

Development of lithium target for accelerator based neutron capture therapy

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Pilot innovative accelerator based neutron source for neutron capture therapy of cancer is now on the threshold of its operation at the BINP, Russia. One of the main elements of the facility is lithium target producing neutrons via threshold ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction at 25 kW proton beam with energies 1.915 MeV or 2.5 MeV. The main problems of lithium target were determined to be: ${}^7\text{Be}$ radioactive isotope activation, keeping lithium layer solid, presence of photons due to proton inelastic scattering on lithium nuclei, and radiation blistering. The results of thermal test of target prototype were presented at previous NCT Congress. It becomes clear that water is preferable for cooling the target, and that lithium target 10 cm in diameter is able to run before melting. In the present report, the conception of optimal target is proposed: thin metal disk 10 cm in diameter easy for detaching, with evaporated thin layer of pure lithium from the side of proton beam exposure, its back being intensively cooled with turbulent water flow to maintain lithium layer solid. Design of the target for the neutron source constructed at BINP is shown. The results of investigation of radiation blistering and lithium layer are presented. Target unit of facility is under construction now, and obtaining neutrons is expected in nearest future.

Keywords: BNCT, Accelerator, Target, Lithium

1. Introduction

Pilot innovative accelerator based neutron source for neutron capture therapy of cancer [1] is now on the threshold of its operation at the BINP, Russia. One of the main elements of the facility is lithium target producing neutrons via threshold ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction at 25 kW proton beam with energies 1.915 MeV or 2.5 MeV. The results of thermal testing of target prototype were presented at previous NCT Congress [2].

In the present report, choice of target is substantiated, the main problems of lithium target are determined, the conception of optimal target is proposed. The results of investigation of radiation blistering and lithium layer are presented. Design of target for the neutron source constructed at BINP is shown.

2. Choice of target

Reactions. 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: ${}^7\text{Li}(p,n)$,

${}^9\text{Be}(p,n)$, ${}^9\text{Be}(d,n)$ and ${}^{13}\text{C}(d,n)$ [3]. Table 1 shows the properties of these reactions.

The ${}^7\text{Li}(p,n)$ 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.

${}^7\text{Li}(p,n){}^7\text{Be}$ 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.

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 × 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.

Table 2.

Material	Li	LiH	Li ₂ O	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

Thus, for neutron generation, just ⁷Li(p,n)⁷Be 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.

Purity. 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.

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

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. Fluence which certainly causes blistering is about 2 10¹⁹ cm⁻² for copper and a hundred times as much for metals that solve hydrogen well. At current of 10 mA and target diameter of 10 cm, blistering appears on copper substrate after several hours, and such a target will require daily replacement. But, copper substrate is not expensive and quite easy to manufacture, its heat conductivity is high, and all these features make this substrate the main solution. Substrate made of metals that solve hydrogen well may be replaced once a week. But heat conductivity of these metals is lower than copper, their manufacturing is harder; and what is the most important, effect of interaction for hydrogen diffusing to substrate surface with lithium is not yet clear.

Induced activity is another problem preventing the target from long-term operation. Every act of neutron production in the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction is accompanied by radioactive nucleus of beryllium isotope. Beryllium isotope ${}^7\text{Be}$ becomes stable lithium isotope ${}^7\text{Li}$ 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. Gate valve used near the target allows increasing operation time, as target may be replaced in specialized room. It allows a clinic to work with radioisotope in close source regime. Another way is realized by solving lithium in a liquid and utilizing liquid radioactive wastes.

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

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.

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 ${}^7\text{Be}$ expansion.

3. Thermal testing

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. 1) 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 .

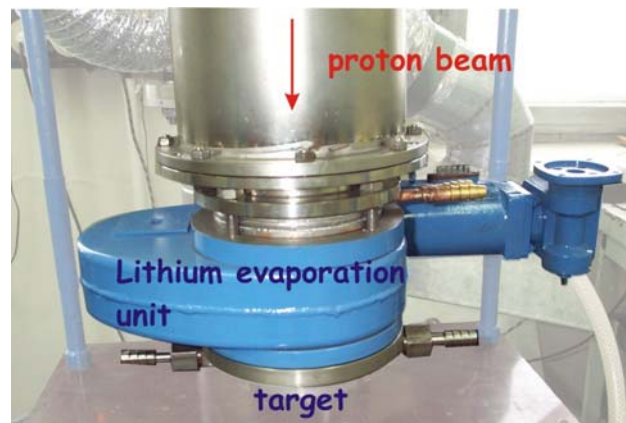


Fig. 1. Lithium evaporation unit.

Visual observation of lithium layer behavior in vacuum and in the air was carried out. 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. Lithium layer composition under different vacuum conditions was investigated at secondary ion mass spectrometer quadrupole-type Riber MIQ-256. Depth profile of elements in lithium layer was determined. It was cleared out that change in lithium layer composition due to interaction between lithium and residual gas could not cause drop in neutron yield.

Radial distribution of lithium layer thickness (Fig. 2) was measured by original technique: several small thin witnesses are placed on target through good thermal contact. After evaporation a lithium layer of a witness is solved in distilled water and then conductivity of water is measured. It was cleared out that conductivity is determined only by solved lithium quantity and does not depend on the compound. This allows operating with the target in air.

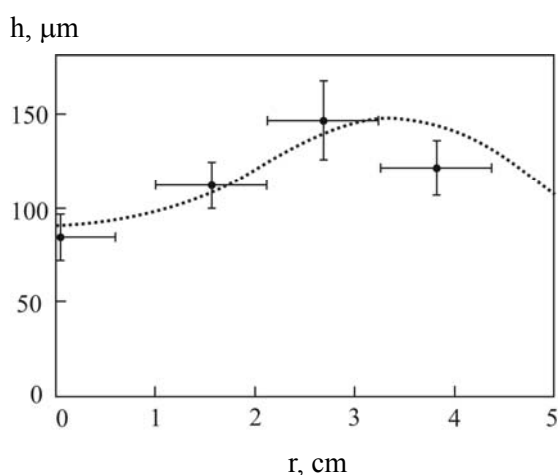


Fig. 2. Radial distribution of lithium layer thickness: ● – measured,– calculated for collisionless flow of lithium vapors from evaporator.

5. Target design for BINP facility

Target design for BINP accelerator neutron source is presented in Fig. 3. In contrast to target prototypes, cooling channels are spiroid for more effective heat removal (four spiral cooling channels, 2 turn). Pressure drop 2 atm. allows turbulent water flow ($Re = 4 \cdot 10^4$) with velocity of 10 m s^{-1} . Water consumption is $3,5 \text{ m}^3 \text{ hour}^{-1}$, heating is $8 \text{ }^\circ\text{C}$. Bayonet connector is used to remove the target substrate automatically that is desirable at operation with radio-active isotopes.

6. Conclusions

For accelerator neutron capture therapy, ${}^7\text{Li}(p,n){}^7\text{Be}$ 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 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.

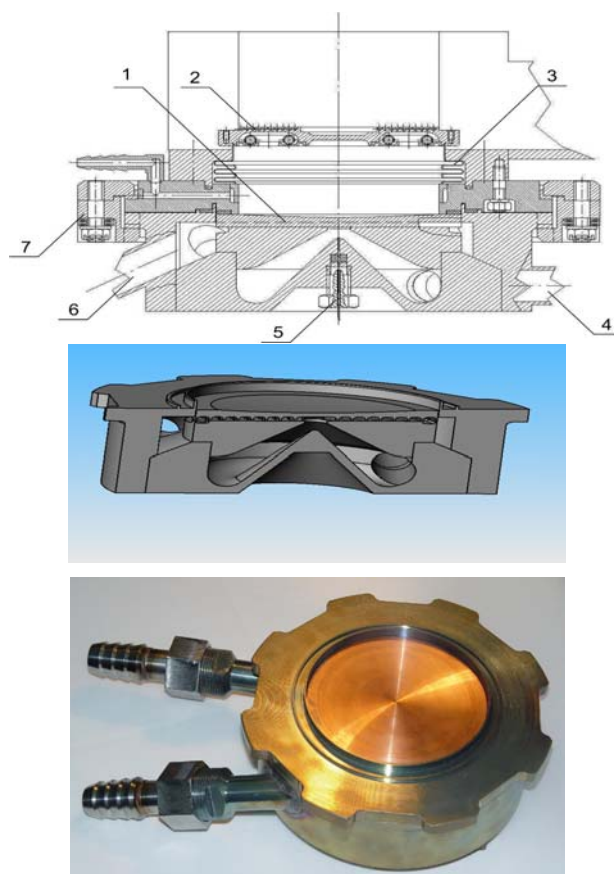


Fig. 3. Target for BINP accelerator neutron source: 1 – backing with lithium layer, 2 – counter-plate with heater and container of lithium evaporation unit, 3 – silphon, 4 – water input, 5 – thermocouple, 6 – water output, 7 – bayonet.

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