

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

A study of gamma-ray and neutron radiation in the interaction of a 2 MeV proton beam with various materials



Applied Radiation and

D. Kasatov^{a,b}, A. Makarov^a, I. Shchudlo^a, S. Taskaev^{a,b,*}

^a Budker Institute of Nuclear Physics, 11 Lavrentiev Avenue, 630090 Novosibirsk, Russia
^b Novosibirsk State University, 2 Pirogov street, 630090 Novosibirsk, Russia

HIGHLIGHTS

- Radiation is measured for interaction of 2 MeV protons with various materials.
- Absorption of protons in molybdenum and tantalum leads to minimal radiation.
- Neutron generating target with thin lithium layer for BNCT must be made of tantalum.

ARTICLE INFO

Article history: Received 1 November 2014 Received in revised form 29 July 2015 Accepted 11 August 2015 Available online 13 August 2015

Keywords: Boron neutron capture therapy Tandem accelerator

ABSTRACT

Epithermal neutron source based on a tandem accelerator with vacuum insulation and lithium target has been proposed, developed and operated in Budker Institute of Nuclear Physics. The source is regarded as a prototype of a future compact device suitable for carrying out BNCT in oncology centers. In this work the measurements of gamma-ray and neutron radiation are presented for the interaction of a 2 MeV proton beam with various materials (Li, C, F, Al, V, Ti, Cu, Mo, stainless steel, and Ta). The obtained results enabled the optimization of the neutron-generating target and the high energy beam transportation path.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

The optimal reaction to generate epithermal neutrons for their application in BNCT is the reaction ⁷Li(p,n)⁷Be. The interaction of protons with lithium nuclei results not only in the generation of neutrons but also gamma photons with an energy of 0.478 MeV. In order to decrease this undesirable gamma flux, the lithium layer should be only as thick as needed to slow down the protons to the threshold neutron generation energy of 1.882 MeV. Further proton absorption must be performed in another substance in which the proton interaction with the substance nuclei as a result of reactions (p, γ), (p,p γ), (p,n γ), and (p, $\alpha\gamma$) does not cause a noticeable gamma and X-ray yield. The aim of this work was the experimental measurement of the dose rate and the gamma radiation spectrum when 2 MeV protons are directed to the target made of various materials.

E-mail address: taskaev@inp.nsk.su (S. Taskaev).

http://dx.doi.org/10.1016/j.apradiso.2015.08.011 0969-8043/© 2015 Elsevier Ltd. All rights reserved.

2. Experimental results and discussion

The studies were carried out on an epithermal neutron source based on a tandem accelerator with vacuum insulation and a lithium target, which was developed to elaborate the BNCT procedure (Aleynik et al., 2014a). A specially manufactured cooled vacuum volume in the form of a cylindrical stainless tube with the inner diameter of 105 mm and a copper disk was installed instead of the lithium neutron-generating target. Various materials, preferably made in the form of disks 95 mm diameter about 1 mm thick, were placed inside the vacuum volume and irradiated by a 2 MeV proton beam with the current up to 500 µA. The absorbed dose rate of electromagnetic radiation was measured by a spherical ionization chamber at a distance of 25 cm from the target under study and a dose rate of neutron radiation was measured by a DKS-96 radiometer dosimeter with a BDMN-96 detecting unit at a distance of 50 cm. The ionization chamber registers not only the radiation from the target under the influence of the proton beam but the bremsstrahlung from the accelerator because of the accompanying current of electrons (Kasatov et al., 2015). To determine the contribution of bremsstrahlung the second ionization chamber was used, which was located far enough from the target and registered only the bremsstrahlung from the accelerator. The

^{*} Corresponding author at: Budker Institute of Nuclear Physics, 11 Lavrentiev Avenue, 630090 Novosibirsk, Russia.



Fig. 1. Time dependences of the dose rate of X-ray and γ -radiation measured by ionization chambers at a distance of 7.5 m from the accelerator and 6.4 m from the target (1) and at a distance of 25 cm from the target (2) under proton beam irradiation of stainless steel, vanadium, tantalum, and titanium. The time dependence of the proton beam current (3) is also shown.

characteristic graphs are depicted in Fig. 1. It is seen that when the proton beam current significantly changes, the radiation dose rate near the target (2 in Fig. 1) practically does not change for tantalum and drastically changes for vanadium, titanium, and stainless steel. This suggests that near the target the emission from tantalum is much less than the bremsstrahlung from the accelerator, while the emission from vanadium, titanium, and stainless steel is comparable.

Table 1 lists the processed results of the electromagnetic radiation dose rates for the studied materials. Note that due to the need for efficient cooling of lithium the measurement of the radiation dose rate on lithium was carried out under somewhat different conditions: with the use of a neutron-generating target (Bayanov et al., 2006) in which a lithium layer was sputtered on a copper substrate in one case, and without it in another. As in the first case the dose was 8.3 times greater, the value for lithium in the table is the value for copper multiplied by 8.3. It is seen that molybdenum or tantalum must be used for the target substrate to decrease the accompanying gamma and X-ray flux.

In order to measure the spectrum of the gamma and X-ray flux BGO (bismuth germanate Bi₄Ge₃O₁₂) and NaI gamma-spectrometers were employed. The BGO scintillator had a height of 100 mm, a diameter of 80 mm; those of the NaI one were 60 mm each. Gamma spectrometers were calibrated using a ⁴⁰K spectral line recorded in the background radiation with respect to radioisotope sources of ⁶⁰Co (with an activity of 5.66×10^7 Bq and gamma quantum energies of 1173 and 1333 keV) and ¹³⁷Cs

Table 1

A	bsor	bed	radiatio	n dose	rate	under	2 M	leV	proton	beam	irrac	liation	of	materi	ial	s.
---	------	-----	----------	--------	------	-------	-----	-----	--------	------	-------	---------	----	--------	-----	----

Atomic weight	Material	Dose rate at 1 m for a proton beam 1 mA, $(\mu Sv/h)$	Error, (%)
7	Li (50 µm)	750	5
12	C	25	10
26	LiF	20,000	20
175	BaF ₂	6500	20
27	Al	150	5
28	Si	23	2
48	Ti	230	8
50	V	270	4
55	Fe	70	10
63	Cu	90	5
96	Мо	< 6	
180	Та	< 6	



Fig. 2. γ -radiation spectrum of various materials irradiated by a 2 MeV proton beam.

 $(2.15 \times 10^8$ Bq, 662 keV). Spectral characteristics of gamma-radiation measured on the BGO spectrometer are presented in Fig. 2. Most graphs exhibit a peak of 511 keV gamma rays caused by the annihilation of generated positrons. The residual activity of materials measured on the NaI gamma-spectrometer is shown in Fig. 3. It was determined that the activation of the graphite results from the ${}^{12}C(p){}^{13}N \stackrel{\rho^+(10\,\text{min})}{\rightarrow} {}^{13}\text{C}$ process, activation of titanium – from the absorption of protons by ${}^{46}\text{Ti}$ and ${}^{47}\text{Ti}$ isotopes followed by β^+ -decay of ${}^{47}\text{V}$, ${}^{48}\text{V}$ nuclei, and electron capture of ${}^{48}\text{V}$, activation of lithium fluoride – from emergence of the radioactive isotope ${}^{7}\text{Be}$ in the reaction ${}^{7}\text{Li}(p,n){}^{7}\text{Be}$. It is worth noting a great gamma yield from fluorine. The reaction ${}^{19}\text{F}(p,\alpha e^+e^-){}^{16}\text{O}$ is characterized by a noticeable cross-section (up to 0.15 b) and is considered for the application in a powerful positron source (Farrell et al., 2002).

It is found that irradiation of stainless steel, titanium, and vanadium is also accompanied by neutron emission. The neutron dose rate is measured by a DKS-96 radiometer dosimeter. For vanadium it was 2500 μ Sv/(h m² mA) and for stainless steel it was 25 times less. Neutron generation is confirmed by the appearance of the characteristic spectrum in the signal of a neutron detector with a GS20 lithium-containing scintillator (Aleynik et al., 2014b). Absorption of a neutron by GS20 scintillator results in ${}^{6}\text{Li} + n \rightarrow {}^{3}\text{H}$ $+\alpha$ reaction with the release of 4.785 MeV. Fig. 4 shows that at the irradiation of vanadium and stainless steel a signal is present in the channels 300-400. This indicates the neutron generation. This figure also shows the signal from the irradiation of copper, from which it is clear that there is no generation of neutrons. Fig. 5 presents the dependence of the neutron count rate on the proton energy. It is seen that neutron generation from stainless steel is caused by the reaction ${}^{55}Mn(p,n){}^{55}Fe$ (reaction threshold is



Fig. 3. γ -radiation spectrum of the residual activity of lithium fluoride (1), titanium (2) and graphite (3) (acquisition time is 100 s).



Fig. 4. Dependence of counting rate of the neutron detector from the channel number (energy) when protons with an energy of 2 MeV irradiate copper (*Cu*), stainless steel and vanadium (*V*).



Fig. 5. Dependence of counting rate of the detector in the area of neutron peak from the proton energy at the absorption of protons in vanadium (V), titanium (Ti), stainless steel and lithium fluoride (Li).

1.034 MeV) due to the presence of manganese as an impurity. From titanium neutrons are generated by the reaction $^{49}\text{Ti}(p,n)^{49}\text{V}$ (reaction threshold is 1.43 MeV), and from vanadium by the reaction $^{51}\text{V}(p,n)^{51}\text{Cr}$ (reaction threshold is 1.562 MeV).

3. Conclusions

In the work the dose rate and the spectrum of gamma radiation

generated by 2 MeV proton irradiation of various materials (F, Li, C, Al, Si, Ti, V, Fe, Cu, Mo, Ta) are experimentally measured. It is revealed that in order to decrease the undesirable accompanying gamma radiation the substrate of a neutron generating target with a thin lithium layer must be made of molybdenum or tantalum. We have previously found that tantalum is also one of three materials having the maximum resistance to radiation damages (blistering) under their irradiation with 2 MeV protons provided that the temperature of the material does not exceed 150 °C (Astrelin et al., 2010; Taskaev, 2014).

It is also established that in order to exclude neutron and γ radiation when the proton beam falls on the construction materials (in case of an emergency situation) they must be protected by tantalum or molybdenum or made of these metals.

Acknowledgments

The study was supported by the grants from the Russian Science Foundation (Project no.14-32-00006) and the Budker Institute of Nuclear Physics.

References

- Aleynik, V., Bashkirtsev, A., Kanygin, V., Kasatov, D., Kuznetsov, A., Makarov, A., Schudlo, I., Sorokin, I., Taskaev, S., Tiunov, M., 2014a. Current progress and future prospects of the VITA based neutron source. Appl. Radiat. Isot, 88, 177–179.
- Aleynik, V., Kasatov, D., Makarov, A., Taskaev, S., 2014b. Measuring the neutron spectrum of the accelerator-based source using the time-of-flight method. Instrum. Exp. Tech. 57, 381–385.
- Astrelin, V., Burdakov, A., Bykov, P., et al., 2010. Blistering of the selected materials irradiated by intense 200 keV proton beam. J. Nucl. Mater. 396, 43–48.
- Bayanov, B., Belov, V., Taskaev, S., 2006. Neutron producing target for accelerator based neutron capture therapy. J. Phys. 41, 460–465.
- Farrell, J., Dudnikov, V., Guardala, N., Merkel, G., Taskaev, S., 2002. An intense positron beam source based on a high current 2 MeV vacuum insulated tandem accelerator. In: 7th International Workshop on Positron and Positronium Chemistry. Knoxville, USA, 7–12 July 2002, p. 47.
- Kasatov, D., Makarov, A., Taskaev, S., Shchudlo, I., 2015. Recording of current accompanying an ion beam in a tandem accelerator with vacuum insulation. Tech. Phys. Lett. 41, 139–141.
- Taskaev, S., 2014. Accelerator Based Epithermal Neutron Source (DSc. thesis). Novosibirsk, Russia.