

**ON THE MRTI EXPERIENCE IN THE
INTERMEDIATE ENERGY NUCLEAR
PHYSICS STUDIES, PERFORMED IN
EXPERIMENTAL VALIDATION OF THE
ACCELERATOR-BASED LARGE-SCALE
NEUTRON PRODUCTION**

R.G. Vassil'kov
(MRI, Moscow)

ON THE MRTI EXPERIENCE IN THE INTERMEDIATE ENERGY NUCLEAR PHYSICS STUDIES, PERFORMED IN EXPERIMENTAL VALIDATION OF THE ACCELERATOR-BASED LARGE-SCALE NEUTRON PRODUCTION

RG. VASSIL'KOV
Moscow *Radiotechnical Institute*
of the *Russia Academy of Sciences*
Warshavskoye chaussee 132, 113519 Moscow, Russia

1. As a matter of **fact**, in the former USSR studies of scientific and technical feasibility of a practical use of the **spallation** reaction neutron potential for the NFC purposes (mainly, novel technologies of producing the **fissile** materials and high-flux steady-state thermal neutron fields) have been initiated in the fall of 1948. This approach required, in particular, to develop a high-current ion accelerator technology and to establish, essentially, a new direction in the intermediate nuclear physics and accumulate the data needed for validating, even though conceptually, nuclear-technology facilities based on such **proton/deuteron** medium-energy accelerators.

As for the accelerator physics field, it had to advance in investigating the very possibility to build a machine providing average ion beam powers of the order of 10-100 MW. Development of a **proton/deuteron** linear accelerator (500 MeV, 100 mA) began at Institute for Chemical Physics of Academy of Sciences of the USSR in 1948-49, then the works have been transferred and concentrated to a place of **Oural** region which one names now as Chelyabinsk-50. The project was not due to the EN problem, and in 1963 it was stopped. A **perfectly** operating injector **PT-500** (500 keV, 150 mA dc) remained, power RF **triode** generators were developed, elements of the **drift tube linac** existed, the proton/deuteron beam dynamics in such a **linac** was developed.

In searching for possible new applications for the suspended **linac** project a rather limited nuclear physics program was started. In studying the problem of an interaction of a light ion beam ($\sim 10^2$ - 10^3 MeV/ion charge unit) with matter, at **ICP-RTI-MRTI** the detailed **neutronics** measurement on primary **bulky** targets and primitive blankets (metal natural/**enriched** U, Pb, Be, reactor grade graphite and their **combinations**), bombarded with accelerated **p, d, ^3He , ^{12}C** were carried out. Also experiments were **being** done on thin targets of various elements (from Be to U).

Our activity aimed, in particular, at elucidating the principal properties of a **spallation** neutron sources and a possible role of the accelerator-based neutron production in the evolving nuclear power industry. It means a large-scale realization of nuclear transmutations : breeding of fuel, incineration of a part of radioactive waste due to nuclear power generation, **tritium** production [1-3].

More specifically, early in the sixties we interested in some experimental nuclear **data**, lacking or unreliable, mainly of integral type, which enabled to judge about **target/surrounding** neutron characteristics : neutron yields and spectra, neutron multiplicities per nuclear absorption of ion, fission cross-section of heavy nuclei by **spallation** neutrons, yields of **n, γ** and **n,f-reactions** inside and around heavy element targets, irradiated with various light ions accelerated to energies **0.3+8 GeV**, as well as to use these data for estimating performances of the hypothetical EN breeder-transmuter and optimizing **energy/current** of its accelerator.

2. First of all, neutron multiplicities **ν** per interaction of high-energy proton (**$E_p = 0.4+3.2$ GeV**) and **deuteron** (**$E_d = 1.48$ and **7,3 GeV**) with nuclei (15 elements in all : from Be up to U) were measured (some examples are presented in fig. 1). The moderation **technique** was **used**, so that neutrons of energies ≤ 15 MeV were **detected**, but later in experiments at the **Synchrophasotron** the time-of-flight (**TOF**) **technique** was supplemented and **$^6\text{Li}/^{12}\text{C}$** ions as projectiles were added. These experimental results were **necessary** for **developing/testing** the **cascade-evaporational** model of **spallation reaction** and for building/testing computer codes [4], which gave integral characteristics of the **high-energy** radiation transport in materials of **quasi-infinite** target assemblies.**

3. From the very beginning of our activity we were aware that lead (or Pb-Bi **eutectic**) was **very** probable substance for the primary **target**, albeit not the only. Therefore neutron emission from thick Pb targets (cylinders **0 10+26x60** 76 cm, **bars/slabs** 16x16x60 cm³) [5] has been studied by hvo quite different experimental methods (neutron moderation and threshold solid state nuclear track **detectors, SSNTD**) ; sometimes, in studying shortened targets, the TOF technique was applied for the sake of check purposes. Measurements of the total yields and **energy/space** distributions of neutrons were done in a broad interval of kinetic energies of **p, d, ^3He , ^4He , ^{12}C** (fig. 2).

The data enabled, in particular, to establish a type of accelerated **ion**, providing the maximum neutron yield per unit of spent energy, determine the ion initial kinetic energy, up to which it would be expediently to accelerate the ion (minimization of expenditures of the primary ion kinetic energy for releasing one neutron) - with accuracy to **E-dependence** of RF power.

4. By the **SSNTD** technique, supplemented in some cases with the TOF **method**, the integral neutron spectra over the target surface (and over various angles) have been measured, thereby **fluences** of neutrons in 8 energy groups, including the tens-hundreds **MeV** range, and a fraction of neutrons was got in the **full**

spallation neutron spectrum, of which energies > 15 MeV. Some examples of the neutron data for Pb target 0 20x60 cm are presented in fig. 3.

5. On the same bulky primary Pb targets the neutron multiplication was studied in general outline in several primitive blankets : the homogeneous ones assembled of continuous metal uranium (both natural and depleted, **with/without** Pb core inside U) ; the heterogeneous ones made of **Pb-Be-graphite/Pb-Be-water** (with neutron traps in some geometries).

In searching for novel approaches to generate steady-state high-intensity thermal neutron fluxes, which are **unachievable** even for the high-flux fission reactors, specially designed for this aim ($\geq 7 \cdot 10^{15}/\text{cm}^2 \cdot \text{c}$), we carried out a series of experiments on the total absorption assemblies composed on the Pb primary target ($16 \times 16 \times 60 \text{ cm}^3$) - a converter of proton beam power into neutrons in the **MeV** range, a Be blanket of **4+20** cm thick ($n, 2n$ reaction multiplier) and a reactor grade graphite moderator/reflector 80 cm thick [7] (fig. 4). This and all other assemblies of such a kind were driven by 0.66 **GeV** proton beam (or lower) of the **JINR** Synchrocyclotron at **Dubna** in 1966-1969. Now this assembly maybe of interest again.

In the first phase of the experiments, an increase of neutrons in the target assembly **Pb+Be** (as compared with Pb only) was determined by the moderation technique, using as thermal neutron detector 5 % solution of **MnSO₄** in water. The number of **spallation** neutrons becomes 1.82 ± 0.20 as much when Be wall is 16 cm thick; the thickness of Be wall 20 cm gives the neutron rise only 3-4 % more. Such a neutron source (**Pb** $16 \times 16 \times 60 \text{ cm}^3$ + Be wall 16 cm thick) provides the thermal neutron flux densities in surrounding graphite moderator (in the maximum of its distribution measured along the proton beam direction, at a distance from Be wall **1+41** cm) from $7.2 \cdot 10^{-3}$ to $3.8 \cdot 10^{-3} \cdot \text{cm}^2 \cdot \text{sec}^{-1} \cdot \text{p}^{-1}$ (± 11 %).

6. Lastly, on the total absorption metal uranium targets (3.2-3.4 ton, natural/depleted, with or without lead converter inside), irradiated by 300-660 **MeV** protons, the data have been obtained [8] concerning the yields of **²³⁹Pu** and fission energy. The data enabled to estimate performances of the EN breeding (the **Pu** production rate, energy spent for its production, accompanying release of energy) and to test computer codes describing the nuclear cascade in large masses of uranium.

7. In the **spallation** neutron **fields** around the same Pb cylinder, absorbing p/d beams of energies 1.0-3.7 GeV, the study of **transuranium actinides** fission has been started, some results (**averaged one group fission cross-sections**) have been obtained for **²³⁷Np**, **²³⁵J238U**, **²³²Th** [9] and quite recently **²⁴³Am**

8. Some our data were used at developing and testing computer code describing the neutron multiplication and balance of energy both in homogeneous **infinite/quasi-infinite** U blocks (**MRTI**, **BNL**, **JAERI**, **JINR**, **ORNL**) and in **full-scale** heterogeneous target/blankets (**BNL**, **JAERI**) ; also for validating the codes **HETC**, describing neutronics of thick Pb cylinder, irradiated with protons of energies -- **GeV** (**CRE Casaccia**, **LAHET**, **ORNL**, **KEK**); for testing the **high-energy** cascade model of **spallation** reaction (**ORNL**, **JINR**, **MRTI**). Agreement between **calculational** and measured results is moderately good, as a rule, but only when the integral values are compared. In the matter of differential values such an agreement is a rarity.

9. About two years ago, in connection with the interest to a creation of safety and cleaner nuclear power industry, to mastering the **Th-²³³U** **NFC** and more efficient reactor systems (for example, **MSR** with **Th-²³³U** fuel and with possible adding **TUA** waste) we have taken up again the assembly **Pb-Be-graphite**. But now Be neutron multiplier is substituted for a mixture **Th (80 %) + ²³⁷Np (20 %)** because the use of Be would be impractical in fields of very hard neutrons. A **behaviour** of metal **Th/salt LiF-BeF₂-ThF₄** in the graphite moderator will be investigated in details. Besides, a possibility to apply a heavier chloride salt **PbCl₂-ThCl₃-[TUA]Cl₄** as the primary target instead of Pb will be studied. **Preliminary** appraisals show, that even at moderate values of k_{eff} (0.86-0.92) current of 1 **GeV** protons can amount to 30-35 **mA**, so that one 100 MW **H⁺/H⁻ CW linac** would support 3 blankets of thermal power 1.2 GW.

Unfortunately, this activity is suspended now completely owing to lack of funding.

REFERENCES

1. Vassil'kov R.G., Goldanskii V.I., Dzhelepov V.P., Dmitrievskii V.P., *The electronuclear method of neutron generation and production of fissile materials*. Atomnaya energ., 29 (1970) 151.
2. Vassil'kov R. G., Goldanskii V. I., Orlov V. V., *On the electrical breeding*. Uspekhi physicheskikh nauk 139 (1983) 434.
3. Vassil'kov R. G., *Free neutron production with hydrogen ion accelerators for nuclear fuel cycle needs*. Muon-catalysed fusion 7(1992) 245.
4. Gelfand E.K., Man'ko B. S., Serov A.Ya., Sychev B. S., *The brief review of the nuclear physics aspects of radiation field simulation on high energy accelerators*. ABC/ATW Conference Chelyabisk-70 (1993).
5. Vassil'kov R. G., Yurevich V. I., *Neutron emission from extended Pb targets under the action of light ions in the GeV region*. Proc. of the ICANS-11 Meeting, 22-26 October, (1990) KEK, Tsukuba, Japan.
6. Nikolayev V. A., Vassil'kov R. G., Yakovlev R.M., Yurevich V.I., *Neutron production in thick lead target by 1-3.7 GeV protons and deuterons*. Proc. of the ICANS-11 Meeting, 22-26 October, (1990) KEK, Tsukuba, Japan.

7. Vassil'kov R. G., Goldanskii V. I., Kogai S. S., Pimenov B. A., Sokolov L. N., a) *Neutron yields in a system Pb-Be-H₂O bombarded by high-energy protons*. Report no 3/743 (1970) ; b) *Thermal neutron flux in a model heterogeneous assembly bombarded by protons with energies 400, 500 and 660 MeV*. Report n^o 2/0368 (1971). Institute for Chemical Physics of the USSR Academy of Sciences and Moscow Institute of Physical Engineering (to be published in Nucl. Instr. Meth.).
8. Vassil'kov R.G., Goldanskii V. I., Pimenov B. A., Pokotilovskii Yu.N., Christyakov L. V., *Neutron multiplication in uranium bombarded by protons with energies 300-660 MeV*. Atomnaya energ., 44 (1978) 329.
9. Nikolayev V.A., Vassil'kov R.G., Yakovlev R.M., Yurevich V.I., *The fission transmutation of ²³⁷Np in a field of spallation neutrons emitted from thick lead target bombarded by 1-3.7 GeV protons and deuterons*. Proc. of the Specialists' Meeting on Accelerator Based Transmutation, 24-26 March (1992), PSI, Villigen, Switzerland.

Table

$\langle \sigma_f \rangle$				
²³⁷ Np	²³⁵ U	²³⁸ U	²³² Th	
PROTON				
1.31 * 0.14	1.57 * 0.17	0.38 ± 0.04	0.14 * 0.02	mod
1.14 * 0.11	1.37 * 0.14	0.33 * 0.03	0.12 * 0.01	SSNTD
DEUTERON				
1.32 ± 0.21	1.55 ± 0.25	0.36 ± 0.06	0.14 * 0.02	mod
1.13 * 0.23	1.35 * 0.25	0.31 * 0.05	0.12 * 0.02	SSNTD
(σ_f)				
0.84	1	0.24	0.09	proton
0.84	1	0.23	0.09	deuteron

Experimental values of averaged one-group fission (transmutation) cross-sections for several key actinides (²³²Th, ²³⁵U, ²³⁸U, ²³⁷Np) [9] and ²⁴³Am. A source of spallation neutrons is Pb target 20x60 cm, irradiated by 1.0-3.7 GeV proton/deuteron beams.

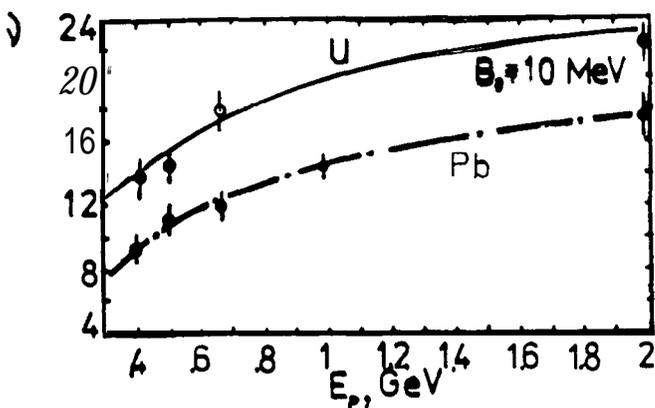


Fig. 1- Experimental data on neutron multiplicities (Y) and (E_n 15 MeV) for Pb/U nuclei absorbing protons Pb target of energy 0.4-2.0 GeV.

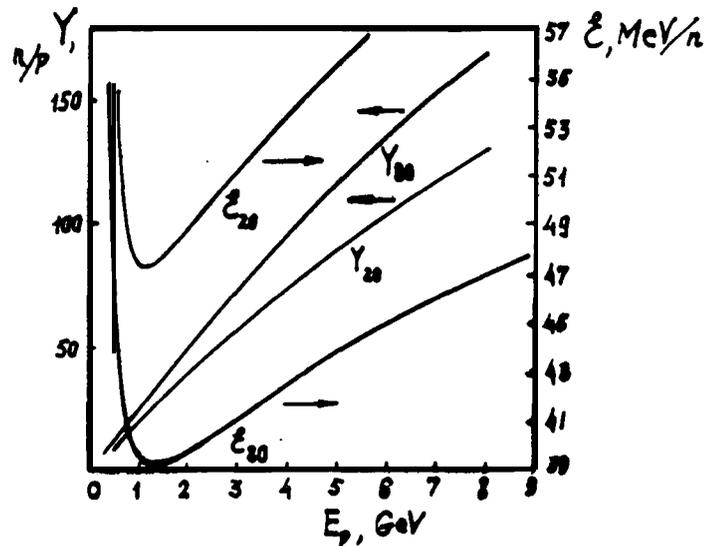


Fig. 2- E_p-dependence of neutron yield energy cost of neutron (E) released from 0 20x60 cm (experiment, moderation technique) and 0 80x100 cm (calculation) under the action of protons.

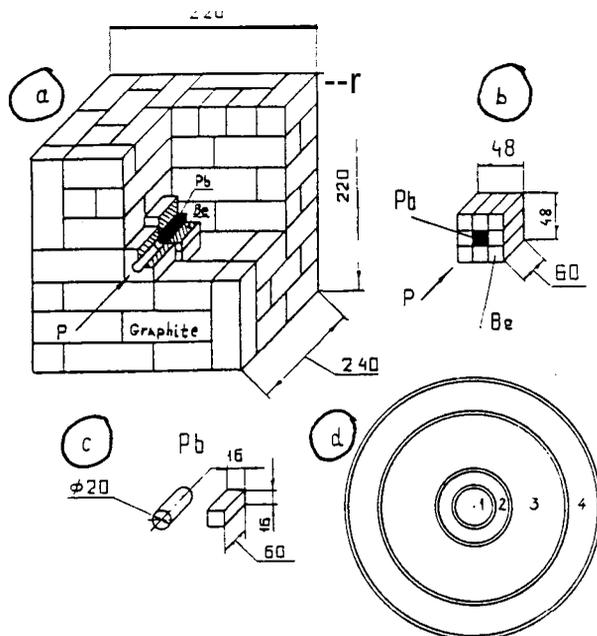
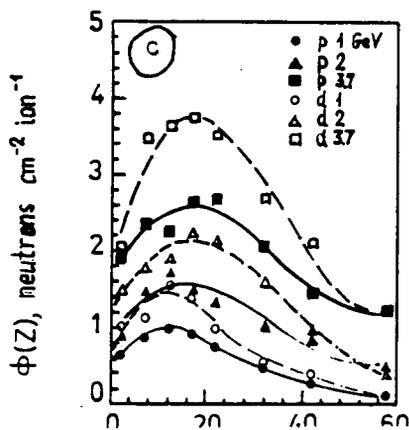
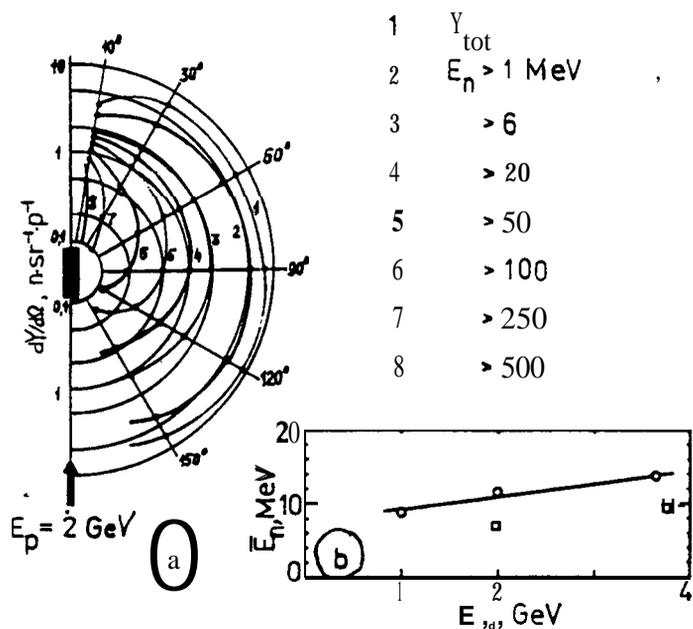


Fig. 4 - Experimental arrangement for studying multiplication of *spallation* neutrons from the Pb **primary** target in Be blanket embodied in graphite moderator-reflector [7] : a) Pb target 16x16x60 cm³ inside Be multiplying cavity, wall thickness 16 cm, both surrounded by a graphite moderator-reflector 80 cm thick ; in the assembly **thermalized** neutron flux densities (3.8-7.2) · 10⁻³/cm³·s.p. (±15%) have been obtained ; b) Pb target 16x16x60 cm³ surrounded by Be blanket-multiplier, its wall being of variable thickness or depleted U wall 8 cm thick ; c) bar Pb targets 16x16x60 cm³ and 010-26 cm ; d) a scheme of an accelerator-driven target-blanket : 1- Pb (Pb-Bi) target (a possible option is heavy chloride molten salts carrying TUA), 2- neutron multiplier (Th.80 % + TUA 20 %) 3- graphite moderator with molten salt **fuel** (1% ²³⁵U), 4- graphite reflector.

Fig. 3- The data obtained by the SSNTD technique for the same Pb target (0 20x60 cm), **bombarded** with proton/deuteron beam ($E_{p,d} = 1,0-3,7 \text{ GeV}$) [6] : a) **energy/angular** distributions of **spallation** neutrons, b) mean neutron energy versus the initial **proton/deuteron** kinetic energy, c) neutron **fluences** over the target surface.

Fig. 5- The yields of ²³⁹Pu (number of ²³⁸U(n,γ) ²³⁹U ²³⁹N_p reactions) and fission reactions ^{235,238}U(n,f) in quasi-infinite metal uranium targets (both natural and depleted, 0.36 %²³⁵U), under the bombardment with protons of energies 0.3-0.66 GeV [8].

