PERGAMON

Available online at www.sciencedirect.com
SCIENCE DIRECT®

Applied Radiation and Isotopes 58 (2003) 169–175

Applied Radiation and Isotopes

www.elsevier.com/locate/apradiso

Application of activation methods on the Dubna experimental transmutation set-ups.

S. Stoulos[a,*], M. Fragopoulou[a], J.C. Adloff[b], M. Debeauvais[b], R. Brandt[c], W. Westmeier[c], M. Krivopustov[d], A. Sosnin[d], C. Papastefanou[a], M. Zamani[a], M. Manolopoulou[a]

[a] *Aristoitle University of Thessaloniki, Physics Deptartment, Thessaloniki 54124, Greece*
[b] *Institut de Recherche Subatomique, F-67037 Strasbourg, France*
[c] *Philipps Universitat, D-35032 Marburg, Germany*
[d] *Joint Institut for Nuclear Research, 141980 Dubna, Russia*

Received 21 October 2002; accepted 8 November 2002

Abstract

High spallation neutron fluxes were produced by irradiating massive heavy targets with proton beams in the GeV range. The experiments were performed at the Dubna High Energy Laboratory using the nuclotron accelerator. Two different experimental set-ups were used to produce neutron spectra convenient for transmutation of radioactive waste by (n,x) reactions. By a theoretical analysis neutron spectra can be reproduced from activation measurements. Thermal–epithermal and fast-super-fast neutron fluxes were estimated using the ^{197}Au, ^{238}U (n,γ) and (n,2n) reactions, respectively. Depleted uranium transmutation rates were also studied in both experiments.

Keywords: Neutron spectra; Spallation sources; Transmutation; Activation analysis

1. Introduction

In the framework of the international concern about long-lived radioactive wastes, many experiments were carried out using proton beams at GeV energies. By these experiments, intense neutron fluxes can be produced through spallation reactions on a solid target, in order to study the effectiveness of a sub-critical Accelerator-Driven System (ADS) to transmute long-lived radioactive wastes by neutron capture or fission (Carminati et al., 1993; Andriamonje et al., 1995; Wan et al., 1998). In both cases it is important to optimize the efficiency of the neutron reaction by maximizing each of the factors affecting the specific neutron reaction rate $R_{\mathrm{n-react}} = \phi\ (\mathring{A})\sigma(\mathring{A})\,\mathrm{d}E$.

To optimize the neutron flux [$\phi(E)$] a lead target must be chosen because neutrons in the lead have a small average lethargy $\xi \approx 0.01$, a high and nearly energy-independent elastic scattering cross-section and very high transparency for energies below 1 keV (Byrne, 1995). The number of neutrons produced increases with the target thickness and reaches saturation after $\sim$10 cm of lead target for proton beams in the GeV range, with an energy cost of production around 20–25 MeV/n (Carminati et al., 1993). Fast neutrons produced by spallation, after moderation by (n,xn) and (n,n′) reactions reach the intermediate energy range and finally having small iso-lethargic steps enter in the resonance energy range. Although this energy cascade they can transmute long lived, radioactive elements started with

*Corresponding author. Tel.: +30-31-099-8202; fax: +30310998058.
E-mail address: stoulos@auth.gr (S. Stoulos).

0969-8043/03/$ - see front matter © 2003 Elsevier Science Ltd. All rights reserved.
doi:10.1016/S0969-8043(02)00312-3

(n,fission) and (n,xn) reactions and closed with (n,γ) reactions. Additional moderators can be used for further slowing of neutrons to the epithermal and thermal energy ranges in order to obtain higher capture cross-section for some elements.

2. Experimental

Two different experimental set-ups were used. In both of them the main spallation target was a lead cylinder covered with a paraffin moderator, in the first experiment, and by a four-section U-blanket, in the second (Figs. 1a and b) (Wan et al., 1998; Krivopustov et al., 2001). Irradiations were made at the nuclotron accelerator in the High Energy Laboratory,at JINR, Russia. The first experiment, using the Pb(Paraffin) target irradiated by a proton beam of 1 GeV with total fluence of $7.6(\pm 0.3)\times 10^{12}$ protons, took place in November 2001. The second one, using the Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV, with total fluence $1.1(\pm 0.1)\times 10^{13}$ protons, took place in December 2001.

In both experiments two general types of measurements were performed:

(A) Neutron fluence measurements with several supplementary techniques such as solid state nuclear track detectors (CR-39) as particle and fission detectors (Adloff et al., 1999) and threshold activation detectors (Au, La, Co, Bi, etc.) for monitoring the γ-rays emitted by any (n,x) reaction (Zhuk et al., 1999).

(B) Transmutation rate measurements for several long-lived radioactive wastes (^{129}I, ^{237}Np, ^{239}Pu) through any (n,x) reaction (Wan et al., 2001a,b; Adam et al., 2002).

In the second experiment, any energy production through the transmutation process was studied using

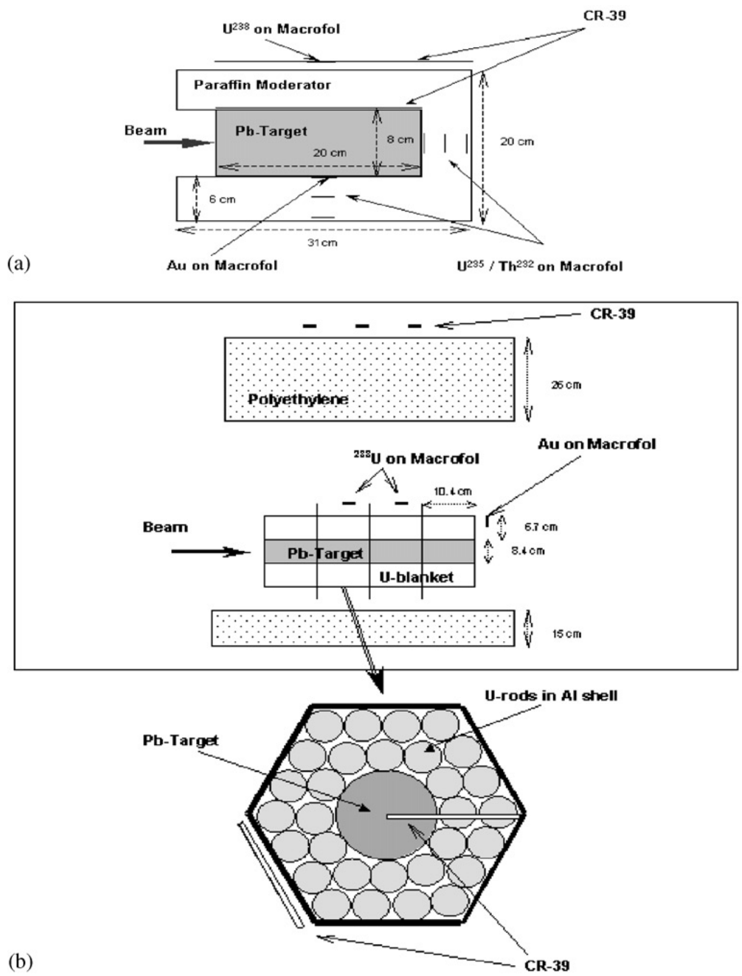

Fig. 1. (a) Pb(Paraffin) target set-up for the first experiment. (b) Pb(U-blanket) target set-up for the second experiment.

micro-thermocouples, semiconductor thermoresistors and methods based on definition of the absolute number of uranium fission events using SSNTD or γ-spectrometry (Wan et al., 1998; Krivopustov et al., 2001).

3. Results and discussion

Our contribution to these experiments was to measure secondary neutron fluence using SSNTDs (as particle and fission detectors) and activation detectors. The target set-ups for these measurements are shown in Fig. 1a and b. In this study only the results from the activation detectors are presented. In addition, from these measurements, the breeding rate of a long-lived radioactive waste such as depleted uranium was determined during these transmutation experiments.

Two types of activation detector were used, i.e. depleted U ($^{235}U/^{238}U = 0.18 \pm 0.01\%$) and natural Au. One sample of U ($m = 5.14 \pm 0.10$ mgr) was irradiated on the top of the Pb(Paraffin) target. Two samples of U ($m_1 = 5.33 \pm 0.14$ mgr, $m_2 = 14.37 \pm 0.27$ mgr) were irradiated on the top of the second and third section of the U-blanket and one of natural Au ($m = 4.67 \pm 0.07$ mgr) on the downstream end of the Pb(four-section U-blanket) target.

After irradiation the samples were measured several times over 1-month period using a low-level γ-ray spectrometry system, consisting of an HPGe coaxial detector (Tennelec model CPVDS30-35190) with 42% efficiency, 2.2 keV resolution at 1.33 MeV photons (^{60}Co), shielded by 4″Pb, 1 mm Cd and 1 mm Cu and a spectroscopy amplifier (Tennelec model TC244 with pile-up rejector). The following reactions were detected:

$$^{238}U(n,\gamma) \rightarrow {}^{239}U \xrightarrow{(\beta-,23.5\ \text{min})} {}^{239}Np \xrightarrow{(\beta-,2.355\ \text{d},E_\gamma=228,278\ \text{keV})} {}^{239}Pu\ldots\ldots$$

$$^{238}U(n,2n) \rightarrow {}^{237}U \xrightarrow{(\beta-,6.75\ \text{d},E_\gamma=208\ \text{keV})} {}^{237}Np\ldots\ldots$$

$$^{197}Au(n,\gamma) \rightarrow {}^{198}Au \xrightarrow{(\beta-,2.69\ \text{d},E_\gamma=412\ \text{keV})} {}^{198}Hg(\text{stable})$$

$$^{197}Au(n,2n) \rightarrow {}^{196}Au \xrightarrow{(6.18\ \text{d},E_\gamma=356\ \text{keV})} \begin{array}{l} EC(92.5\%) \rightarrow {}^{196}Pt(\text{stable}) \\ \beta-(7.5\%) \rightarrow {}^{196}Hg(\text{stable}) \end{array}$$

The total experimental neutron fluence $F_n(E)_{exp}$ (neutrons cm^{-2} per proton) was estimated using the formula

$$F_n(E)_{exp} = \frac{N_{produced}}{N_{irradiated} F_p \sigma_{eff}}.$$

$N_{produced}$ is the total number of nuclei produced, $N_{irradiated}$ the total number of irradiated nuclei, F_p the proton beam fluence and σ_{eff} the effective cross-section of each (n,x) reaction for the corresponding energy to the experimental neutron fluence.

The calculations of the total number of produced nuclei were determined by using the decay curve of selected γ-rays emitted by them, after the essential summation correction factor had been calculated for the sample surface geometry ($\varnothing$: 2.2 cm).

The effective cross-section σ_{eff}(b) was estimated as follows:

$$\sigma_{eff}(n,x) = \frac{\int_{E_1}^{E_2} \varphi_n(E)\sigma_{(n,x)}(E)\,dE}{\int_{E_1}^{E_2} \varphi_n(E)\,dE}$$

$\phi_n(E)$ is the energy-dependent neutron fluence (dF/dE) and $\sigma_{(n,x)}$(E) is the energy-dependent cross-section. For this estimation it is necessary to know at least the shape of the neutron spectrum, so in both experimental set-ups the neutron fluence was determined theoretically using the high-energy transport code DCM-DEM developed in Dubna. These calculations of the secondary neutron fluence referred at the point where the ^{238}U (on Macrofol) detectors were placed. Using the calculated fluence (F_{theor}) a fitting procedure was applied at the energy-dependent neutron fluence (dF/dE). The energy-dependent neutron fluence of the Pb(Paraffin) target followed an hyperbolic background function for the energy range from 10^{-8} to 35 MeV (Fig. 2). Three peaks appeared, the first in the thermal–epithermal area (centroid at 0.16 eV) and the others in the fast–superfast area (centroid at 1.7 and 12.3 MeV). The energy–dependent neutron fluence of the Pb(four-section U-blanket) target followed the same pattern, with a hyperbolic background function (Fig. 3), in which two peaks appeared, the first in the thermal–epithermal area (centroid at 0.06 eV) and the other in the intermediate–fast area (centroid at 0.5 MeV). Another peak, in the fast–superfast area (centroid at 12.5 MeV) also appeared.

A similar procedure was applied to the data for the energy-dependent cross-section obtained by the ENDF/B-VI (300 K) library by fitting an $E^{-1/2}$ function for energies up to 1 eV, a Gaussian–Lorenztian function on each resonance peak and a Chester–Cram Peak function for the (n,2n) reaction. In the resonance area of the (n,γ) reaction only those which appeared from 1 to 100–200 eV have a strong effect on the estimation of the σ_{eff} of the nuclides examined.

The results of the effective cross-section measurements for each reaction according to the corresponding energy range for both experimental set-ups are presented in Table 1. The respective total experimental fluence of neutron $F_n(E)_{exp}$ and the number of neutrons produced per primary proton are presented in Tables 2 and 3 for each experimental set-up, respectively.

The results of the total experimental fluence measurements of neutron $F_n(E)_{exp}$ for energies up to 200 eV produced from a Pb(Paraffin) target irradiated by a proton beam of 1 GeV (Table 2) indicate that at the middle of the moderator cylinder (position A: at a distance of ~15 cm from upstream perpendicular to the beam axis), the number of thermal–epithermal neutrons is 3 (±1) times higher than the number of fast–superfast neutrons (energy range 6–35 MeV).

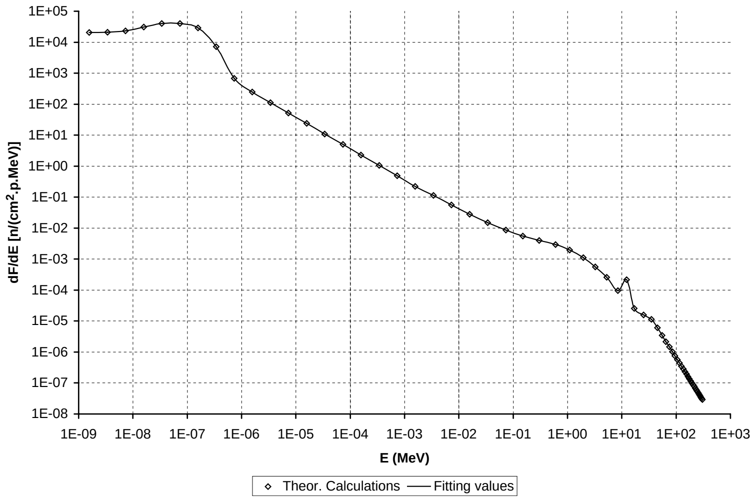

Fig. 2. Theoretical calculation of the energy-dependent neutron fluence using the Dubna Cascade Model for the Pb(Paraffin) target irradiated by a proton beam of 1 GeV.

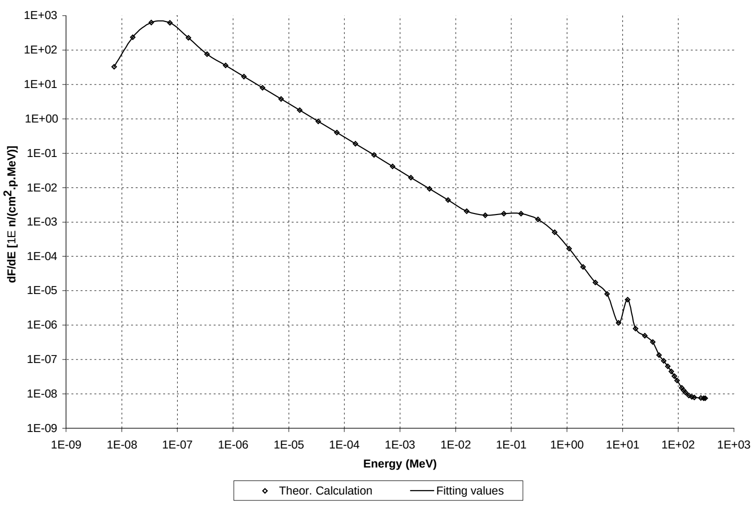

Fig. 3. Theoretical calculation of the energy-dependent neutron fluence using the Dubna Cascade Model for the Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV.

Table 1
Effective cross-section of each reaction according to the corresponding energy range for both experimental set-ups [Pb(Paraffin) target irradiated by a proton beam of 1 GeV and Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV]

Reaction	Energy range	Target	Effective cross-section σ_{eff} (b)
$^{238}U(n, \gamma)\ldots \rightarrow {}^{239}Np\ldots$	Up to 150 eV	Pb(Paraffin)	9.5 ± 1.8
		Pb(four-section U-blanket)	29.3 ± 4.0
$^{238}U(n, 2n) \rightarrow {}^{237}U$	6–35 MeV	Pb(Paraffin)	1.0 ± 0.1
		Pb(four-section U-blanket)	0.9 ± 0.1
$^{197}Au(n, \gamma) \rightarrow {}^{198}Hg$	Up to 100 eV	Pb(Paraffin)	98 ± 18
		Pb(four-section U-blanket)	193 ± 27
$^{197}Au(n, 2n) \rightarrow {}^{196}Pt$	6–35i MeV	Pb(Paraffin)	1.4 ± 0.1
		Pb(four-section U-blanket)	1.4 ± 0.1

Table 2
Total experimental fluence of neutron $F_n(E)_{exp}$ and the number of produced n per primary p for a Pb(Paraffin) target irradiated by a proton beam of 1 GeV

Position along the target at angle 90° to the beam direction (cm)	Energy range	Experimental neutron fluence $F_n(E)_{exp}$ ($\times 10^{-3}$ n cm^{-2} per primary p)	Produced neutrons per primary proton (n/p)
0 (centre)	Up to 150 eV	4.8 ± 0.9	9.4 ± 1.7
	6–35 MeV	1.6 ± 0.4	3.1 ± 0.8

Table 3
Experimental fluence of neutron $F_n(E)_{exp}$ and the number of produced n per primary p for a Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV

Position along the target (angle to the beam direction)	Energy range	Total experimental neutron fluence $F_n(E)_{exp}$ ($\times 10^{-3}$ n cm^{-2} per primary p)	Produced neutrons per primary proton (n/p)
Second section of U-blanket (110°)	Up to 150 eV	1.7 ± 0.3	7.8 ± 1.2
	6–35 MeV	1.3 ± 0.1	5.9 ± 0.6
Third section of U-blanket (110°)	Up to 150 eV	1.6 ± 0.3	7.3 ± 1.1
	6–35 MeV	1.3 ± 0.1	5.9 ± 0.6
Downstream (120°)	Up to 100 eV	1.1 ± 0.2	4.8 ± 0.7
	6–35 MeV	0.7 ± 0.1	3.1 ± 0.3

The same results for a Pb(4-section U-blanket) target irradiated by a proton beam of 1.5 GeV (Table 3.) indicate that the distribution of n produced per primary p is uniform at the middle area of the U-blanket (position B: at a distance of between 20 and 30 cm from upstream perpendicular to the beam axis) and then drops at the downstream position for both thermal–epithermal and fast–superfast neutrons. In the middle area of the U-blanket, the number of thermal–epithermal neutrons (energy up to 150 eV) is approximately the same as the number of fast–superfast neutrons (energy range 6–35 MeV).

The total experimental fluence of thermal–epithermal neutrons at position A of the Pb(Paraffin) target irradiated by a proton beam of 1 GeV is 3 (± 1) times higher than the total experimental fluence of thermal–epithermal neutrons at position B of the Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV.In contrast, the total experimental fluence of fast–superfast neutrons is approximately the same for the exact positions A and B.

Finally, the effectiveness of these experiments for the transmutation process of a long-lived radioactive waste such as depleted uranium was determined in terms of the breeding rate B_{exp} defined as follows (Wan et al., 2001a,b):

$$B_{exp}(A^Z) = \frac{N_{produced}}{m_{irradiated} F_p}.$$

Table 4
Breeding rate for the produced ^{239}Np [through the ^{238}U (n,γ) reaction] and for the produced ^{237}U [through the ^{238}U (n,2n) reaction] for both experimental set-ups

Experimental set-up	Position along the target	Reaction	Breeding rate B_{exp} ($\times 10^{-4}$)
Pb(Paraffin) target irradiated by a proton beam of 1 GeV	Centered at angle 90° to the beam direction	^{238}U(n, γ)… → ^{239}Np…	1.20 ± 0.04
		^{238}U(n, 2n) → ^{237}U	0.04 ± 0.01
Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV	Second section of U-blanket (at angle 110° to the beam direction)	^{238}U(n, γ)… → ^{239}Np…	1.3 ± 0.1
		^{238}U(n, 2n) → ^{237}U	0.03 ± 0.01
	Third section of U-blanket (at angle 110° to the beam direction)	^{238}U(n, γ)… → ^{239}Np…	1.2 ± 0.1
		^{238}U(n, 2n) → ^{237}U	0.030 ± 0.004

$N_{produced}$ is the total number of nuclei produced with A^Z, $m_{irradiated}$ the mass of irradiated target (gr), F_p the proton beam fluence. The breeding rate for the ^{239}Np produced [through the ^{238}U (n,γ) reaction] and for the ^{237}U produced [through the ^{238}U (n,2n) reaction] is presented in Table 4 for both experimental set-ups.

The depleted uranium transmutation process at both experimental set-ups originates mainly from the ^{238}U(n,γ) reaction. The breeding rate seems to be identical for the exact positions A and B at each experimental set-up. This is due to the effective cross-section (N_{eff}) of each experimental set-up. According to Table 1 the N_{eff} of ^{238}U(n,γ) reaction at position B of the Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV is 3 ($\pm$1) times higher than the respective σ_{eff} at position A of the Pb(Paraffin) target irradiated by a proton beam of 1 GeV, as opposed to the σ_{eff} of ^{238}U(n,2n) reaction which is identical for the exact positions A and B at each experimental set-up. Correlating the resulting total experimental fluence for thermal–epithermal and fast–superfast neutrons with the respective calculated σ_{eff} values of the ^{238}U(n,3) and ^{238}U(n,2n) reactions, the predicted specific neutron reaction rate $R_{n-react} = \phi\ (\mathring{A})\sigma(\mathring{A})\,dE$ and therefore the measured breeding rate for each reaction has to be identical for the exact positions A and B at each experimental set-up. The determined breeding rates (Table 4) are in a good agreement with the results from previous experiments on the same targets with the same proton energy (Wan et al., 1998; Krivopustov et al., 2002).

4. Conclusion

To optimize the specific neutron reaction rate by $R_{n-react} = \phi\ (\mathring{A})\sigma(\mathring{A})\,dE$ during any transmutation process of long-lived radioactive wastes using spallation reactions, an appropriate target and proton beam must be selected to obtain the maximum neutron fluence for the energy range where the transmutated radioactive wastes appear to have the maximum cross-section for a specific (n,x) reaction. An experimental set-up, like the Pb(Paraffin) target irradiated by a proton beam of 1 GeV, is very effective for the transmutation of radioactive waste with a strong capture or fission cross-section at the thermal–epithermal energy range and weak resonance. At variance, an experimental set-up, like Pb(four-section U-blanket) target irradiated by a proton beam of 1.5 GeV, is very effective for transmutation of a radioactive waste with strong capture or fission resonance and a normal to low cross-section at the thermal–epithermal energy range. In order to maximize the contribution of the (n,xn) reactions to the total transmutation rate a proper experimental set-up must be used to produce a total fast–superfast neutrons fluence (energy > 6 MeV) higher than the fluence of neutrons with energy < 10 keV by one or two orders of magnitude. In addition, at these transmutation experiments the total neutron fluence in the specific energy range must be determined using several supplementary techniques (^{3}He, ^{4}He proportional counters or Bonner spheres) to confirm any theoretical calculations. The confirmation is necessary for correct estimation of the effective cross-section used in the evaluation of the experimental results.

References

Adam, J., et al., 2002. Transmutation of ^{239}Pu and other nuclides using spallation neutrons produced by relativistic protons reacting with massive U- and Pb targets. Radiochim. Acta 90, 431.

Adloff, J.C., et al., 1999. Secondary neutron production from thick Pb target by light particle irradiation. Radiat. Meas. 31, 551.

Andriamonje, S., et al., 1995. Experimental determination of the energy generated in nuclear cascades by a high energy beam. Phys. Lett. B 348, 697.

Byrne, J., 1995. Neutrons, Nuclei and Matter. Institute of Physics Publishing, Bristol and Philadelphia.

Carminati, F., et al., 1993. An Energy Amplifier for cleaner and inexhaustible nuclear Energy production driven by a particle beam accelerator. CERN, Geneva, Print CERN/AT/93-47 (ET).

Krivopustov, M.I., et al., 2001. Experimental studies of electronuclear method of energy production and transmutation of radioactive wastes using relativistic beams from JINR(Dubna) syncrophasotron/nuclotron. In: Baldin, A.M., Burov, V.V., Malakhof, A.I., (Eds.), Proceedings of the XV International Seminar on High Energy Physics Problems, "Relativistic Nuclear Physics and Quantum Chromodynamics" JINR (Dubna), Russia, Vol. II, p. 3.

Krivopustov, M.I., et al., 2002. First experiments with a large uranium blanket within the installation "energy plus transmutation" exposured to 1.5 GeV protons. Kerntechik, in press.

Wan, J.S., et al., 1998. Transmutation of radioactive waste with the help of relativistic heavy ions. Kerntechnik 63, 167.

Wan, J.S., et al., 2001a. Transmutation of ^{129}I and ^{237}Np using spallation neutrons produced by 1.5, 3.7 and 7.4 Gev protons. Nucl. Instr. and Meth. A 463, 634.

Wan, J.S., et al., 2001b. Transmutation of ^{239}Pu with spallation neutrons. J. Radioanal. and Nucl. Chem. 247, 151.

Zhuk, I., et al., 1999. Determination of spatial and energy distributions of neutrons in experiments on transmutation of radioactive waste using relativistic protons. Radiat. Meas. 31, 515.