# Experience gained during 10 years transmutation experiments in Dubna

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**Abstract** Transmutation, the procedure of transforming long-lived radioactive isotopes into stable or short-lived, was proposed for reducing the amount of radioactive waste resulting from technological applications of nuclear fission. The Accelerator Driven Systems (ADS) provide the possibility to generate intense neutron spectrum yielding in an effective transmutation of unwanted isotopes. Such experiments are being carried out for the last 10 years in Synchrophasotron / Nuclotron accelerators at the Veksler-Baldin Laboratory of High Energies of the Joint Institute for Nuclear Research in Dubna, Russia. Thick Pb and Pb-U targets, surrounded by moderators, have been irradiated by protons in the energy range of 0.5-7.4 GeV. Neutron fluence measurements have been performed by different techniques of passive detectors (neutron activation detectors, solid state nuclear track detectors). Transmutation of <sup>129</sup>I, <sup>237</sup>Np, <sup>239</sup>Pu was studied. The results of these experiments are presented and discussed.

# 1. Introduction

Highly radioactive waste is being accumulated during the last 60 years due to the operation of commercial nuclear power plants and other nuclear facilities. Large quantities of such waste constitute a serious problem for mankind both from a commercial as well as from an ecological and safety point of view. The development of subcritical nuclear assemblies for the transmutation of long lived radioactive waste, have been suggested by several teams last decade. The new approach is the development of Accelerator Driven Systems (ADS) [1]. In these systems high-energy light particles (practically protons) from powerful accelerators produce spallation neutrons by hitting heavy targets. The neutrons produce subsequently copious fission and fission chain reactions. In this way, nuclear fission energy is produced and the energy of the input particles is amplified [2]. Neutron reactions can destroy fission products into short-lived fission fragments or to transform long-lived radioactive waste nuclei into shorter-lived nuclides. The energy spectrum of spallation neutrons extends up to a few hundred MeV. The exact shape of the neutron spectrum depends on wide range parameters, such as the position within the target assembly, the target composition, dimensions and others. Studies of these and associated problems have also been carried out during last decade -among others- at the Laboratory of High Energies (LHE), Dubna. Parameters having strong

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influence on the neutron spectrum were examined in order to investigate best operation conditions of such a set up regarding transmutation efficiency. The results presented in this work are referred to the target and projectile masses and energy. As target mass we have experienced on Cu, Pb, U and combined targets of U/Pb. The dimensions of the targets were almost constant. Projectiles were delivered from Synchrophasotron and from 2000 Nuclotron accelerators. Most of experiments were performed with protons but also deuterons, alphas and carbon were used [3, 4]. The beam energy was varied from 0.5 up to 7.4 GeV. Spatial neutron distributions around the target have been measured as well as calculated in every case by the LAHET code (Los Alamos) and the DCM (Dubna Cascade Model) developed in Dubna [5, 6]. Typical examples of long-lived and highly radiotoxic nuclides that can be transmuted to short-lived or stable nuclei as <sup>237</sup>Np and <sup>129</sup>I were used in these experiments [6, 7, 8]. The case of <sup>239</sup>Pu was also studied in some of the experiments [9, 10]. The results concern neutron production, transmutation efficiencies of the setup and cross sections under the conditions examined. Perspectives and future plans are also presented in this work.

## 2. Experimental

#### 2.1. The experimental setup

The experiments were carried out at the Synchrophasotron accelerator and last years at the new Nuclotron accelerator. Five scintillation detectors were used to monitor the beam. Polaroid films were used for the definition of position, size and shape of the beam on the target. Al activation foils were used to determine the integral proton fluence on the target. The Al monitor foil stack was placed approx. 60 cm upstream the target in order to avoid activation from backward emitted particles. The beam intensity was determined via <sup>27</sup>Al(p,3pn)<sup>24</sup>Na reaction in the central foil. A diagram of the setup using for last years experiments (GAMMA 2 setup) is presented in figs.1a and 1b. Details of beam monitoring and measurements of beam profile are given in Ref [7]. Target had a cylindrical shape of final length 20 cm and diameter of 8 cm, fig. 1b. The system was surrounded by a paraffin moderator, 6 cm thick. The beam side was open.

In all experiments the target was irradiated by a fluence of  $10^{12}$  to  $10^{13}$  particles.



Figure 1. a) Irradiation set up with beam monitors and the target. b) Pb target with the paraffin moderator.

#### 2.2. Measurement of the spatial neutron distribution

Neutron measurements were carried out by activation methods and Solid State Nuclear Track Detectors. a) Activation detectors

<sup>139</sup>La(n, )<sup>140</sup>La reaction was used as neutron monitor (also as transmutation sample). Samples containing 1 g of <sup>139</sup>La each were placed on top of the target assembly at distances of 5, 10, 15, 20 and 25 cm from the front side of the paraffin block. After irradiation the radioactive samples were measured by using a HPGe detector in order to study gamma activity. From the measured activity and the calculated "effective cross-section" [6] neutron distribution was determined along the moderator surface.

 $^{238}$ U of about 4 mg was placed on the middle of the moderator surface. The reaction  $^{238}$ U(n, ) was used for thermal-epithermal and  $^{238}$ U(n,2n) for 6-20 MeV neutrons.  $^{197}$ Au of about 4 mg was placed at the same position with  $^{238}$ U. The reaction  $^{197}$ Au(n, ) was used for thermal-epithermal and  $^{197}$ Au(n,2n) for 6-30 MeV neutrons.

<sup>nat</sup>Cd foils were also used as sensors for thermal-epithermal neutron determination along the paraffin surface. For neutron determination an "effective cross-section" was calculated for each reaction [11].

b) Solid State Nuclear Track Detectors. Each set of SSNTDs contains CR39 d

Each set of SSNTDs contains CR39 detectors that act as particle detector in contact with Kodak LR115 Type 2B which is the neutron converter. The system was partially covered by 1 mm Cd. So each set was able to detect thermal-epithermal neutrons and intermediate-fast neutrons. A free CR39 foil was also served to detect proton recoils and give supplementary information about fast neutrons namely in the neutron energy range of  $0.3 < E_n < 3$  MeV which is the range with about constant response to protons. Details on the operation of these systems and their response are given in refs. [12, 13]. The detectors were positioned every 3cm along the paraffin surface (so parallel to the target).

Sets of SSNTDs as fission detectors with  $^{235}$ U,  $^{238}$ U and  $^{232}$ Th were placed on the middle of the moderator surface for measuring thermal and fast neutrons at energies above 1 MeV. They consist from about 200 µg/cm<sup>2</sup> of target evaporated on Makrofol detector. Details for the method are given in ref. [14].

#### 2.3. Transmutation samples

Transmutation samples examined during last years experiments concern to <sup>139</sup>La, <sup>129</sup>I, <sup>237</sup>Np and <sup>239</sup>Pu. <sup>139</sup>La is not a radioactive nuclide and so has several advantages making it suitable for transmutation studies with lowenergy neutrons. The induced radioactivity is short lived and has strong and clear -ray lines. On top of the moderator 0.425 g <sup>129</sup>I, 0.742 g <sup>237</sup>Np and 0.45 g <sup>239</sup>Pu were placed. <sup>129</sup>I, under the form of NaI, had an isotopic composition of iodine 15% <sup>127</sup>I and 85% <sup>129</sup>I. <sup>237</sup>Np were in the form of NpO<sub>2</sub>. The iodine isotope samples were obtained from the Bochvar Institut (VNIINN), Moscow. The radioactive samples I, Np and Pu were prepared by the Institute of Physics and Power Engineering, Obninsk, Russia. As both samples of I and Np were radioactive even without activation, the irradiated samples were measured at a distance 20 cm from an HPGe detector. Especially for Np sample a Pb plate 1cm thick is placed between the sample and the detector for reducing considerably the low energy gamma activity. Analysis of gamma spectra is carried out using state of art techniques described in ref. [8]. For transmutation of <sup>239</sup>Pu fission fragments were identified in the analysis of gamma-ray spectra. Knowing the relative fission yields for thermal-epithermal neutron induced fission of <sup>239</sup>Pu it is possible to calculate transmutation via fission [7, 9].

## 3. Results and discussion

#### 3.1 Spatial neutron distribution

Neutron distribution along the paraffin moderator surface was studied by SSNTDs and La activation. In figs 2a and 2b the thermal-epithermal and fast neutron fluences measured by SSNTDs are presented. The results correspond to Pb target irradiated with protons of 0.65 to 3.7 GeV. Both distributions of thermal-epithermal and fast neutrons are peaked at around the middle of the moderator length reflecting nuclear processes taken place in the target i.e. the competition of particle production and beam attenuation. The distance of the distribution



Figure 2. a) Thermal-epithermal neutron and b) fast neutron distribution along the outer paraffin moderator surface.

maximum from the side of the beam entrance in the target is related to the proton mean free path and is independent on the target length as it is confirmed by experiments in longer targets [15, 16]. In figs 2a and 2b the role of the beam energy is also shown. Higher neutron fluences were produced by more energetic protons. The result is expected by the study of particle production in high energy physics and was attributed mainly to the higher energy of the primary and secondary particles [17, 18]. At these beam energies reaction cross section remains almost constant with beam energy. Indeed the effect can be observed by the maximum of the longitudinal distribution which is presented at the same position along the paraffin moderator, about one mean free path in the target. Similar results are deduced by <sup>139</sup>La sensors, fig. 3. By <sup>139</sup>La activation detectors the symmetry of the neutron distribution around the moderator was studied. The influence of the beam alignment along the target is also discussed in ref [7].



Figure 3. <sup>139</sup>La activation giving the neutron distribution along the outer paraffin surface.

## 3.2 The target

At the GeVs energies reaction cross section depends only by the target and projectile masses. Several targets such as Cu, Pb, U and combined targets as U/Pb were irradiated with various projectiles. As an example in fig. 4 the neutron fluences measured at the moderator surface are given as a function of the atomic number of the target for <sup>12</sup>C projectile. The results are given in row data i.e. in tracks.cm<sup>2</sup>.proton<sup>-1</sup> because the conversion of track number to neutrons requires calculation of the neutron spectrum which was not known for all experiments. Each point in the figure corresponds to the maximum of the longitudinal distribution along the moderator surface. The increasing behaviour of the neutron production with increasing target atomic number follows the cross section law.



Figure 4. Neutron production on the outer paraffin surface as a function of target atomic mass for <sup>12</sup>C projectile.

By similar studies on the neutron production by spallation reactions on heavy targets is known [18, 19] that the great part of neutrons escaping the target surface is in the intermediate–fast neutrons. The role of the moderator in the presented experiments can also be seen by the production of thermal-epithermal neutrons which are about of the same order comparing to fast neutrons. In the case of combined targets U target was surrounded by Pb with total target dimensions of about the same of the single target.

## 3.3 The projectile

A lot of experiments were performed by others with various projectiles in the same energy range as our experiments and selected data are given in literature [18-20]. In most of these experiments proton beams were used because protons are of importance in Energy production by Accelerator Driven Reactors [1, 2]. In fig. 5 thermal-epithermal and fast neutron components are presented as a function of the projectile atomic number for protons, deuterons, alphas and carbon. The given results were obtained by Pb target and correspond to the maximum of the longitudinal distribution along the moderator surface. As it was expected from the cross section law heavier projectiles produce more neutrons. The dependence on projectile atomic number is clearer by the slope of the almost linear behaviour at higher projectile energies (in log scale).



Figure 5. Neutron production in the outer moderator surface as a function of the projectile atomic number for Pb target.

#### 3.4 Beam energy dependence

The experiments were performed at beam energies from 0.55 GeV to 7.4 GeV. Two sets of curves are given in figs 6 for thermal-epithermal and fast neutrons for Pb and U/Pb targets irradiated with proton beams. The increasing neutron production with beam energy is mainly attributed to the higher energy of secondary particles and their ability to produce secondary reactions (and less of higher order reactions) [17, 20].



Figure 6. Energy dependence of neutron production in the outer moderator surface for Pb and combined U/Pb targets.

Comparing the results in fig 6 it is deduced that U/Pb target produce about two times more neutrons than Pb target. However spallation cross section of U is not two times the Pb cross section, so secondary reactions in the U target must be considered. Secondary reactions on the Pb target are of less importance.

#### 3.5 Neutron cost

The neutron cost is referred in literature [2, 18] as the neutron production in the target per incoming ion. It was proved that the maximum production and so the best neutron cost is around 1 GeV. For transmutation experiments the most important factor is the neutron spectrum concerning the possibility to (n,f) or (n, ) reaction exploitation. Alternatively a hard neutron spectrum can be useful if the transmutation experiment is based on (n,xn) reactions. In our experiments the neutron production at the moderator surface as a function of the proton energy are given in fig. 7. The results are given as the mean value of the neutron production relatively to the total neutrons escaping the moderator surface. It is shown that the neutron economy was obtained at lower energies than 1 GeV.



Figure 7. Neutrons escaping the outer moderator surface as a function of the proton beam energy for Pb target

The same result was observed concerning transmutation efficiencies. As examples, <sup>129</sup>I and <sup>237</sup>Np results are shown in figs 8a and 8b. Some of <sup>239</sup>Pu and <sup>139</sup>La results are given in refs [6, 7]. The results of figs 8a and 8b were obtained by Pb target irradiated with protons. Transmutation efficiencies are presented by the B-value which is defined as the number of exited nuclei per g of irradiated sample and per incoming proton.

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Figure 8. B-values of <sup>238</sup>Np and <sup>130</sup>I as a function of the proton beam energy.

#### 3.6 Transmutation Efficiency

The experimental set up containing paraffin moderator is a unique set up which was studied in detail. It is interesting to show the transmutation rates of such a set up because they have used as a guide for further calculations and experiments. A representative B-value for proton beams and Pb target is given in table 1 [21] assuming a 10 mA beam in order to be compared with calculation and data disposed in literature. Account taken the target mass of the transmutation set up in our experiments (about 12 Kg of Pb) we can conclude that transmutation efficiency of the specific set up is high. Increasing target dimensions however more neutrons were produced. However there are limitations in the target growth as it was calculated but also verified experimentally. This means that almost saturation in neutron production is reached in the target diameter as well as in length [18, 19].

Table 1. Transmutation efficiency		
Nuclear reaction	<b>B</b> (Transmutation)	T (mg product)/ (1 g target )
<sup>239</sup> Pu • fission (destruction of Pu )	$3.2 \ 10^{-3}$	201 mg/month
$^{238}$ U • $^{239}$ Np (breeding of Pu)	5.5 10-5	3.3 mg/month
$^{237}$ Np • $^{238}$ Np (destruction of Np)	$3.3 \ 10^{-4}$	21 mg/month
<sup>139</sup> La • <sup>140</sup> La (practical sensor)	$1.1  10^{-4}$	4 mg/month
$^{129}$ I • $^{130}$ I (destruction of I)	8.1 10 <sup>-5</sup>	3 mg/month
<sup>239</sup> Pu • fission	$5.0 \ 10^{-3}$	-
(with SSNTD in CERN, using µg Pu)		
$^{129}$ I • $^{130}$ I( radiochemistry in CERN,	$5.0 \ 10^{-4}$	-
64.7 mg <sup>129</sup> I		

# 4. Conclusions

The knowledge of the neutron yield in spallation process and the understanding of the behaviour of these neutrons in a sub-critical assembly as accelerator driven systems are the most important and determining factors in the design and operation of these systems. Spatial and energy distribution plays a critical role for production and destruction of particular isotopes and finally in transmutation process. In this study the target was surrounded by a moderator in order to shift the hard neutron spectrum to lower energies because the neutron energy region of resonances is of main importance in transmutation process.

The experience collected from our experiments in Dubna during last ten years can be summarised to the spatial neutron production as function of the target mass and projectile mass and energy. Target dimensions are also of importance for transmutation experiments but we have use information coming from previous or parallel experiments [18-20]. Although the moderator complicates the comparison with nuclear data taken with a single Pb target, the results show that neutron production measured at the moderator surface follows the reaction cross section concerning the target and the projectile masses. Indeed heavier targets give more neutrons. Also, a larger amount of neutrons escapes the moderator surface as the projectile masse increase. Concerning the projectile energy the higher neutron production with increasing energy is attributed to the higher energy of secondary particles produced in the target because reaction cross section is independent on

projectile energy at these energies. However higher order reactions can also transfer energy in the target and then produce some reactions but their contribution is of less importance. The role of higher order reactions in the target can be considered at higher projectile energies than the energies at which these experiments were performed.

The spatial neutron distribution reflects the processes taken place on the target and presents a maximum at about one mean free path indicating the competition between particle production and beam attenuation. Nevertheless the fluctuation maximum to minimum which was observed at the end of the target (and of the moderator) permits to use the entire moderator surface for transmutation.

## 5. Perspectives

The experience accumulated from transmutation experiments in Dubna served to prepare new experiments. Already from 2000 a new set up consisting from Pb/U target was developed. The design was supported by calculation based on the idea that although U target has a slightly higher spallation cross section, secondary reactions in the surrounding Pb are very few. In contrast when the target is Pb, secondary reactions are more probable in U-Blanket. The produced neutron and proton spectra by the target for both targets are almost the same. Especially a great part of the neutron spectrum contains neutrons between 1-100 MeV. These neutrons can induce fission reactions when the surrounding material is U while is not probable in Pb with low fission cross section at these neutron energies. In addition, most of the secondary protons having energies in the region of 40-60 MeV (and less up to 100 MeV) are able to induce fissions in U, with a cross section of about 0.6 barns while fissions in Pb have too small cross section. So the design of a new set up contains a central Pb core as in previous experiments but 50 cm length. The Pb target is surrounded by m<sup>at</sup>U rods (the U blanket), fig. 9. The set up is very efficient to fast neutron production [22] and favourites (n,xn) and (n,f) reactions.

Another set up is now under construction in continuation of the idea of the fast neutron thermalization. The Pb target used in previous experiments will be placed in graphite moderator of 120x120x60 cm. In various distances from the source (the target) different neutron spectra can be obtained giving different transmutation efficiencies in different places inside target. Calculations by Fluka and MCNPX codes have completed and first run with proton beam is expected at the beginning of 2006.



Figure 9. A new set up consisting from Pb target with U blanket.

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