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Proton irradiation effects on beryllium – A macroscopic assessment☆

Nikolaos Simos [a,*], Mohamed Elbakhshwan [a], Zhong Zhong [b], Fernando Camino [c]

[a] Nuclear Sciences & Technology Department, Brookhaven National Laboratory, Upton, NY, 11973, USA
[b] Photon Sciences, NSLS II, Brookhaven National Laboratory, Upton, NY, 11973, USA
[c] Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY, 11973, USA

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ABSTRACT

Beryllium, due to its excellent neutron multiplication and moderation properties, in conjunction with its good thermal properties, is under consideration for use as plasma facing material in fusion reactors and as a very effective neutron reflector in fission reactors. While it is characterized by unique combination of structural, chemical, atomic number, and neutron absorption cross section it suffers, however, from irradiation generated transmutation gases such as helium and tritium which exhibit low solubility leading to supersaturation of the Be matrix and tend to precipitate into bubbles that coalesce and induce swelling and embrittlement thus degrading the metal and limiting its lifetime. Utilization of beryllium as a pion production low-Z target in high power proton accelerators has been sought both for its low Z and good thermal properties in an effort to mitigate thermos-mechanical shock that is expected to be induced under the multi-MW power demand.

To assess irradiation-induced changes in the thermal and mechanical properties of Beryllium, a study focusing on proton irradiation damage effects has been undertaken using 200 MeV protons from the Brookhaven National Laboratory Linac and followed by a multi-faceted post-irradiation analysis that included the thermal and volumetric stability of irradiated beryllium, the stress-strain behavior and its ductility loss as a function of proton fluence and the effects of proton irradiation on the microstructure using synchrotron X-ray diffraction. The mimicking of high temperature irradiation of Beryllium via high temperature annealing schemes has been conducted as part of the post-irradiation study. This paper focuses on the thermal stability and mechanical property changes of the proton irradiated beryllium and presents results of the macroscopic property changes of Beryllium deduced from thermal and mechanical tests.

1. Introduction

Beryllium, exhibiting excellent neutron multiplication and moderation properties in conjunction with its good thermal properties, is under consideration for use as plasma facing material in fusion reactors and as a very effective neutron reflector in fission reactors [1,2]. While its relatively low thermal neutron absorption cross section makes beryllium the material of choice for reactor reflectors, long term exposure to neutrons may result in gradual buildup of transmutation gases such as helium and hydrogen the buildup of which is linked to swelling and degradation of its mechanical properties. The irradiation generated transmutation gas products exhibit low solubility leading to supersaturation of the beryllium matrix and to eventual precipitation into bubbles that coalesce resulting in swelling and embrittlement, degradation of the metal properties and the limiting of its lifetime [3–6].

In addition to its consideration as plasma facing material in fusion reactors and as reflector in fission reactors beryllium has been also considered in hybrid spallation neutron source targets or as moderators/reflectors and as primary pion production target in a number of high power accelerator initiatives [7–13] where low-Z target material is sought for a desired pion spectrum. With the ever-increasing accelerator power required to support high-energy physics initiatives, the pool of materials forming production targets or other critical components capable to withstand the pulse intensities associated with these multi-MW class machines is depleted. MW-class accelerator targets, in addition to the high proton fluence they must endure during their operational life,

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* Corresponding author.
E-mail address: simos@bnl.gov (N. Simos).

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fluence that may approach those of reactor-based materials typically exposed to high neutron fluxes, must be able to continuously absorb and diffuse the severe thermo-mechanical shock resulting from the intense proton pulses. Tightly focused, high-energy proton pulses must be intercepted by the envisioned accelerator targets within several nanoseconds leading to extremely high deposited energy densities that are far beyond what most materials can tolerate. Densities of energy deposited under these conditions on the target as well as other beam-intercepting elements like collimators are estimated to be extremely high and far beyond what common materials, even those with extensive reactor-based track record, can tolerate. Further, the thermo-mechanical fatigue issue, direct result of the pulsed nature of these MW-class accelerators, adds additional performance requirements on these materials because of the need to extend the operational life of these components. Fatigue limits, even under the best of circumstances in which materials can resist irradiation-induced degradation of their properties, can be reached in short time stemming from the fact that accelerator pulse frequencies are of the order of 60 Hz. Therefore, utilization of beryllium as a pion production low-Z target in high power proton accelerators has been sought for two reasons namely (a) its low Z which desirable to produce a certain pion spectrum and (b) its good thermal properties that can be relied upon to mitigate the intense thermo-mechanical shock expected to be induced under the multi-MW power demand and the proton pulse structure.

Whether in a high energy neutron or energetic proton environments impinging protons and/or neutrons will knock out atoms in the beryllium crystal lattice generating defects, such as vacancies. Coalescence of vacancies leads to void formation and swelling adding to the swelling from the gaseous nuclear reaction products of He and H that are formed. A number of studies [3–6] have assessed that transmutation gas production plays a more important role than radiation-induced damage in the form of interstitial and vacancy formation in limiting the lifetime of the material. When irradiated at elevated temperatures, neutron or energetic proton irradiation is expected to cause He bubbles to form and embrittle grain boundaries. Studies [2] have also observed that beryllium irradiated at low temperatures (50 °C) but subsequently annealed to 750 °C exhibited significant swelling (~27%) attributed to large bubble formation. Embrittlement and helium swelling during low temperature irradiation according to studies [3–6,14] should be limited for beryllium.

Beryllium being a hexagonal close-packed structure has only two operating slip planes (0001) & (1010) significantly limiting the ductility of the metal [15]. For this reason important engineering properties such as fracture toughness, ultimate strength, and plastic elongation has been thought to be poor. It appears however that some of these properties, particularly ductility, are functions of fabrication process and controllability of grain boundary integrity which is associated with the removal of oxygen.

Several studies focusing on the properties of beryllium over a range of temperatures including irradiation effects from neutrons and in some cases energetic protons have been conducted in recent years. An excellent summary of unirradiated beryllium properties are presented by K. A. Walsh [16] where the physical, elastic and crystallographic properties as functions of temperature are described. The microstructure of out-of-pile annealed neutron irradiated beryllium was studied in Ref. [17] by X-ray tomography. Specifically, beryllium reflector fragments irradiated for 15 years in a research reactor at low temperatures were post-irradiation annealed and porosity was deduced after annealing using synchrotron X-ray micro-tomography. Studies of irradiated BeO have shown the recovery of the lattice parameters ***c*** and ***a*** [18].

This paper focusses on the effects of energetic protons and the damage they induce on Beryllium. Irradiation of beryllium samples that was one of the materials comprising a large array of candidate materials under study was performed at the Brookhaven National Laboratory Linac using 200 MeV protons. Following an exposure to peak proton fluence of ~1.2×10^{20} p/cm^2 the beryllium samples underwent post-irradiation evaluation focusing on the effects of irradiation damage on stress-strain characteristics, ductility and thermal expansion coefficient. In addition, and through high temperature annealing, the damage reversal as well as oxidation and transmutation gas mobilization and diffusion on volumetric changes were studied. X-ray diffraction studies on the effects of irradiation and macroscopic stress on the microstructural evolution and crystallographic role in the deformation of the beryllium were also conducted as part of the experimental campaign. This paper, however, presents only the effects of irradiation damage on the properties of beryllium observed macroscopically. The X-ray diffraction analysis findings of the damage induced by the 200 MeV protons on beryllium microstructure are presented in Ref. [19].

The experimental details and configuration of the beryllium sample array in the irradiating proton beam accompanied by estimated energy deposition and damage in the form of displacements-per-atom (DPA) are presented in the subsequent section. Estimates of energy deposition in the beryllium array are used to estimate through a 3-D thermo-mechanical analysis of the experiment in an effort to establish a good basis for comparison with studies by other researchers. The experimental details section is followed by the summary on the stress-strain behavior of the unirradiated Beryllium, the effects of the irradiation as a function of proton fluence and the role of the ***hcp*** crystal structure of Beryllium in the macroscopic elasto-plastic behavior. Subsequently, the effects of proton irradiation on the dimensional stability and the thermal expansion coefficient are presented and discussed along with the findings on volumetric changes and damage reversal via high temperature annealing. In the summary section the observations on proton irradiation effects on beryllium are summarized and discussed within the context of available data from similar or relevant studies.

2. Experimental

2.1. Microstructural characterization and sample preparation

Brush-Wellman standard grade of beryllium S-200-F produced through the consolidation of beryllium powder via vacuum hot pressing was obtained and used in the experiment in the form of ready samples that were specifically designed for the experiment. The fabrication process utilized and measurement or specification of the composition of the actual material was unavailable upon receipt of the produced samples. In addition, prohibitive regulations in effect at the home institution where the experiments were conducted associated the handling of beryllium, prevented microstructural and metallurgical analysis upon receipt of the material specimens. Microstructural studies, however, were performed on the received S200-F beryllium grade utilizing both scanning electron microscopy and EDS on unirradiated/unpolished samples and X-ray diffraction on irradiated and as received S200-F beryllium. Fig. 1(a; b; c and d) depict SEM micrographs of the unpolished surface of as-received beryllium obtained using the JOEL 7600F analytical high resolution electron microscope with the integrated INCA X-ray microanalysis system (EDX) at the Center of Functional Nanomaterials (CFN) revealing grain size, porosity and the presence of impurities at grain boundaries. Energy Dispersive Spectroscopy (EDS) analysis results are shown in Fig. 2 (a–d). The spectral analysis revealed that oxygen is the dominant impurity in the form of BeO, followed by carbon, aluminum and iron as well as traces of