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Neutron producing target for accelerator based neutron capture therapy

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Abstract. Pilot innovative accelerator based neutron source for neutron capture therapy of cancer is under construction now at the Budker Institute. One of the main elements of the facility is lithium target producing neutrons via threshold ^{7}Li(p,n)^{7}Be reaction at 10 mA proton beam with energies of 1.915 MeV or 2.5 MeV. In the present report, choice of target was substantiated. The main problems of lithium target were determined to be: ^{7}Be radioactive isotope activation, keeping lithium layer solid, presence of photons resulted from proton inelastic scattering on lithium nuclei, and radiation blistering. The results of thermal testing of target prototype, investigation of radiation blistering and several simulations are presented. It becomes clear that water is preferable for cooling this target, and that the lithium target 10 cm in diameter is able to run up to 25 kW proton beam before melting. The conception of optimal target is proposed: thin and easy to detach metal disk 10 cm in diameter, evaporated with thin layer of pure lithium from the side of proton beam exposure: its back is intensively cooled with turbulent water flow to maintain lithium layer solid. Design of target for the neutron source constructed at BINP is shown. Conceptions of radiation protection and neutrons, γ-rays and α-particles diagnostics are presented. The immediate plans on obtaining epithermal neutron beam are declared.

1. Introduction

Pilot innovative accelerator based neutron source for neutron capture therapy is under construction now at the Budker Institute of Nuclear Physics, Novosibirsk, Russia [1]. One of the main elements of the facility is lithium target [2], that produces neutrons via threshold ^{7}Li(p,n)^{7}Be reaction at 10 mA proton beam with energies of 1.915 MeV or 2.5 MeV.

In this report, choice of target is substantiated, the main problems of lithium target are determined, results of thermal testing, investigation of radiation blistering and several simulations are presented. The conception of optimal target is proposed and design of target for the neutron source constructed at BINP is shown.

2. Choice of target

2.1.1. Reaction. Neutrons with energies of 0.5 eV – 10 keV are necessary for neutron capture therapy. Neutrons with higher or lower energies and γ-radiation are extremely undesirable. The following neutron-producing charged particles reactions are considered mainly for use in accelerator based neutron capture therapy: ^{7}Li(p,n), ^{9}Be(p,n), ^{9}Be(d,n) and ^{13}C(d,n). Table 1 shows the properties of these reactions.

Table 1. Characteristics of reactions considered for accelerator based neutron capture therapy.

Reaction	Bombarding energy, MeV	Neutron yield at 10 mA, s^{-1}	Average neutron energy at 0°, MeV	Maximum neutron energy at 0°, MeV	Target melting point, °C	Target thermal conductivity, W/(m K)
^{7}Li(p,n)	2.5	8.9×10^{12}	0.55	0.786	181	71
	1.915	2.9×10^{11}	0.04	0.113		
^{9}Be(p,n)	4.0	10×10^{12}	1.06	2.12	1287	201
^{9}Be(d,n)	1.5	$2.1 \times 10^{12*}$	2.01	5.81		
^{13}C(d,n)	1.5	1.8×10^{12}	1.08	6.77	3550	230

*Varies by a factor of three in the literature.

The ^{7}Li(p,n) reaction is excellent neutronically: neutron producing is high and relatively soft spectrum requires less moderation than those generated in other reactions. Unfortunately, lithium melting point is low, its thermal conductivity is poor, and finally, lithium is a very reactive metal, forming compounds immediately upon exposure to air. Alternate targets from beryllium and carbon overcome these difficulties in manufacture and cooling. However, more extensive moderators are required, and higher accelerator current is needed.

^{7}Li(p,n)^{7}Be reaction is a threshold one and it is characterized by unusually high increase in reaction cross section near threshold. This allows to consider an addition opportunity for near threshold operation when proton energy exceeds 30 – 40 keV the reaction threshold (1.882 MeV). Neutron beam with mean energy of 40 keV kinematically collimated is generated in this case. To decrease the neutron energy to epithermal value, thin water moderator (about 2 cm thick) is sufficient, therefore patient may be placed near the target. This provides the same treatment time that the standard mode at proton energy of 2.5 MeV.

Thus, for neutron generation, just ^{7}Li(p,n)^{7}Be reaction is proposed to be used with proton beam energy of 1.915 or 2.5 MeV, in spite of poor lithium properties. This choice intends developing much more complicated lithium target than the beryllium-9 or carbon-13 ones.

2.1.2. Pure lithium is more efficient in neutron generating than lithium hydride, oxide, nitride, or fluoride [4] (Table 2) and possesses higher thermal-conductivity, but incomparably lower melting temperature, therefore it requires efficient heat removal at as low lithium layer temperature as possible. Use of target with liquid lithium layer is also possible, but considerable lithium evaporation results in decrease in high voltage electric durability due to lithium vapor inflow and expansion of nascent radioactive beryllium over the whole facility.

Table 2.

Material	Li	LiH	Li_2O	LiF
Target melting point, °C	181	690	1500	850
Target thermal conductivity, W/(m K)	71 (s. 182 °C) 43 (l. 182 °C)	5.5 (200°C) 4 (500 °C)		12 (0°C) 3 (850 °C)
Neutron yield, arb. units	1	0.7	0.493	0.304

2.1.3. Thickness. Inelastic proton scattering on lithium nuclei leads to considerable γ-rays flux with energy of 478 keV that sometimes exceeds neutron flux [5]. In Table 3 the gamma yield is shown depending on proton energy for thick lithium target which stops a proton, and for thin one, braking a proton only to 1.882 MeV (energy of neutron generation reaction threshold). It is seen that thin target decreases considerably the gamma flow. In case of thin target protons should further brake in tungsten, molybdenum or any other substance whose inelastic scattering does not result in gamma radiation. This condition is met for almost all nuclei harder than aluminum one [6].

Table 3. 478 keV gamma yield for thick and thin natural lithium target.

Proton energy, MeV	478 keV gamma yield at 10 mA, s^{-1}	
	thick target	thin target
2.5	$3.66\ 10^{12}$	$2.1\ 10^{12}$
1.915	$1.6\ 10^{12}$	10^{11}

*2.1.4. Lithium target lifetime.*Radiation blistering is main processes determining lifetime of a target. Appearance of blisters increases the lithium layer evaporation due to rise of temperature because of swelling and flaking, and generally spoils the target. At current of 10 mA and target diameter of 10 cm, lower bound in hydrogen blistering of 10^{18} cm^{-2} [7] is achieved during 20 min. Fluence of 10^{19} cm^{-2}, which certainly causes blistering is achieved during 3.5 hour. It is clear that the time of blister appearance is comparable to the time of planned radiation treatment. That is why it is desirable to find materials as resistant to blistering as possible to prolong target operation time up to 10 hours. Jet liquid lithium target, of course, solves the blistering problems, but it causes the other ones.

Induced activity is another problem preventing the target from long-term operation. Every act of neutron production in the ^{7}Li(p,n)^{7}Be reaction is accompanied by radioactive nucleus of beryllium isotope. Beryllium isotope ^{7}Be becomes stable lithium isotope ^{7}Li due to capture of orbital electron with half-life of 53.6 days. The capture causes no radiation in 89,7 % cases, and in 10.3 % it radiates gamma-quantum with energy of 478 keV. Operation with open source of activity higher than 10^9 Bq is allowed in an isolated room only. As 10^9 Bq activity may be achieved quite quickly (12 min at the operation mode of 2.5 MeV 10 mA proton beam, and 6 hours at near threshold mode), than it is highly desirable to localize the radiation source by maintenance of lithium layer in solid state. Therefore, the simplest way to solve the problem of induced activity is to provide substrate with neutron-generating layer simple and easy to exchange.

As estimation shows, lithium layer dispersion caused by protons and decreasing of lithium layer thickness due to thermal evaporation at temperature below 200 ° are negligibly low. For example, at lithium temperature $T = 178$ °C, the velocity of the layer thickness decrease is $1.5\ 10^{-12}$ cm/s (Tabl. 4).

Table 4. Dependence of lithium pressure of saturating on temperature.

T, °C	178	204	234	268	306	350	402	464	538	629
, Torr	10^{-10}	10^{-9}	10^{-8}	10^{-7}	10^{-6}	10^{-5}	10^{-4}	10^{-3}	10^{-2}	0,1

2.1.5. Target diameter. The target dimensions are not so significant for 2.5 MeV mode requiring extensive moderator. This is not so for near threshold mode, with patient disposed very closely, optimal target diameter is 10 cm. At smaller diameter, local dose on skin increases; and the neutron flow density decreases rather significantly at bigger diameter of target.

2.1.6. Design of ideal target. Ideal stationary target seems to be thin metal disc 10 cm in diameter covered with thin layer of solid lithium (on the side of proton beam) and cooled by turbulent flow of water (on the opposite side). Shortening of lifetime of target due to blistering of target surface and target induced activity makes it clear that the target should be simple and easy to replace.

In this conception, pure lithium is required for maximum of neutron yield, thin lithium layer to decrease temperature and to reduce γ-ray flux, solid lithium to decrease lithium evaporation and to prevent radioactive isotope ^{7}Be expansion.

3. Thermal experiments

The first prototype of neutron producing stationary target was made [8] and tested under 20 kW 1.4 MeV electron beam [9]. This target consisted of molybdenum plate that diffusely welded on an ARMCO steel disk. Rectangular grooves were on the disk for cooling. In the process of examination, heat removal up to 650 W cm^{-2} was provided using water, and liquid metal cooling resulted in the target destruction due to high chemical interaction of gallium with ARMCO steel.

These experiments and calculations showed the way to improve the target and cooling system. A new variant of target is presented in Fig. 1 [2]. It is a tungsten disk 80 mm in diameter, 3 mm thick with cooling rectangular channels 3 mm × 2 mm at an interval of 3.7 mm, pressed to titanium body without diffuse welding. Laborious diffuse welding was refused, which resulted in possibility to obtain more homogeneous temperature field on the surface of the target at the expense of decrease of the size for the rib, to which the plate was stuck earlier, and at the expense of increase of the distance from the target surface to the heat carrier.

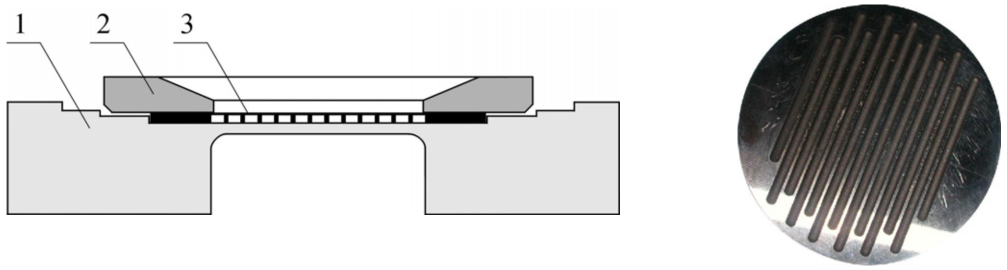

Figure 1. Target design (left): 1 – titanium body, 2 – clamping flange, 3 – beam absorber (tungsten disk with 13 channels for cooling). Proton beam falls onto the beam absorber from above. Beam absorber photo (right); view from the side opposite the one which is exposed to the proton beam.

This neutron producing target was tested under 20 kW heater. The heater is manufactured from niobium plate 1 mm thick, 45 mm × 45 mm, by spark cutting 2 mm stripes with 0.2 mm gap. Then using electrochemical micro-arc technique it was covered with thin (5-10 μm) layer of Nb_2O_5 to provide isolation of not less than 300 V. The heater resistance is of order of 1 Ohm. It is pressed to the target through 1 mm thick BeO plate. The heater is fed from a stabilized power source with current up to 100 A. In Fig. 2, dependence of target surface temperature on heating is presented. 3 regimes of heat removal are seen: i) up to 250 W cm^{-2} – turbulent flow with good correlation to calculation, ii) with bubble boiling (temperature does not practically change when heating increases), and iii) with film boiling more then 450 W cm^{-2} (heat-transfer drops, and temperature increases sharply). Wide plate presenting allows to detect target overheating without this undesirable dropping of heat removal.

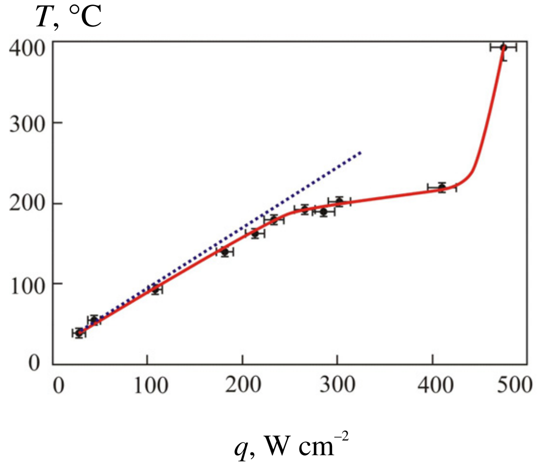

Figure 2. Dependencies of target surface temperature at water velocity 3 m s^{-1} on heat density: solid line – measured, touch line – calculated.

So, thermal investigations and calculations [2] allow us to speak with confidence that the lithium target in 10 cm diameter could run up to 10 mA proton beam before melting using turbulent flow of water with velocity about 10 m s^{-1} for cooling.

4. Lithium layer

Lithium is a very reactive metal, forming compounds immediately upon exposure to air. A unit for lithium evaporation using industrial gate valve (Fig. 3) was developed to generate lithium layer directly at the facility. Heater and lithium containing volume were placed at movable counter-plate. For lithium evaporation, the counter-plate with heater and container is pushed to proton channel and is moved several millimeters to the direction of proton beam up to heat-insulated silphon restricting the volume of evaporation. Further heat of the counter-plate and container allows to deposit practically all amount of lithium uniformly over the target substrate.

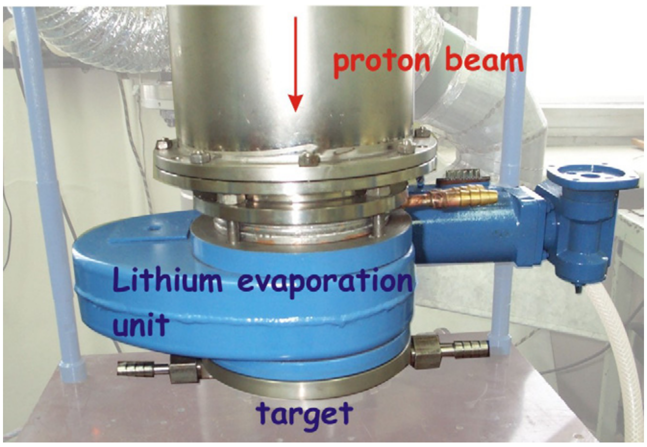

Figure 3. Lithium evaporation unit.

Immediately after the exposure to air the lithium layer from silver-white turns to practically black (it must be Li_3N). Several days later, this layer becomes gray (probably, it becomes Li_2O), it distends here and there and easily comes off the substrate (Fig. 4).

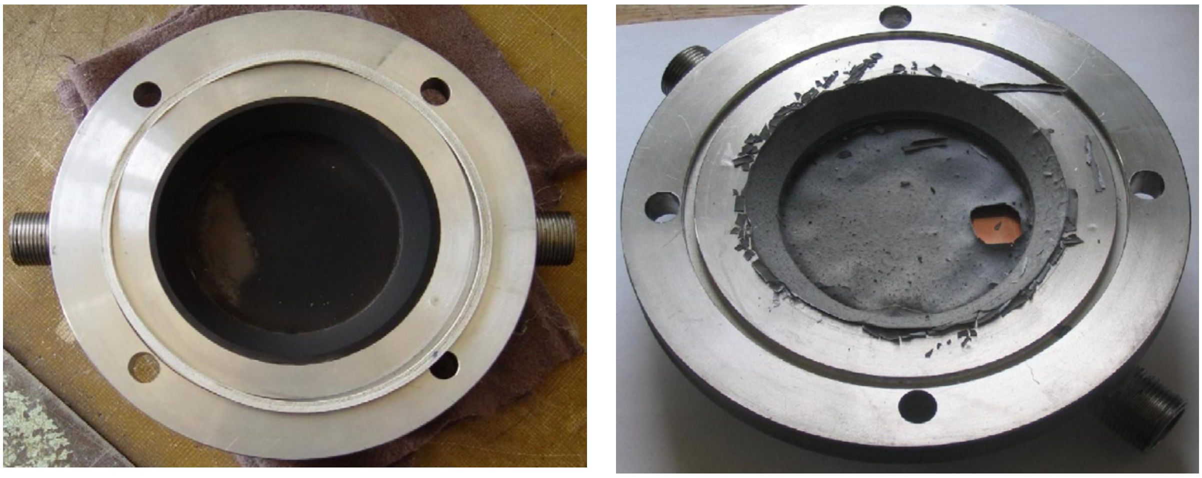

Figure 4. Target at once after opening (left), after 3 days in air (right).

5. Target design for BINP facility

Target design for BINP accelerator neutron source is presented in Fig. 5. In contrast to target prototypes described in subsection 3, cooling channels are spiroid for more effective heat removal (four spiral cooling channels, 2 turn). Pressure drop 2 atm. allows turbulent water flow (Re = $4\ 10^4$) with velocity of 10 m s^{-1}. Water consumption is 3,5 m^3 $hour^{-1}$, heating is 8 °C.

Bayonet connector is used to remove the target substrate automatically that is desirable at operation with radio-active isotopes.

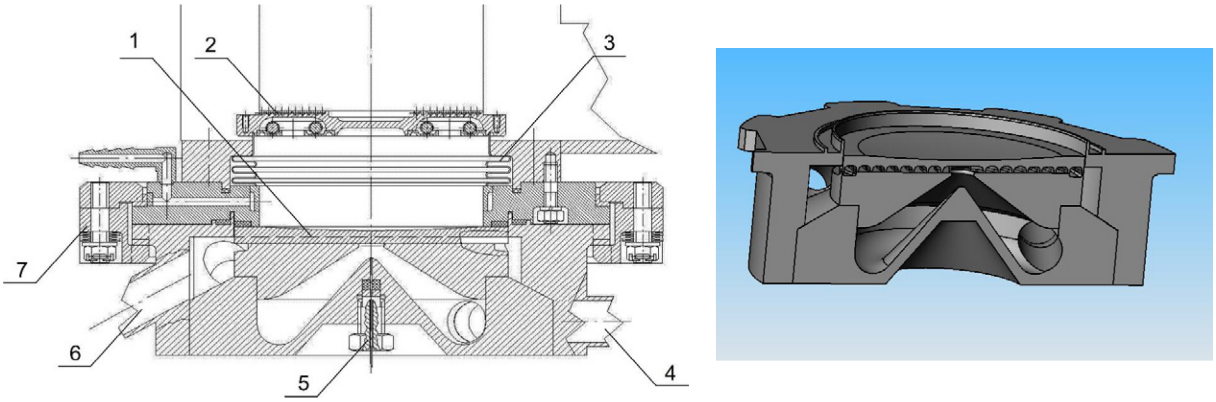

Figure 5. Target for BINP accelerator neutron source: 1 – backing with lithium layer, 2 – counter-plate with heater and container of lithium evaporation unit, 3 – silphon, 4 – water input, 5 – thermocouple, 6 – water output, 7 – bayonet.

6. Conclusion

For accelerator neutron capture therapy, ^{7}Li(p,n)^{7}Be reaction is preferable, in spite of poor lithium properties.

Neutron-generating target is developed for accelerating source of epithermal neutron that is under construction at BINP. The target is thin metal disc 10 cm in diameter, evaporated with thin layer of pure solid lithium from the direction of proton beam, and its opposite side is cooled intensively with turbulent flow of water. It became clear that using water is preferable for cooling this target, and that the lithium target could run up to 10 mA proton beam before melting. Such simple and easy to replace target allows us to solve the problem of blistering and induced activity.

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