n \leq 20$ MeV, half-thickness values are 25 cm and 18.5 cm for (Pb,Pb,0) and (Pb,C,0) systems respectively.

Table 2. Fitting parameters of Eq.1 for the variation of the neutron fluence F(x) as a function of x for neutrons of En ≤ 20 MeV. Parameters x_0, t and $L_{1/2}$ are in units of cm.

(Pb,Pb,0)*	(Pb,C,0)**
$y_0 = 0 \pm 0$ $x_0 = 15 \pm 0$ $A = (731.327 \pm 7.578)\times10^6$ $t = 36.086 \pm 0.647$ $L_{1/2} = 25.013 \pm 0.448$	$y_0 = 0 \pm 0$ $x_0 = 15 \pm 0$ $A = (825.991 \pm 7.662)\times10^6$ $t = 26.717 \pm 0.454$ $L_{1/2} = 18.519 \pm 0.315$

* (Pb,Pb,0) – Pb-target, Pb-moderator without neutron multiplier

** (Pb,C,0) – Pb-target, graphite moderator without neutron multiplier

Note that the number of the primary neutrons produced in these two cases are not the same. In the (Pb,Pb,0) system Y = 66.23 ± 0.07 (stat.) neutrons are produced per proton of 2.5 GeV/c, while in the case of the (Pb,C,0) system Y = 52.38 ± 0.16 (stat.), which is 21% less and is due to the smaller size of the Pb-target in the (Pb,C,0) system and the difference in the type of the material surrounding the target [12].

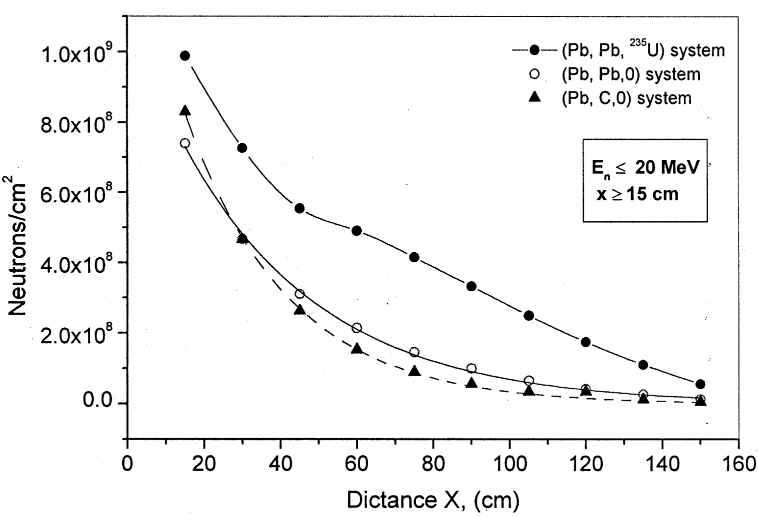

Figure 7. Variation of fluence of neutrons of energy ≤ 20 MeV as a function of distance from the center of the assemblies. Distance x, was measured on the X-axis (See Fig. 1). For the cases that there is no neutron multiplier in the system the fluence as a function of distance can be described with an exponential attenuation curve as shown in the figure. The line joining the solid circles is for the guide of the eye only.

From Fig.7 it can be seen that in the case of the (Pb,Pb,^{235}U) system, the variation of the neutron fluence F(x) with x, is very different from the cases of non-multiplying systems and it cannot be described by an exponential attenuation curve.

In Fig.8 the variation of the neutron fluence as a function of distance, for the (Pb, Pb, 0) system and for $E_n \leq 20$ MeV is shown which also includes the fluence-values for $x < 15$ cm. When all fluence-values with $x \geq 0$ cm are included, the results cannot be expressed in terms of a first order exponential attenuation curve (Eq. 1). However the data points shown in Fig.8 have a near perfect fit to a two-component exponential attenuation curve of the form;

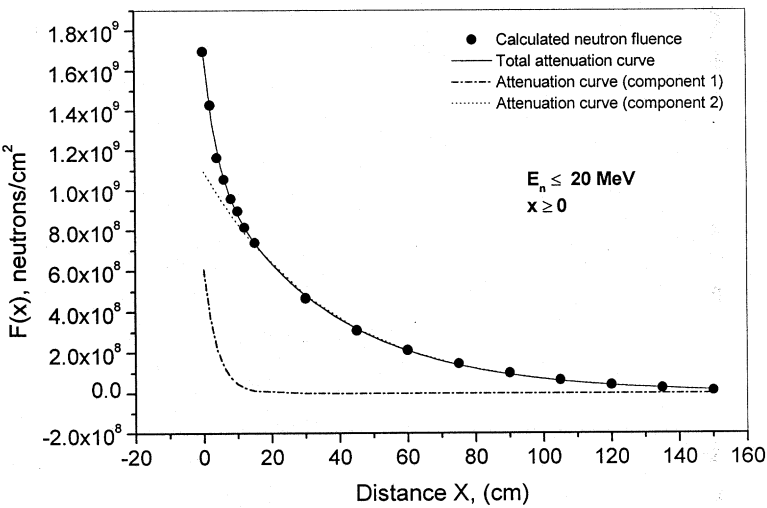

Figure 8. Fluence of neutrons of $E_n \leq 20$ MeV as a function of distance x (on the X-axis, Fig. 1) from the centre of the (Pb, Pb,0) system. The data point fit to a two component exponential attenuation curve of the form given by Eq. 2.

$$F(x) = y_0 + A_1 e^{-(x-x_0)/t_1} + A_2 e^{-(x-x_0)/t_2} \qquad x \geq 0 \qquad (2)$$

The parameters of the Eq.2 for 10^9 incident protons of 2.5 GeV/c are as follows; $y_0 = 0 \pm 0$, $x_0 = 0 \pm 0$ cm, $A_1 = (60.693 \pm 2.933)\times10^7$, $t_1 = 3.882 \pm 0.350$ cm, $A_2 = (109.619 \pm 2.876)\times10^7$ and $t_2 = 36.381 \pm 1.139$ cm.

In Fig.8 the best fit curve to the calculated fluences as well as the curves describing the two components of the exponential attenuation curve are shown. The first component of the Eq.2 has an attenuation length of $t_1 = 3.88$ cm and a half-length of $(L_1)_{1/2} = 2.69$ cm, while for the second component the attenuation length is $t_2 = 36.38$ cm and $(L_2)_{1/2} = 25.22$ cm.

The attenuation and half-length of the more penetrating component are identical (within statistical errors) to those obtained form Eq.1 for fluences corresponding to $x \geq 15$ cm (Table 1). The first component of the attenuation curve dies away for $x \geq 15$ cm.

Detailed analysis of the results showed that the presence of two exponential components in Eq. 2, is not the consequence of the difference in the size of the calculation cells at $x < 15$ cm and $x \geq 15$ cm. We deliberately used larger cell sizes for $x \geq 15$ cm to reduce the statistical errors of the calculations.

Figure 9 shows the variation of the fluence of the low energy neutrons ($E_n \leq 1$ eV) as a function of distance x, for the all three systems studied. As expected and already shown in Fig. 5 the number of slow neutrons in the case of the graphite moderator is much higher than the other two systems. In the case of the (Pb,Pb,0) system the variation of the F(x) with x ($x \geq 15$ cm) can be described by a second order polynomial. However this is not the case with (Pb,C,0) and (Pb,Pb,^{235}U) systems . The slow neutron fluence in the vicinity of the multiplying region is highly depressed (as expected)

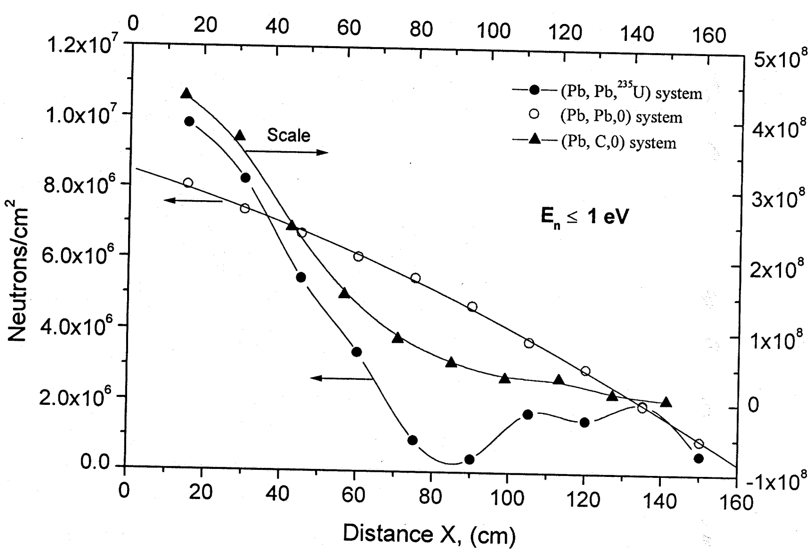

Figure 9. Fluence of neutrons of energy ≤ 1 eV as a function of distance x, from the centre of the assemblies. Distance x, was measured on the X-axis (See Fig. 1). In the case of the Pb, non-multiplying medium ,the (Pb, Pb,0) system, the fluence of the slow neutrons as a function of distance can be fitted to a second order polynomial for $x \geq 15$ cm as shown in the figure. For the two other cases shown in the figure (solid circles and upward triangles) the line joining the data points are to guide the eye only.

Figure 10 illustrates the variation of the neutron fluence with distance x for neutrons with energy in the range of 1 eV < $E_n \leq 1$ keV. Such neutrons are important for nuclear transmutation by adiabatic resonance crossing method [4,5]. As seen in Fig. 10 the existence of a neutron multiplying region highly enhances the number of the neutrons that are relevant to TARC. Also from Fig. 10 it is evident that, as for neutrons with $E_n \leq 1$ eV (Fig. 9) the neutron fluence within and in the neighbourhood of the multiplying region (64< x <107 cm) is highly depressed. This results in F(x) values that are significantly less than the case when no neutron multiplier was present. Therefore in order to exploit the increased neutron fluence (because of the presence of a neutron multiplier), one has to avoid the vicinity of the fissile material region. Thus in the case of the (Pb,Pb,^{235}U) system discussed in this paper we are restricted to the volume with x < 60 cm (Fig. 10).

In the case of the (Pb,C,0) system effectively regions with x < 30 cm will be useful for transmutation by resonance capture and indeed in this region the neutron fluence can compete with (Pb,Pb,^{235}U) system. Note that for the (Pb,C,0) system at x-values of < 30 cm the fluence of neutrons relevant to transmutation via resonance capture is higher by a factor of one to two, compared to the (Pb,Pb,0) system.

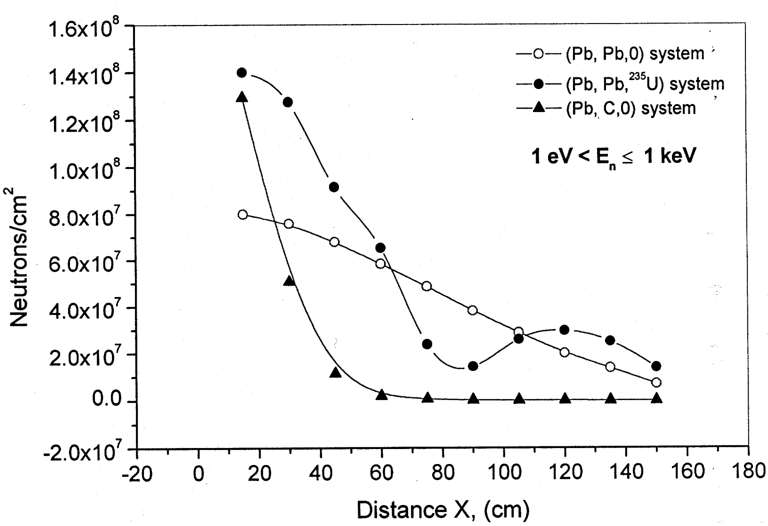

Figure 10. Variation of fluence of neutrons of energy 1 eV < $E_n \leq 1$ keV as a function of distance from the centre of the assemblies. Distance x, was measured on the X-axis (Fig. 1). Lines joining the data points are to guide the eye only.

As already mentioned, not only the fluence of the slow neutrons (< 1 eV) but in general the fluence of all neutrons of energy less than ~ 1 keV is highly depressed (within and in the neighbourhood of the neutron multiplying region (Figs 9, 10)). This is expected, and is a common feature of the heterogeneous nuclear systems. Thus in an accelerator driven system, using large volume of lead as a neutron moderator-storage medium (for a given k_{eff}

-value) one needs to minimise the volume of the neutron multiplying region (the fissile material) to increase the available high-flux volume in the system, for transmutation applications. Therefore if an ADS is designed for nuclear waste transmutation by means of TARC, a heterogeneous lattice structure with fuel elements spread throughout the moderating environment does not seem to be a suitable choice. One will not have such restrictions in homogenous systems where the fissile and moderating materials are uniformly mixed.

5. Nuclear waste transmutation

Given the above results, we studied the transmutation rates of the some radioactive waste nuclei in (Pb, Pb,0) and (Pb, C,0) systems. In this regard the transmutation rates of the ^{99}Tc ($t_{1/2} = 2.1\times10^5$ y) and ^{129}I ($t_{1/2} = 2\times10^7$ y) were calculated. The ^{99}Tc and ^{129}I samples were placed at x = 15 cm and 12 cm respectively. For neutron transport in ^{99}Tc we used the ENDF/B-VI.0 data library and for its (n,γ) transmutation calculations we used the LLLDOS, the dosimetry data library of MCNP-4B2 [11]. For ^{129}I, the ENDF/B-VI.0 data library was used both for neutron transport and transmutation rate calculations. The calculated transmutation rates, B (transmutation per gram of the given waste isotope per incident 2.5 GeV/c proton) and R_B are given in Table 3 where for a given waste isotope. R_B, is defined as;

$$R_B = \frac{B - value \;\; in \;\; system}{B - value \;\; in \;\; (Pb, Pb, 0) \;\; system}$$

Table 3. Transmutation rates (B-values) for ^{99}Tc and ^{129}I in ADSs with graphite and lead moderating environments, at proton energy of E_p =1.73 GeV.

Type of ADS	B-value at energy range			$R_{B(E_n \le 20\ MeV)}$
	$E_n \le 1$ eV	1 eV < $E_n \le 20$ MeV	$E_n \le 20$ MeV	
^{99}Tc at x = 15 cm				
(Pb, C,0)	4.39×10^{-3}	2.29×10^{-3}	6.68×10^{-3}	4.81
(Pb, Pb,0)	4.30×10^{-5}	1.34×10^{-3}	1.39×10^{-3}	1
^{129}I at x = 12 cm				
(Pb, C,0)	4.89×10^{-3}	3.84×10^{-4}	5.27×10^{-3}	14.36
(Pb, Pb,0)	4.52×10^{-5}	3.22×10^{-4}	3.67×10^{-4}	1

It must be noted that in calculating the B-values the ^{99}Tc and ^{129}I samples were embedded in the corresponding moderating media (lead or graphite). In such an irradiation geometry the calculated B-values are somewhat higher than the cases when activation samples were placed in a void channel (e.g. an empty tube) within the moderator. The ^{99}Tc and ^{129}I samples used for calculations were in the form of hoop of thickness 1mm centred the beam axis on XY-plane (Fig. 1). These cannot be considered thin, and there will be self-shielding effects, nevertheless the results given in Table 2 are sufficient for comparison purposes.

From these calculations (Table 3) we should note the following:

- In (Pb, C,0) system 65.7% of ^{99}Tc and 92.8% of ^{129}I transmutations were caused by neutrons of $E_n \leq 1$ eV.
- 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.
- B-values in the (Pb, Pb,0) system, i.e transmutation by adiabatic resonance crossing (TARC) are much less than B-values in the (Pb, C,0) system, i.e. transmutation in a slow neutron dominated neutron field. For identical geometrical locations within the moderators and for the same nuclear waste composition, size and mass the (Pb, C,0) system transmutes ^{99}Tc by factor of 4.8 and ^{129}I by a factor of 14.4 higher than the (Pb,Pb,0) system.
- The above observations lead to the conclusion that in an accelerator driven system designed for nuclear waste transmutation, graphite moderators and those of similar properties are much more suitable than lead, at least for transmutation of ^{99}Tc and ^{129}I. With such moderators the spallation neutrons are used much more efficiently and much higher transmutation rates for ^{99}Tc and ^{129}I are obtained, compared to lead and other comparable moderators.

In the (Pb, Pb,0) system the B-values of ^{99}Tc and ^{129}I for protons of momentum 3.57 GeV/c were also calculated and values of $(2.14 \pm 0.06)\times10^{-3}$ for ^{99}Tc and $(5.51 \pm 0.16)\times10^{-4}$ for ^{129}I were obtained at the sample locations x = 15 cm and x = 12 cm respectively.

The B-values for ^{129}I and ^{99}Tc in a geometrical set-up and location, identical to that in ref. 5 (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. 5. We obtained a B-value of $(3.87 \pm 0.27)\times10^{-4}$ for ^{129}I that is equivalent of $(2.50 \pm 0.18)\times10^{-5}$ 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 $(2.61 \pm 0.26)\times10^{-5}$ [5]. In the case of ^{99}Tc, B-values close to the experimental results of ref. 5 are obtained only for thick and dense samples of ^{99}Tc. We consider a spherical volume of 1 cm^3. If we distribute 216 mg of metallic ^{99}Tc throughout this volume (average density 0.216 gcm^{-3}) we find a B-value of $(3.13 \pm 0.25)\times10^{-3}$. As we increase the mass to 11500 mg (density now 11.5 g cm^{-3}) the B-value becomes $(1.40 \pm 0.10)\times10^{-3}$. The B-values reported in Ref. 5 are 2.34×10^{-3}, 1.52×10^{-3} and 1.24×10^{-3} for ^{99}Tc samples of mass 14.3, 100.9 and 216.1 mg respectively.

6. Conclusions

We used the LAHET code system to study the behaviour of spallation neutrons resulting from interaction of 2.5 GeV/c proton with a massive lead target within large (3.3×3.3×3 m^3) lead and graphite moderating environments.

It is shown that with lead moderator the peak of the energy distribution of neutrons appears at ~520 keV in the presence or absence of a neutron multiplying medium in the system. This is independent of the location within the moderator.

In the case of the graphite moderator as expected the neutron field is dominated by slow neutrons ($E_n < 1$ eV) while in the lead moderator the slow neutron fluence does not exceed 8.4% of the total neutron fluence at a given location.

The variation of the neutron fluence ($E_n \leq 20$ MeV) for both lead and graphite moderators (in absence of any fissile material in the system) can be described very well using a first order exponential attenuation curve for $x \geq 15$ cm where x is the distance from the centre of the assembly in direction normal to the proton beam line. We obtained attenuation length of 36 cm and 26.7 cm for lead and graphite moderators respectively.

In the case of the lead moderator when the fluence values corresponding to $x < 15$ cm are included in the plots, a two component exponential attenuation curve gives an excellent fit to the data.

The LAHET calculations lead to the conclusion that in the accelerator driven systems studied, the transmutation rates of ^{129}I and ^{99}Tc by adiabatic resonance crossing within a lead moderator are significantly less than the case of transmutation in a slow neutron dominated field, within a graphite moderator.

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Исследование ADS-систем методом Монте-Карло.
Энергетическое и пространственное распределения нейтронов
в размножающих и неразмножающих средах

Поведение полей нейтронов расщепления, рождаемых в результате взаимодействия протонов при 2,5 ГэВ/с с массивной свинцовой мишенью, окруженной большим объемом (~ 30 $м^3$) свинцового и графитового замедлителя, изучено с помощью программы LAHET. Исследованы пространственное и энергетическое распределения нейтронов в присутствии и в отсутствие делящихся материалов в системе. Рассчитана скорость трансмутации долгоживущих ядерных отходов, ^{99}Tc и ^{129}I, в свинцовой и графитовой замедляющей среде. Показано, что в исследуемых ADS-системах скорость трансмутации ^{99}Tc и ^{129}I посредством адиабатического резонансного рассеяния (TARC) существенно меньше, чем в случае, когда трансмутация производится нейтронным полем, в котором доминируют тепловые нейтроны.

Работа выполнена в Лаборатории высоких энергий ОИЯИ.

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Hashemi-Nezhad S.R. et al. E1-2000-291
Monte Carlo Studies of Accelerator Driven Systems.
Energy and Spatial Distribution of Neutrons
in Multiplying and Non-Multiplying Media

The LAHET code system was used to study the behaviour of the spallation neutrons resulting from the interaction of 2.5 GeV/c protons with a massive lead target within a large (~ 30 m^3) lead and graphite moderating environments. The spatial and energy distributions of neutrons with presence and absence of a fissile material in the system were investigated. Transmutation rates of long-lived nuclear wastes, ^{99}Tc and ^{129}I, in lead and graphite moderating environments were calculated. It is shown that in the accelerator driven systems studied, the transmutation rates of ^{99}Tc и ^{129}I by Adiabatic Resonance Crossing (TARC) are significantly less than in the case of transmutation in a thermal neutron dominated, neutron field.

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

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