Neutron Source of INR RAS, the status and the further developments

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Abstract. The current status of the spallation neutron source of the Institute for Nuclear Research (INR RAS) is given shortly. Probable next steps of modernization of the neutron source and its targets to increase the neutron yield and the thermal neutron flux, by creation of new target modules and increase of reflector efficiency, are described. The proposal for targets based on ^{237}Np (motivation, characteristic property, results of preliminary researches and calculations) is discussed.

1. Linac

Regular work of the accelerator on physical and applied problems has begun in 1993. The maximum energy 502 MeV was reached in 1996 and was limited by available number of klystrons which production was temporary stopped. Because of the limited lifetime of klystrons, the available maximum energy was decreasing to 209 MeV. The manufacture of klystrons is renewed now.

Table 1. Linac parameters [1, 2]

	The design parameters	Running characteristic	Maximum reached value
Accelerated particles	P, H^-	p, H^-	p, H^-
Maximal proton energy, MeV	600	209	502 (1996 г)
The maximal average current, μA	500	150	150
Pulse current, mA	50	16	16
Pulse repetition rate up to, Hz	100	50	50
The maximal pulse duration up to, μs	100	200	200

In these conditions the accelerator mainly operates for the isotopes production, the lead slowing down neutron spectrometer, the medical complex and the source of epithermal neutrons.

The nearest aims are directed to maintenance and development of the accelerator. Increase of the pulse repetition rate up to 100 Hz is planned. It will allow to double the beam intensity ~ 300 μA. Increase of proton energy up to 500 MeV with obtaining of klystrons is planned.

2. Experimental complex

The experimental complex has the next structure:

- Complex of neutron sources;

- Thermal neutron source with TOF spectrometers
- Source of epithermal neutrons (RADEX) with TOF spectrometers
- 100 t lead slowing down neutron spectrometer. Power will be increase up to 3 kW after creation of new target with air cooling
- The free space boxing at the radiating shield designed for the second source of neutrons

- Complex of proton therapy.
- Channels for transportation of proton beams.
- Power supplies and support systems of the experimental complex (special ventilation, water supply, storage of radioactive isotopes, temporal storages of nuclear waste and irradiated structural elements of experimental complex etc.)

Simultaneous work of the different proton lines will be possible after start of the H- ions source.

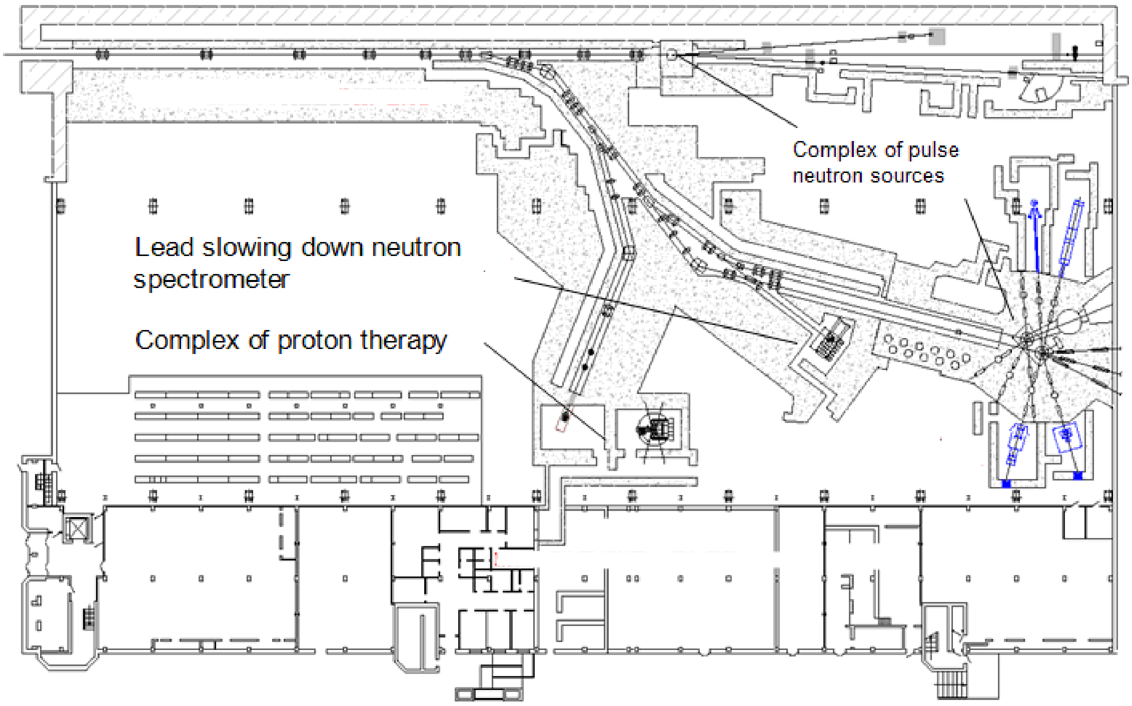


Fig. 1. Layout of the main objects of the experimental complex: beam lines, radiation shield, neutron sources, and neutron spectrometers.

3. Pulsed sources of thermal neutrons

The pulsed source of thermal neutrons is located in the first box of a radiation shield and is intended for researches in condensed matter physics [3,4]. Its scheme is presented on fig. 2. The design of a neutron source is flexible enough, allows to use modules with different targets (on the basis of tungsten and other materials) and moderators to carry out full replacement of all equipment of the central part for modernization of the source.

Within the last three years the source of neutrons is modernized according to the new requirements for radiation objects:

- Replacement of steel shot by lead fraction in constructional elements over source has been made for increase of shielding properties and compatibility of materials
- Neutron guide collimators are inserted into the shield
- Pick-and-place systems and containers are created and tested
- Works on assemblage and tests of the new target module with Be-reflectors were carried out

- Some spectrometers are created at the source
- Debugging sessions for devices and spectrometers are carried out with use of the existing target module

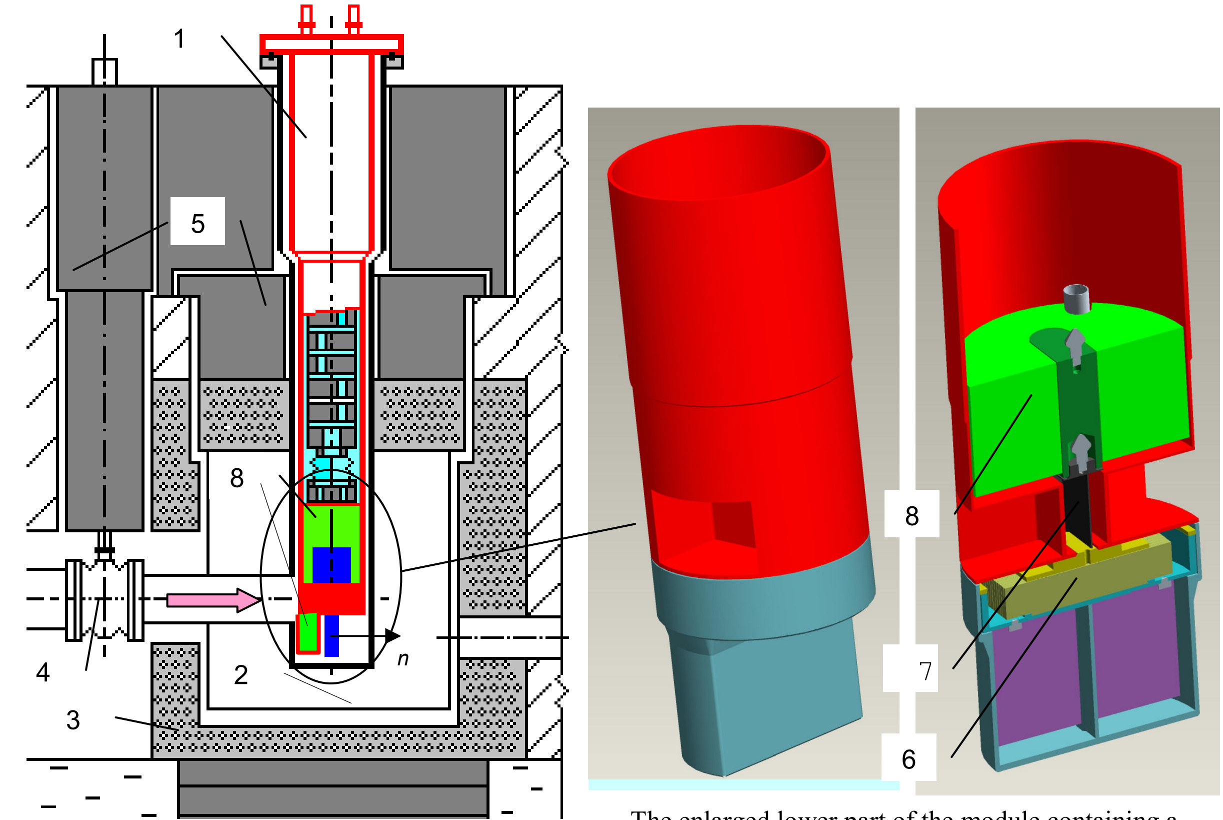
The second box of radiation shield of the neutron complex is empty now. Various possibilities of its use are studied.

4. New target module

The new target module with beryllium reflectors will allow us to increase thermal neutron flux at the upper moderator more than twice in comparison with operating module and ~ 6-8 times at lower moderator in comparison with operating module.

Substantial growth of density of the thermal flux at the lower moderator is caused by an increase of the moderator thickness from 2 to 5 cm and by effect of the beryllium reflector at the opposite moderator side [5]. Lower moderator of the new module is intended for work with thermal neutrons mainly but the epithetmal neutron flux will be increased too.

This module can be used without a storage ring effectively too. The scheme of the module with reflectors is shown on figure 3. Calculated density of the thermal neutron flux at the upper moderator at presence of the 2 cm water premoderator is given in the Table 2.



Position of a target module in the neutron source and the arrangement of basic elements

The enlarged lower part of the module containing a target, moderators and reflectors, cut of the lower part of the module.

Fig. 2. Scheme of the pulsed neutron source with new module: 1 - the ampoule (the module) with shielding plug, W – core, Be - reflector and moderators, 2 – the gas tanks, 3 - heat shield, 4 – the remote-controlled vacuum sealing, 5 - the removable steel plugs, 6 – tungsten target, 7 – moderators, 8 – beryllium reflector.

Table 2. Calculated density of the thermal neutron flux at the upper moderator

Proton energy, MeV	400	500	600
Average thermal neutron flux : **per proton** - n /($cm^2 \cdot p$) / **for current 100 μA** - n/($cm^2 \cdot s$)			
On the surface of top Moderator	$2.24 \cdot 10^{-3}$ / $1.4 \cdot 10^{12}$	$3.24 \cdot 10^{-3}$ / $2.0 \cdot 10^{12}$	$3.97 \cdot 10^{-3}$ / $2.5 \cdot 10^{12}$
At distance ~ ***10 m*** from the Surface of top moderator	$3.65 \cdot 10^{-8}$ / $2.3 \cdot 10^{7}$	$5.15 \cdot 10^{-8}$ / $3.2 \cdot 10^{7}$	$6.35 \cdot 10^{-8}$ / $3.9 \cdot 10^{7}$

After these modifications the further improvements of the moderator-reflector system are almost impossible under the given spatial module limits.

5. Development and modernization of the neutron source

Installation of the beryllium reflector in all volume of the gas tank with neutron channels inside is planned. In this case thermal neutron flux at surfaces of moderators will increase additionally.

Creation of targets with increased neutron yield per proton is considered. Possibility of creation of the target on a basis ^{237}Np is studied.

Creation of a neutron source in the second free boxing of the shield is planned.

6. The possibility of using neptunium for targets of high-current proton accelerators

^{237}Np has rather low threshold of fission reaction (~ 0.5 MeV) in comparison with ^{238}U (~ 1 MeV), therefore the neutron yield from a neptunium target will be higher, than from targets on the basis of the normal or depleted uranium.

^{237}Np cannot be used in the military aims like the ^{235}U and ^{239}Pu in spite of its sphere critical mass (without a reflector) is about ~ 61 kg [6].

Neutron yield from neptunium target will be increased by a multiplication factor (booster target). To create now a similar target on a basis of the high enriched uranium (~ 90% ^{235}U) like that, as it was used in IPNS ANL [7] is inexpediency and unrealistically because of nuclear safety problems, high price and related problems.

^{237}Np is not produced specially like other materials such as ^{235}U and ^{239}Pu. It is an inevitable by-product produced in nuclear reactors along with minor actinides (Am, Cm). Tons of neptunium are accumulated now (< 100 kg as metal, a rest as NpO_2 and solutions). For example, PWR -1000 (MW el.) produces about 14 kg of ^{237}Np per year [8]. Np is extracted from irradiated fuel along with isotopes U and Pu.

Table 3. Nuclear Properties of Target Materials

Isotope	Fission Yield - ν	Fission Threshold Energy, MeV	Delayed Neutron Fraction – β	Sphere critical mass (without reflector), kg
^{237}Np	~ 2.47	~ 0.5 MeV	**0.0041**	~ 61 [6]
^{238}U	2.6	~ 1.0 MeV	**0.0157**	No
^{235}U	2.5	0	**0.0068**	52 (93.8% ^{235}U) [9]

Neptunium has no wide application now. The small part of neptunium is used for ^{238}Pu manufacturing (as the energy source for nuclear space batteries). Neptunium can be used as the addition to fuel for fast nuclear reactors in the future. However only two fast reactor works now in the world: the power fast breeder reactor BN-600 MW el. and research fast reactor BOR-60 (Russia). Therefore it is too far prospect.

Considerable progress take place in technologies of the fuel elements (with metal fuel) providing high burn-out.

7. Some features of neptunium target

Some part of fast neutrons is slowed down and absorbed under the threshold of fission reaction in a presence of a coolant. However in case of loss of coolant these neutrons will cause fissions. It means that a positive effect of reactivity at loss of coolant take place $+\Delta K_{eff-cool}$.

For minimization of this effect, the volume fraction of coolant in a target and its cross-sections of elastic and inelastic scattering should be minimal.

D_2O and PbBi eutectic are most comprehensible from the point of view of a combination of technological and nuclear properties. Sodium was not considered because of fire danger.

It is necessary to notice that PbBi and D_2O coolants with equal volume fraction form practically equal neutron spectra in a tight lattice of cylindrical fuel elements. These targets will have practically equal multiplication factors [10, 11]. However the neutron yield from a target with D_2O coolant will be higher than from a target with PbBi eutectic. It is connected with additional losses of energy of a primary proton to ionization and, as a consequence, quantity of energy spent for neutron yield is decreased. Therefore PbBi coolant has been excluded from the further consideration. Besides, its use complicates technology of a neutron source.

The critical condition is not reached in the presence of water (H_2O). The water filling up of target is enough for conversion to a safe condition at any emergency.

The maximal multiplication factor for a subcritical assembly (target) without control system and emergency protection should not exceed 10 in any situation; including the maximal design failure connected with loss of a coolant (positive reactivity at loss of coolant is $\Delta K_{eff} \sim +0.05$ for neptunium target). It means that maximal multiplication factor in working position will have next value:

$$1 \Big/ 1-\left(k_{eff\,\max}+\Delta k_{eff}\right) \cong 10 \rightarrow k_{eff\,\max} \approx 0.85 \rightarrow K_{m\,\max} \cong 6 \div 7$$

The second reason of the multiplication factor restriction is connected with background between pulses caused by multiplication of delayed neutrons. The acceptable multiplying factor from the point of view of background is ~ 6÷7.

The background relation of neptunium target will be lower than for target on basis of enriched uranium, other things being equal.

8. Technological and project features of fuel elements and a target based on ^{237}Np

Radiotoxicity of neptunium is below than that of plutonium in view of smaller specific radioactivity. However, inevitable presence of the constantly nascent protactinium complicates process of machining of metal neptunium.

Fuel elements on the basis of the metallic not alloyed neptunium (α – phase, $\rho_{theory} \sim 20.45$ g/cm^3) can be used under following conditions:

- The temperature of fuel is less than temperature of phase transition - $t < t_{\max} = 280^0 C$.
- Burning out is ~ 1 % at.
- Cylindrical cladding of fuel elements is used for most successful resisting of swelling.
- The form of the fuel core should have the special form for compensation of the fuel swelling.
- The density of fuel under cladding is about ~ 80 % of the theoretical density, i.e. ~ 17 g/cm^3.
- There should be a reliable thermal contact between fuel and cladding to prevent fuel overheat. The AlSi alloy layer provides the desired thermal contact here.
- The maximum temperature can be lowered by expansion of beam cross-section (if necessary).

Additional factors for maintenance of these conditions;

- Fuel elements with metallic neptunium are disposed outside of the beam.
- The fuel elements with NpO_2 or dispersion type elements with high fuel content are placed under beam. The placing of the tungsten insert under beam is possible for protons with energies ~ 300 ÷ 450 MeV.

The dispersion type elements of novel generation with high fuel content. Fuel elements with uranium volume fraction in fuel rod up to 72% are considered as examples [12-14].

Structurally the dispersion fuel core consists of uniformly distributed higher density fuel granules of U–Mo, U–Nb–Zr or U_3Si alloys that are metallurgically bonded between themselves and to fuel cladding with specially developed Zr-based matrix alloys having the melting temperatures of 790–860 ^{0}C. The fuel element fabrication process involves vibrofilling of a zirconium cladding of a fuel element with blended fuel and matrix granules and a capillary impregnation i.e. a short-term (1–5 min) annealing at 840–910 ^{0}C. In this case a fuel core retains controllable porosity in the range of ~ 14% to accommodate fuel swelling. The volume fraction of fuel alloy is up to 72%, zirconium matrix – 14%, pores – 14%. The uranium content of a fuel alloy under the cladding is up to 11.7–12.9 g/cm^3.

High thermal conductivity of the fuel composition (18–22 W/m K) and metallurgical bond between the fuel and the cladding shifts down the maximum temperature of fuel (cold fuel).

Fuel composition in zirconium matrix has high resistance to aqueous corrosion.

The porosity of the fuel core allows the accommodation of swelling up to the burn-up of 1.0 g fiss/cm^3 (120 MW d / kgU).

Metallurgical bond between the fuel and the cladding makes fuel elements serviceable under transients and improvements of their operation reliability and safety.

The melting temperature of neptunium is 640 ^{0}C. In this case other matrix with a lower melting point is used. The volume fraction of neptunium in such matrix can be more than 72 %. This technology practically excludes machining of neptunium.

9. Comparison of neutron sources with tungsten and neptunium targets

The comparison of a neutron source with a tungsten target of the optimum thickness ~ 4 cm and targets based on metallic neptunium with cylindrical fuel elements of dispersive and metal type, and also their combination (the dispersive fuel elements are placed under the beam, the elements with metal fuel are out of the beam) is shown in Table 4.

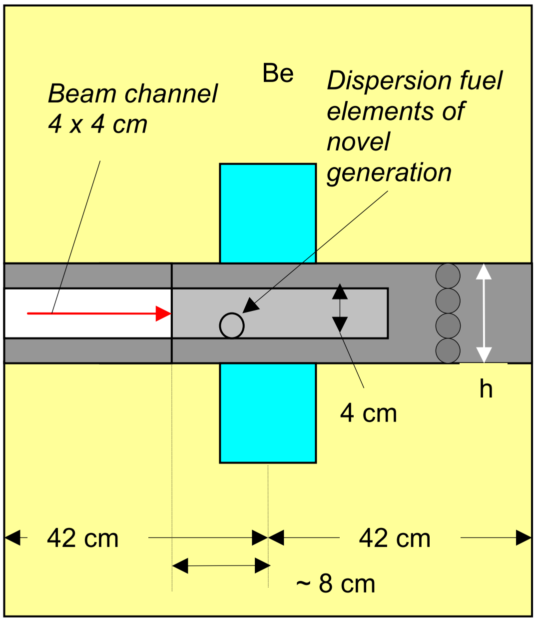


Fig. 3. The scheme of neutron source for the comparison. Moderator dimensions - 12 x 12 x 5 cm. Zones with dispersive and metal fuel elements are shown separately (for the combined target).

The considerable preference for a tungsten target is made in the given calculations. Real diameter of disks of tungsten targets is equal ~ 8÷10 cm, and so the gain factor will be ~ **4.**

For the target based on metallic natural uranium with cylindrical fuel elements the corresponding factor is equal to ~ 1.4 [3, 4], that is about ~ 3 times less than for neptunium target of the limited multiplication ~ 6.7. The increase of flux in ~ 1.4 times does not justify efforts and the cost spent for the uranium target.

Table 4. Multiplication factor – K_m and gain factor – $G = \varphi_{Np} / \varphi_W$ (relation of thermal neutron fluxes on a moderator surface of a neutron source with Np and W targets) depending on neptunium target thickness. E_p = 600 MeV.

Main target material and type of target	Target thickness h, cm	k_{eff}	K_m	$G = \varphi_{Np} / \varphi_W$
90%vol.W, 10%vol.H_2O	4	-	-	**1**
Cylindrical dispersion fuel elements (Np) of novel generation **G_{max} ~ 2.9,** $K_m \approx 6.7$	4	0.486	1.94	**1.36**
	6	0.598	2.48	**1.60**
	8	0.683	3.15	**1.84**
	10	0.750	4.00	**2.09**
	12	0.802	5.05	**2.41**
	14	0.847	6.53	**2.73**
	16	0.882	8.47	**3.30**
Cylindrical fuel elements with metal Np **G_{max} ~ 3.8,** $K_m \approx 6.7$	4	0.588	2.43	**1.72**
	6	0.714	3.49	**2.23**
	8	0.806	5.15	**2.96**
	10	0.878	8.19	**4.07**
Dispersion fuel elements under beam Fuel elements with metal Np out of beam **G_{max} ~ 3.5,** $K_m \approx 6.7$	4	0.536	2.15	**1.68**
	6	0.639	2.77	**2.00**
	8	0.719	3.56	**2.39**
	10	0.781	4.56	**2.75**
	12	0.830	5.88	**3.22**
	14	0.872	7.81	**3.89**

For the uranium target with plate-type fuel elements the corresponding factor is ~ 1.7 [15]. It is more than ~ 2 times below the possibilities of neptunium target. However targets with plate-type fuel elements have the short mean time of life (3 – 6 months), therefore such targets are too expensive for operation [16-18].

Further increase of G - factor may be done by using of booster target on basis of high enriched uranium (~ 90 % ^{235}U). This material however causes many problems.

Table 5. Comparative characteristics of targets

Target	Coolant	Type of fuel elements	Maximal multiply factor	Nucl. safety problems	G – in comparison with W target
W	H_2O, D_2O	Plate		No	1
Np by-product	D_2O, H_2O for rotated target only	Cylindrical	~ 7	No	Up to ~ 4
U natural	H_2O, D_2O	Cylindrical Plate (short lifetime)		No	1.4 too low 1.7 (short lifetime)
^{235}U	H_2O, D_2O	Cylindrical Plate (short lifetime)	Up to ~ 7	Yes	~ 5 too many problems

Besides, there are problems with nuclear safety. So, for example, any water cavity near the target or in the course of its assemblage can lead to a criticality set on thermal neutrons before the necessary factor of fast neutron multiplication will be reached. The background between pulses from delayed neutrons also limits the multiplication factor by value ~ 6÷7. Comparative characteristics of targets are shown in the Table 5.

It is necessary to notice, that manufacture of fuel elements is expensive enough process in spite of the fact that the basic material (Np) is a by-product. Therefore it is important to provide the long term operation of such target. Besides there is always a beam window problem, i.e. duration of service of the target case.

The cardinal increase of an average operating time of a target and window is possible only by means of a rotating target or by means of rotation of a target part being under a beam. For instance it can be Be-disk for protons of low energy, W-disk for protons with energy 300 ÷ 450 MeV or a rotating part of a source with U or Np elements for proton with energy 500 – 600 MeV. Diameters of boxes of neutron sources in radiation shield (at INR RAS) are ~ 1.6 m. It is more than is enough for creation and placing of such target, with time of life about ~ 10 years and with effective system of reflectors.

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