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INFLUENCE OF DIFFERENT MODERATOR MATERIALS ON CHARACTERISTICS OF NEUTRON FLUXES GENERATED UNDER IRRADIATION OF LEAD TARGET WITH PROTON BEAMS

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Introduction

It is shown /1-2/ that effective transmutation of radioactive wastes accumulated over the decades of the employment of nuclear power plants and other nuclear facilities worldwide using intensive neutron fluxes is fulfilled only in the fields of neutrons with spectrum varying from thermal up to rather high energies depending on the ratio of neutron capture and fission cross sections for each radioactive isotope. It means that in case of accelerator driven facilities, i.e. the facilities which include a high current proton accelerator (intensity of the proton beam about 10 mA, proton energy ~ 1 GeV) in combination with an assembly of an extended heavy (lead, lead-bismuth etc) target and fissile blanket providing rather high neutron generation, multiplication and utilization characteristics, development of some more or less optimal transmutation scenario requires employment of different moderator materials. The scope of such materials is normally limited to those used in the present nuclear reactor technologies; these materials show rather large variety in neutron moderation and absorption properties, therefore it seems reasonable at the present stage of the research activity to explore the influence of as many reflector materials as possible both experimentally and theoretically.

In recent years some experimental studies of characteristics of neutron generation and transport inside small target systems (lead primary target ∅8 cm × 20 cm surrounded with 6 cm paraffin moderator) were carried out at LHE JINR, Dubna, Russia /3/. Sometimes this heavy target was modified by inserting a natural uranium rod as a central core of the target assembly. Samples of several radioactive isotopes (^{129}I, ^{237}Np and ^{239}Pu) as well as some neutron detectors (^{139}La and ^{nat}U) were placed on top of the paraffin to study transmutation and neutron flux properties. These setups were irradiated with protons provided by LHE JINR Synchrophasotron/Nuclotron at energies in the range from 0.5 up to 7.4 GeV.

Another experimental approach is to introduce a fissile blanket surrounding the primary target /4,5/ (in case of the experiments performed at LHE JINR this blanker consisted of 206 kg of natural uranium). Due to neutron safety and shielding considerations this more sophisticated assembly was surrounded with a box containing polyethylene moderator and neutron (cadmium) and gamma-quanta (lead) absorbers. This setup was irradiated with a proton beam at 1.5 GeV.

In an earlier work (S.R.Hashemi-Nezhad et al. Nucl. Instr. and Meth. A 482 (2002) 547-557) we have shown that in large set-ups (~ 10 m^3) graphite is much more effective than lead for in transmuting the transuranic and long-lived fission-fragment nuclear waste isotopes. This article is aimed to study by means of Monte-Carlo simulation (DCM/CEM /6/) the influence of another moderating material such as graphite on the neutronics of a small setup consisting of the lead neutron-generating target surrounded with a layer of a moderator material. Comparison between the neutron spectra at the surface of such assemblies in case of paraffin and graphite is performed. Neutron fluxes at the surface of a rectangular graphite block covering the upper surface of the lead target are investigated depending on the thickness of the moderator material.

Cylindrical setup

It is well known that materials with high content of light atoms (e.g. hydrogen) display the best moderating ability due to large energy transfer per collision as compared with heavy materials. One of the best moderators in this row is paraffin. However its physical properties, in particular very low melting point, don't allow it to find wide use in reactor technology and limit its employment to laboratory applications. Therefore it is been proposed to try graphite (a well known moderator material widely used in nuclear reactors) as a replacement to paraffin in the setups used in transmutation experiments.

Prior to the experiments, the neutronics of the lead-paraffin and lead-graphite assembly has been performed by means of Monte-Carlo simulation using the computer program DCM/CEM /5-8/. This code is based on modelling of the intranuclear cascades, described in the frame of the Bertini model and pre-equilibrium evaporation model and includes the channel of high-energy fission of medium and heavy nuclei (A>100). Neutron transport in heterogeneous media at lower energies (E<10.5 MeV) is simulated by means of the Monte-Carlo method in the transport approximation. Properties of neutron collisions with atoms of the moderator material at thermal energies are simulated considering the thermal motion of atoms (Maxwell-Boltzmann spectrum) with subsequent recalculation of the cross section. Both high energy and low energy part of the calculation procedure take into consideration heterogeneous structure of the setup. Simulation of the neutron transport is performed basing on the neutron cross-section library ARAMAKO /10/.

The setup used at the first stage of the calculations is similar to that used in experiments held in recent years at LHE JINR to study transmutation properties of radioactive waste samples /3/. The schematic drawing of the set-up is shown in Fig.1. The setup consists of a primary lead target, which converts the proton beam into secondary particles resulting from spallation reactions. The size of this target is ∅8 cm × 20 cm. This target is covered with a paraffin cylinder with the size ∅20 cm × 30 cm. Protons strike the front plate of the lead target, which is shifted by 4 cm backwards with respect to the front plate of the moderator. Proton energy is assumed to be 1 GeV. To compose the neutron spectra the neutrons traveling through the target-moderator volume are registered when crossing the surface of the detector volumes, defined as cylindrical shell of infinitesimal thickness at the surface of the moderator. This approach is different from the shape of the detectors used in the experiments (small cylindrical vials filled with Lanthanum or Uranium salts), however it takes the advantage of the azimuthal symmetry of the setup and significantly improves the statistics of the calculations. Usually the positions of the detecting volumes in calculations coincide with the positions of the samples and detectors used in real experiments. However, for the purpose of this article the position of the detecting surfaces is of no special importance and we use them just for the sake of comparison. In the case under discussion the width of the detector shell was taken to be 0.7 cm, which was located at a distance of 7.15 cm from the front plate of the moderator. In our calculations we considered that the setup includes a steel reflector to compensate the influence of the shielding and equipment materials present in the experimental hall. It has been taken to be a steel cylinder with 50 cm thick walls at a distance of 3 meters from the surfaces of the moderator. This reflector is not shown in Fig.1 for the sake of simplicity. Of course this assumption is rather crude as the configuration of the experimental hall and especially the equipment around the installation is rather complicated but at this stage of the calculations it seems to be enough.

In the case of the graphite moderator the size and the shape of the moderator was taken to be exactly same as that of the paraffin moderator. The densities of the paraffin and graphite were taken to be 0.89 g/cm^3 and 2.65 g/cm^3, respectively. Estimated neutron spectra are presented for both cases in Fig.2. The resulting shape of the neutron spectrum shows no presence of the thermalization peak in the region of lower energies, which means that to obtain the same densities of thermal flux one needs to use considerably thicker layers of the graphite moderator. As for the integral values characterizing the effectiveness of the setup as a neutron generating assembly some of the figures are shown in Table 1.

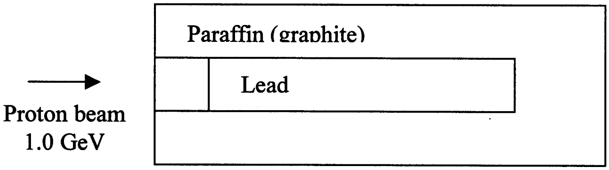

Fig.1. Schematic drawing of the setup used for calculations and comparison.

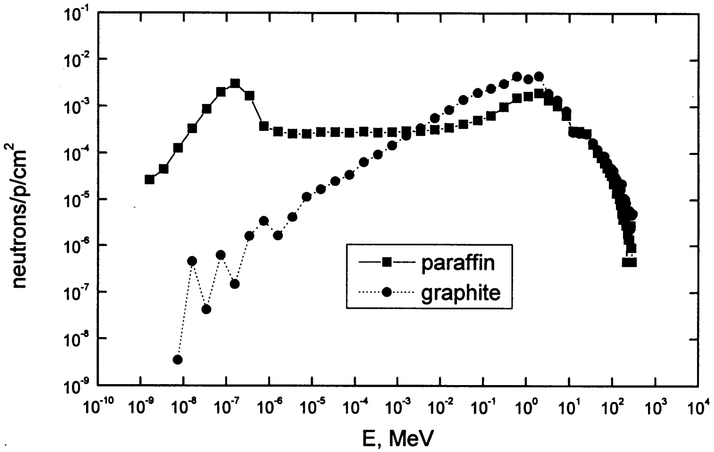

Fig.2. Neutron spectra at the surface of paraffin (graphite) moderator.

Total number of neutrons (see Table 2) is evidently the same in both systems (56.8 and 55.9 for paraffin and graphite respectively); still they are redistributed in a sense that much larger amount of neutrons is absorbed in paraffin on hydrogen atoms.

Table.1 Multiplicity of neutrons and some of nuclear properties

	Total number of neutrons	Number of captures	Neutron leakage
Paraffin	56.8	35.8	21.0
Graphite	55.9	19.6	36.3

For the setups considered here the thermal and epithermal neutron density is much less in the case of the graphite as compared with the paraffin setup.

Rectangular setup

It seems that in the future experiments the appearance of the setup will be different from the one assumed at the first stage of the calculations. The proposed setup may look like the assembly shown in Fig.3. In this case the primary lead target (∅8 cm × 50 cm) is cutting into the volume of the graphite moderator to the size of its radius. The size of the graphite block is taken to be 20 cm × 10 cm × 50 cm, i.e. the length of the target and the moderator block are identical and equal to 50 cm. It means that the effective thickness of the moderator in the vertical direction is equal to about 6 cm and it can be further enlarged. The front plate of the moderator block coincides with that of the target. The shape of the assembly is not symmetric. As it is supposed that the moderator block will be placed only on top of the target and the calculation algorithm does not allow one to introduce concave target geometry the target-moderator assembly was supplemented with a symmetric rectangular block filled with vacuum. The supplement does not influence the neutron distribution inside the assembly. On the other hand one can estimate and compare the neutron densities at the same distance, i.e. at the top surface of the moderator block and at the opposite imaginary surface behind the layer of vacuum. Both surfaces are "cut" into a lattice with the size 10 cm along the beam direction for the sake of simplicity we will denote this direction as Z-axis. The coordinate system is tied to the center of symmetry of the section perpendicular to the beam line, i.e. the zero coordinates are located in the central point where the beam strikes the target surface. One can take the advantage of the "left-right" symmetry of the neutron flux in respect to the negative and positive values of the coordinate. Thus the horizontal coordinate (see right part of Fig.3, called X-coordinate) was "cut" into a grid with a step of 2 cm. The neutrons are registered when crossing the respective element of the surface considering the absolute value of this horizontal X-coordinate and the value of the Z-coordinate. Again, a similar approach was taken when the upper graphite and lower vacuum surfaces were considered to register the neutron spectra.

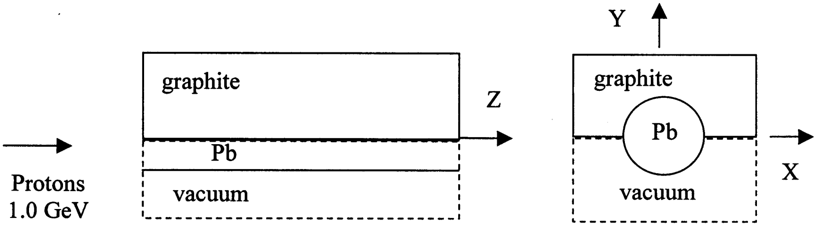

Fig.3. A scheme of the rectangular setup considered in our calculations.

First of all let's consider the development of the neutron flux at both upper and lower detecting surfaces along the beam trajectory. In Fig.4a,b one can see the neutron spectra at the surface of graphite and vacuum blocks depending on the distance from the beam entrance into the target (Z axis) in the 4 cm wide strip (X from -2 cm to +2 cm). In principle they show the same behavior and about the same energy dependence. In fact, they look similar to the spectrum at the cylindrical setup surface. Some dropdowns in the spectrum at lower energies are coming from rather poor statistics (only few neutrons are registered in each of these energy bins). At the first 20 cm the density of the neutron flux keeps almost flat then it starts to decrease. In both cases the density of the neutron flux falls down by one order of magnitude along the 50 cm from the front plate to the back end of the assembly. To simplify comparison between the spectra on the graphite and vacuum surfaces a combined graph demonstrating the neutron flux at 10 to 20 cm along the Z-axis and at -2 to 2 cm along the X axis is shown in Fig.5. Data obtained by means of the renowned computer codes LAHET /11/ and MCNP-4B /12/ describing particle production and transport at high energies and neutrons at energies below 20 MeV in extended media are also shown in Fig.5.

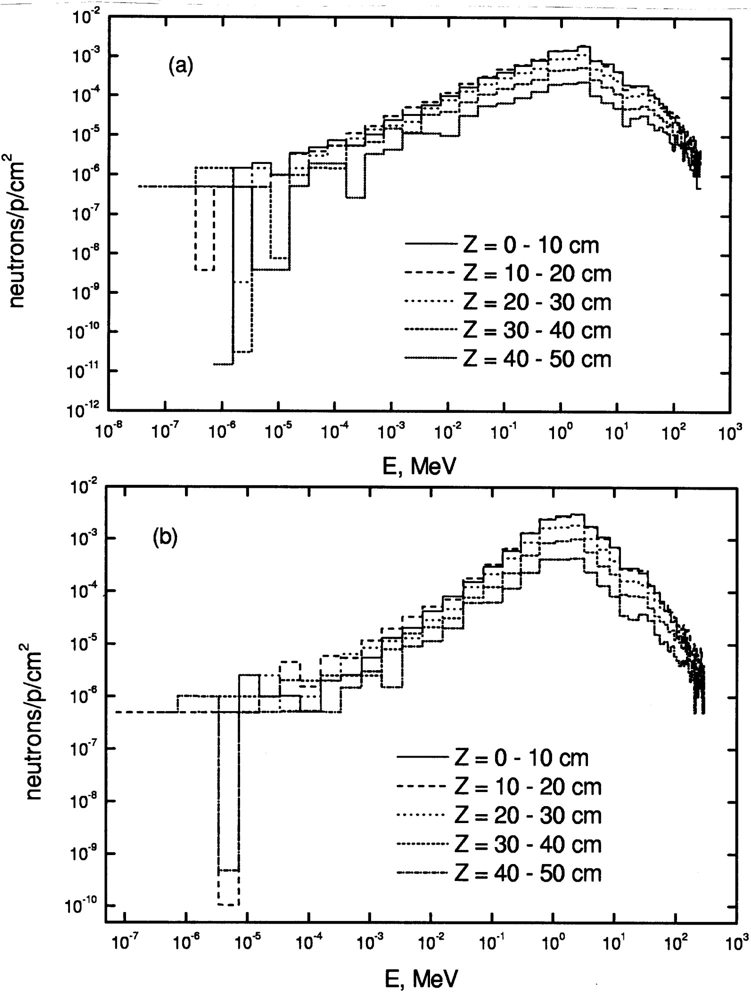

Fig.4. Dependence of the neutron spectra at the top surface of the graphite (a) and bottom surface of the vacuum (b) blocks on the position of the surface element along the Z-axis.

Both computer codes give similar description of the neutron spectrum except that MCNP predicts somewhat softer spectrum behind the moderator, which can be explained by the fact that the MCNP calculation does not consider the presence of the steel reflector. On the other hand the DCM/CEM calculation does not take into account crystal lattice in the graphite, which is probably distorting the simulation of the process of neutron moderation. As it can be seen from Fig. 5, the flux at the surface without the moderator (vacuum) appears to be slightly harder than that behind the moderator, however the difference is not as large. The reason of this difference in the spectrum is possibly explained by the fact that the neutron flux at a distance of 10 cm from the center of symmetry of the lead target in absence of moderator is influenced not just by the particles escaping the primary lead target surface but also by the neutrons leaking (reflected) from the bottom surface of the graphite moderator.

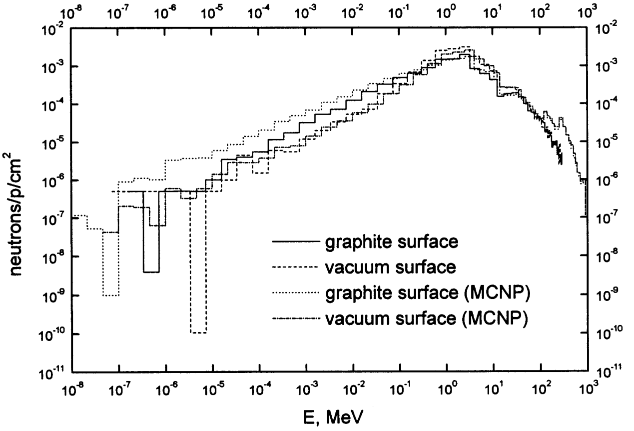

Fig.5. Comparison of neutron spectra (surface element with X from -2 to 2 cm and Z from 10 to 20 cm) on top of the graphite and bottom of the vacuum blocks.

The geometry of the rectangular moderator block is rather complicated in comparison with the cylindrical case where the surfaces of the target and the moderator are equidistant and one can foresee azimuthal symmetry of the neutron flux. Here the effective thickness of the moderator material is changing with the displacement along the X-axis. Dependence of the neutron spectra on the position of the surface element on the displacement of the detecting strip along the X-axis is shown in Fig.6 (again a detecting strip from 10 to 20 cm along the Z axis is considered). Surfaces of the graphite and vacuum blocks are considered (see Fig.6 (a) and Fig.6 (b), respectively).

The density of the neutron flux changes by a factor of about 1.7 on the vacuum surface (see Fig.6 (b)) when the detection strip is moved from interval of X=0-2 cm to X=8-10 cm. On the surface of the moderator this factor raises to 2.7. Comparison of the spectra behind the graphite and vacuum block leads to a rather interesting conclusion. In such asymmetric setups graphite works rather as a moderating reflector and the neutron flux behind the moderator is a little softer than that without the moderator. Neutron spectra are essentially not dependent on the position at the "vacuum" side.

Finally, it is interesting to see the change of the neutron spectrum with an increase of the moderator thickness. We estimated the neutron flux at the surface element with Z from 10 to 20 cm and X from -2 to 2 cm depending on the moderator thickness that changed from 10 to 16 cm in 2 cm steps. Results of the comparison are presented in Fig.7. As expected, the contribution of the low energy tail of the neutron flux is getting larger with the growing thickness of the moderator and the effect around epithermal energies is around one order of magnitude.

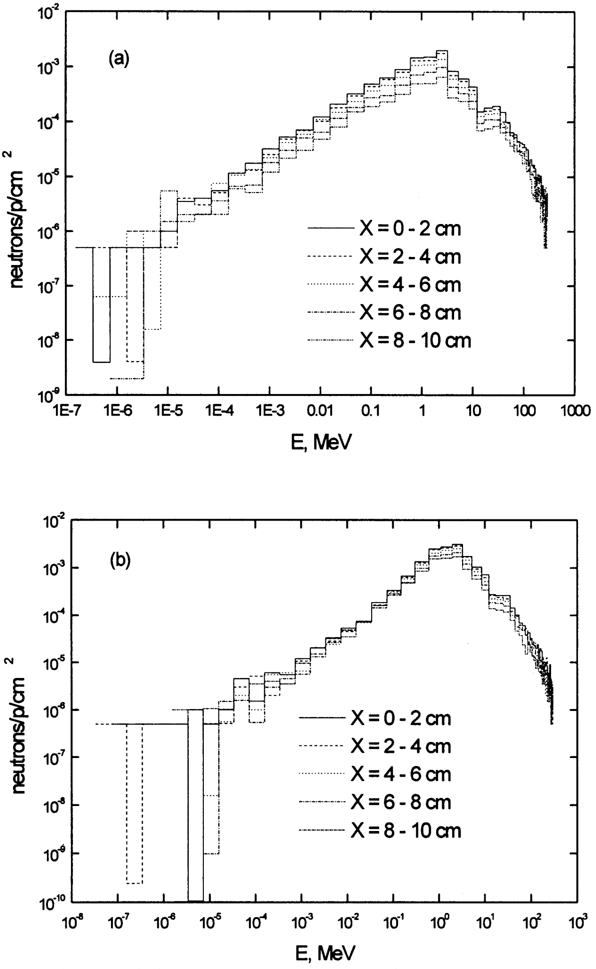

Fig.6. Dependence of the neutron spectra (surface elements from 10 to 20 cm along the Z axis) on the position along the X-axis on top of the graphite (a) and bottom of the vacuum (b) blocks.

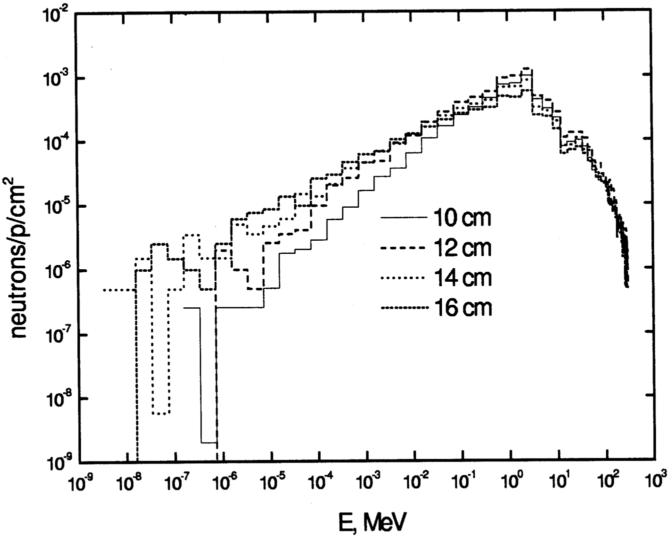

Fig.7. Dependence of neutron spectra on the moderator thickness.

Conclusions

The influence of different types of moderator materials (paraffin, graphite) on the neutron spectra at the surface of the cylindrical lead target – moderator setups is studied. It is shown that for the setups considered here the spectrum obtained with the graphite moderator is harder than the spectrum from paraffin moderator and it lacks the thermalization peak at lower energies.

As expected, the neutron spectrum becomes softer with increasing thickness of the moderator. Neutron spectrum in such asymmetric setup as the one considered in the present work (see Fig.3) appears to be slightly harder on the "vacuum" side than on the top surface of the graphite block. Neutron spectrum at the "naked" side of the primary target is influenced by the neutrons reflected from the bottom of the graphite block.

Acknowledgements

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Влияние различных замедляющих материалов
на характеристики потоков нейтронов, генерируемых
при облучении свинцовых мишеней пучками протонов

Исследуются поля нейтронов, генерируемые в протяженных мишенях ($Z \geq 82$) при облучении протонными пучками с энергией в области 1 ГэВ. Сравнивается влияние различных замедлителей на спектры и множественность нейтронов, вылетающих с поверхности сборки, состоящей из свинцовой мишени (⌀ 8 см × 20 см или ⌀ 8см × 50 см), покрытой слоями полиэтилена или графита различной толщины. Показано, что эффективность графита, используемого в таких сборках в качестве материала, замедляющего нейтроны до тепловых энергий, значительно хуже, чем эффективность полиэтилена.

Работа выполнена в Лаборатории информационных технологий ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2002

Sosnin A. N. et al. E2-2002-258

Influence of Different Moderator Materials
on Characteristics of Neutron Fluxes Generated
under Irradiation of Lead Target with Proton Beams

Neutron fields generated in extended heavy ($Z \geq 82$) targets under irradiation with proton beams at energies in the range of 1 GeV are investigated. Influence of different moderators on the spectra and multiplicities of neutrons escaping the surface of the assembly consisting of a lead target (⌀ 8 cm × 20 cm or ⌀ 8cm × 50 cm) screened by variable thickness of polyethylene or graphite, respectively, was compared in the present work. It is shown that the effectiveness of graphite as a material used in such assemblies to moderate spallation neutrons down to thermal energies is significantly lower than that of paraffin.

The investigation has been performed at the Laboratory of Information Technologies, JINR.

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