IN SITU OBSERVATIONS OF BLISTERING OF A METAL IRRADIATED WITH 2 MeV PROTONS *

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Abstract

A vacuum-insulated tandem accelerator was used to observe in situ blistering during 2-MeV proton irradiation of metallic samples to a fluence of up to 6.7 10^{20} cm^{-2}. Samples consisting of copper of different purity, tantalum, and tantalum-copper compounds were placed on the proton beam path and forced to cool. The surface state of the samples was observed using a CCD camera with a remote microscope. Thermistors, a pyrometer, and an infrared camera were applied to measure the temperature of the samples during irradiation. After irradiation, the samples were analyzed on an X-ray diffractometer, laser and electron microscopes. The present study describes the experiment, presents the results obtained and notes their relevance and significance in the development of a lithium target for an accelerator-based neutron source, for use in boron neutron capture therapy of cancer.

INTRODUCTION

To enable the development of a promising cancer therapy, boron neutron capture therapy [1, 2], an accelerator-based epithermal neutron source was designed and made at the Budker Institute of Nuclear Physics [3, 4]. Neutrons are generated as a result of a ^{7}Li(p,n)^{7}Be threshold reaction by directing a proton beam, with an energy of 2 MeV and a current of up to 5 mA obtained in a tandem accelerator with vacuum insulation, to a lithium target with a diameter of 10 cm. The target is made of a thin lithium layer deposited on an efficiently-cooled substrate (in this study, copper) [5, 6]. The thickness of the neutron-generating lithium layer is chosen so that the proton energy coming out from the layer is slightly below 1.882 MeV (the ^{7}Li(p,n)^{7}Be reaction threshold). Then, protons are decelerated and absorbed into a construction material that must satisfy the following requirements: firstly, this material must either have a high thermal conductivity, or be thin enough for the lithium temperature to not exceed the melting point of 182 °C, to prevent the propagation of radioactive beryllium-7 nuclei that are formed and lithium vapour. From this standpoint, copper is the best material. Secondly, the proton deceleration in this material should not cause a noticeable increase in undesired X-ray and gamma-radiation. As a result of the studies performed [7], the best materials are found to be molybdenum and tantalum. Thirdly, this material must be sufficiently resistant to radiation blistering [8-10]; defined as the deformation of the surface layer in the form of numerous blisters (uplifting and peeling off of a thin layer of the material), which leads to a decrease in thermal conductivity. Experimental data on the critical dose of blistering have been extremely scarce and are absent for a proton energy of about 2 MeV. The aim of this study was to investigate the blistering, under 2-MeV proton irradiation, of samples made of copper, tantalum and copper-tantalum alloys.

EXPERIMENTAL APPARATUS

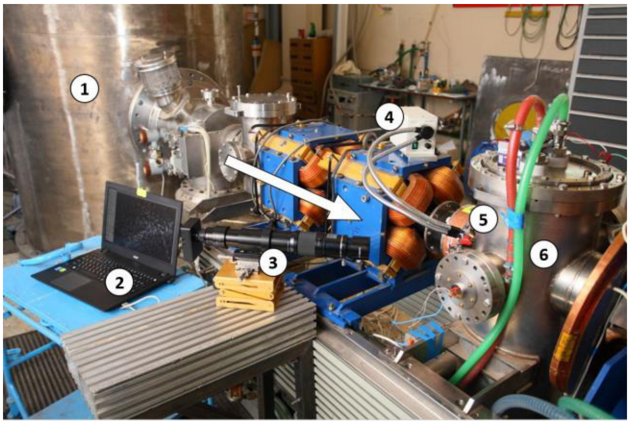

Figure 1: Experimental apparatus: 1 - tandem accelerator with vacuum insulation; 2 - computer; 3 - remote microscope with a CCD camera; 4 - booster-light; 5 - fused quartz window; 6 - diagnostic vacuum chamber. The arrow schematically shows the direction of 2-MeV proton beam propagation.

The studies were carried out on a tandem accelerator with vacuum insulation [3] that generates a proton beam with an energy of 2 MeV, a current of 0.5 mA, and a transverse dimension of about 1 cm. Figure 1 shows an image of the experimental apparatus. A disc-shaped sample with a diameter of 30 mm and a thickness of 3 mm was placed

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on the axis in the diagnostic vacuum chamber of the proton beam path. Via a gallium-indium alloy, the sample was tightly pressed to the heat-removing surface in the form of a copper plate with closely spaced holes to allow coolant flow. The sample temperature was measured to an accuracy of 1 °C with thermistor inserted inside. The sample surface temperature was measured with an Optris pyrometer through a fused quartz window and a FLIR T650SC infrared camera through a barium fluoride window. The current of the proton beam incident on the sample was measured with an ohmic divider when 100 V positive voltage was applied to the sample and 300 V negative voltage was applied to a suppressor ring at a distance of 50 mm in front of the sample.

Continuous real-time monitoring of the sample surface was performed through a fused quartz window by means of a CCD camera Mightex with a remote Infinity K2 microscope installed at an angle of 42° to the normal to the sample surface. Through an additional fused quartz window, the samples were enlightened by a LFP-10WP-R halogen lamp with a power of 10 W. A computer was installed near the CCD camera to collect data from the camera, save images in the local base, and display the real-time state of the target.

After irradiation, the samples were analyzed on a SHIMADZU XRD-7000 diffractometer (Shimadzu Co., Japan), a KEYENCE VK-X200 laser scanning 3D microscope (Keyence Co., USA), a Jeol JCM-5700 electron microscope (Jeol, Japan), and a FIB-SEM Helios G3 UC focused ion beam electron microscope (FEI, USA).

EXPERIMENTAL RESULTS

Eight types of samples were studied. Four of them were prepared from copper of different purity: M0 copper (State Standard GOST 859-2014, Russia), 99.996% fine-grained copper (OFC-1 JIS H3150 C1011, SH Copper Products Co., Ltd., Japan), 99.99996% coarse-grained and 99.99996% fine-grained coppers (High Purity Copper, Mitsubishi Materials Co., Japan). In the next three samples, ~100 μm thick tantalum foil was welded to copper using different techniques (explosion, diffusion and soldering). Sample 8 contained a ~100 μm thick upper layer made of tantalum and copper powders in a 1:1 volume ratio.

The characteristic dynamics of the appearance of blisters is shown in Fig. 2. A typical image of blister is shown in Fig. 3.

The proton beam profile was determined from the spread of the blistering region boundary with irradiation time. The proton beam size at the half-height was 8.9 mm horizontally and 12.4 mm vertically with an accuracy of 5%. The beam effective area, defined as a ratio between the current and the maximum current density, amounted to 75 ± 7 mm^2. Taking the beam area to be 75 mm^2, we determined that a 1 mA·h integral of the current corresponds to a particle fluence of (3 ± 0.3) 10^{19} cm^{-2}.

We summarize the key results of the performed studies on 2-MeV proton irradiation of different samples as follows:

The blistering threshold of the copper surface depends on the copper purity. The purer the copper, the higher the threshold is. The maximum threshold is 3×10^{19} cm^{-2}; the minimum value is seven times lower.

The size of the blisters on the copper surface depends on the copper purity. The purer the copper, the larger the blisters are. Blister size ranges from 40 ± 20 to 160 ± 50 μm.

No dependence of the blistering threshold on the copper crystallite orientation was found.

Once blisters appear on the copper surface, further irradiation does not cause any more surface modification, which can be due to the formation of holes and cracks when blisters emerge.

The attachment of a thin tantalum foil to copper by explosion or diffusion welding as well as soldering is resistant to a heat load of up to 1 kW/cm^2.

Tantalum is much more resistant to the formation of blisters than copper. The threshold of blisters in the form of bubbles or flakes on the tantalum surface exceeds 6.7×10^{20} cm^{-2}. At a fluence of 3.6×10^{20} cm^{-2} the surface is modified in the form of a relief with a characteristic cell size of 1 μm.

During tantalum irradiation, an increase in the sample surface temperature was detected. This could be due to a decrease in the thermal conductivity because of the appearance of cavities and hydrogen incorporation into the tantalum crystal structure.

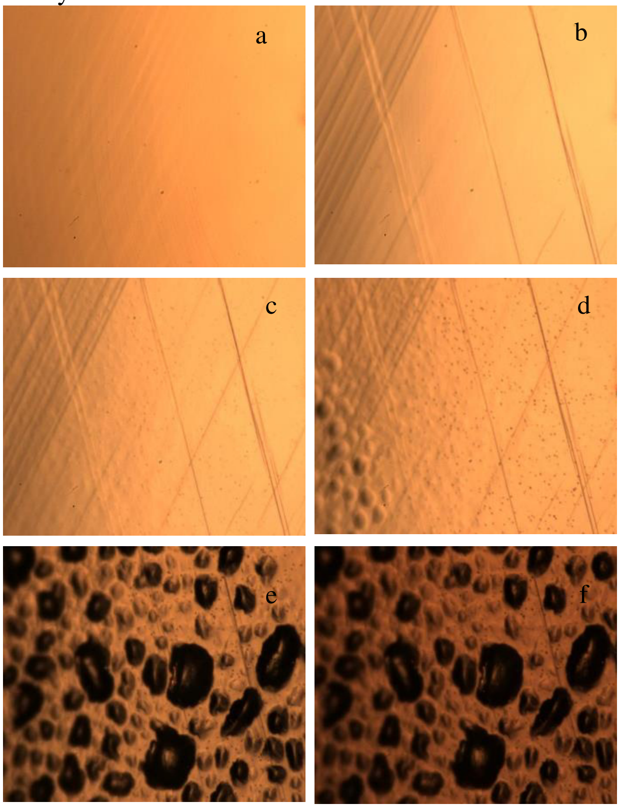

Figure 2: Images of the surface of 99.99996% coarse copper during irradiation by a 2-MeV proton beam with varying integrals of the current: (a) 0, (b) 0.04, (c) 0.25, (d) 0.50, (e) 1.00, (f) 1.89 mA h. The frame size is 7.3 mm horizontally and 3.8 mm vertically.

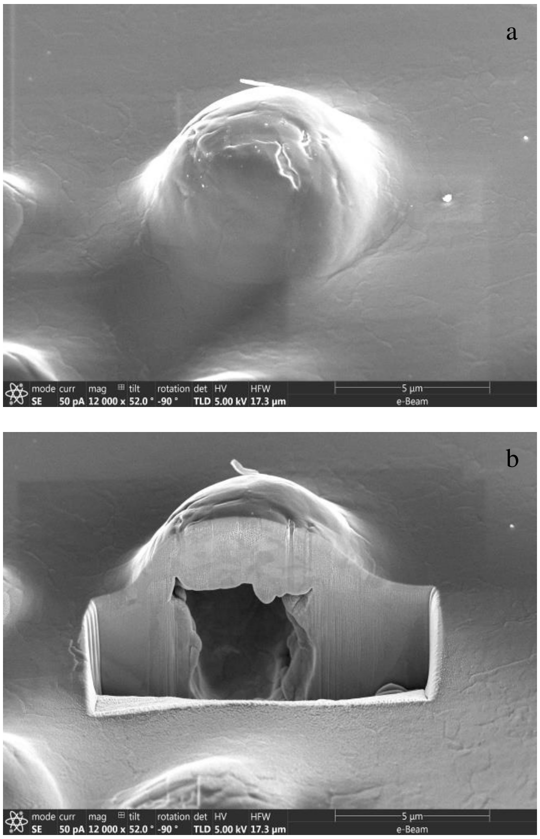

Figure 3: Images of the surface of 99.99996% coarse copper, which were taken on a FIB-SEM Helios G3 UC focused ion beam electron microscope. In (b) a blister specially cut by an ion beam is seen.

CONCLUSION

With the use of a CCD-camera and a remote microscope, the in situ observation of blistering of samples prepared from copper and tantalum was performed during their irradiation with a 2-MeV proton beam to a fluence of 6×10^{19} cm^{-2} (and in one case, a fluence that was 10-times larger). The sample temperature was measured by a thermistor, pyrometer and an infrared camera. The surface of the irradiated samples was investigated by means of an X-ray diffractometer, laser and electron microscopes.

It is found that among all copper samples studied, ultra-pure (99.99996%) fine-grained copper (Mitsubishi Materials Co., Japan) is most resistant to blistering. At a sample temperature of 150 °C, the blistering threshold is 3×10^{19} cm^{-2}. If this copper is used for preparing a neutron-producing target 10 cm in diameter for boron-neutron capture therapy of malignant tumors, then at a proton current of 10 mA, the target is resistant to blistering for 10 h. Since the planned therapy time is half an hour, the target can be applied in the therapy of several patients (approximately 20).

Ultra-pure (99.99996%) coarse-grained copper (Mitsubishi Materials Co., Japan) is two-times less resistant to blistering in comparison with ultra-pure fine copper. At a sample temperature of 150 °C, the blistering threshold is 1.5×10^{19} cm^{-2}. Since there were samples with different orientations of crystallites among the coarse copper samples studied, it is revealed that the crystallite orientation did not affect blistering.

M0 (State Standard GOST 859-2014, Russia) and OFC-1 JIS H3150 C1011 99.996% fine copper (SH Copper Products Co., Ltd., Japan) are seven-times less resistant to blistering in comparison with ultra-pure (99.99996%) fine copper. At a sample temperature of 150 °C, the blistering threshold is 0.45×10^{19} cm^{-2}.

The samples prepared by four different techniques of depositing tantalum on copper (explosion and diffusion welding, soldering and plasma arc deposition of tantalum and copper powders) are mechanically resistant to both stationary and pulsed thermal loads up to 1 kW/cm^2.

Tantalum is much more resistant to blistering than copper. The blistering threshold at 160 to 200 °C exceeds 6.7×10^{20} cm^{-2}. At a proton fluence of 3.6×10^{20} cm^{-2}, tantalum surface modification is observed in the form a relief (grid) with a cell size of about 1 µm.

It is established that during tantalum irradiation, the sample surface temperature increases, which may be due to a decrease in the thermal conductivity because of the formation of cavities inside tantalum and hydrogen incorporation into the tantalum crystal structure.

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