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# Irradiation effects in beryllium exposed to high energy protons of the NuMI neutrino source

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ABSTRACT

A beryllium primary vacuum-to-air beam ‘window’ of the "Neutrinos at the Main Injector" (NuMI) beamline at Fermi National Accelerator Laboratory (Fermilab), Batavia, Illinois, USA, has been irradiated by 120 GeV protons over 7 years, with a maximum integrated fluence at the window centre of $2.06\ 10^{22}$ p/cm$^2$ corresponding to a radiation damage level of 0.48 dpa. The proton beam is pulsed at 0.5 Hz leading to an instantaneous temperature rise of 40 °C per pulse. The window is cooled by natural convection and is estimated to operate at an average of around 50 °C. The microstructure of this irradiated material was investigated by SEM/EBSD and Atom Probe Tomography, and compared to that of unirradiated regions of the beam window and that of stock material of the same PF-60 grade.

Microstructural investigations revealed a highly inhomogeneous distribution of impurity elements in both unirradiated and irradiated conditions. Impurities were mainly localised in precipitates, and as segregations at grain boundary and dislocation lines. Low levels of Fe, Cu, Ni, C and O were also found to be homogeneously distributed in the beryllium matrix. In the irradiated materials, up to 440 appm of Li, derived from transmutation of beryllium was homogeneously distributed in solution in the beryllium matrix.

## 1. Introduction

It is now recognized that materials degradation by radiation damage is one of the most challenging factors in the design and operation of next generation multi-megawatt high intensity proton accelerator facilities [1]. The existing database on materials response to radiation effects relevant to proton accelerator environments is very limited and this complicates material selection and lifetime predictions for beam windows and targets. An international research collaboration, Radiation Damage In Accelerator Target Environments (RaDIATE), was recently launched in order to explore radiation damage issues in different candidate materials under the relevant environment [2]. Beryllium is an excellent beam window material due to its high strength, low atomic number, low nuclear interaction cross-section and high melting point, and this paper reports the study of one such successful application. Beryllium is an alternative to graphite in applications where a low-Z particle production target material is appropriate (e.g. the NuMI pion production target). Consequently Be has been selected as one of very few candidates for beam windows and target components in a new generation of proton accelerator driven particle sources such as, for example, the Long Baseline Neutrino Facility (LBNF) [1,3,4], a higher power version of NuMI.

Industrial purity beryllium grades used in accelerator components contain a wide variety of trace impurities, typically oxygen, iron, aluminium, nickel, copper, silicon, carbon and magnesium. The majority of these impurities have very limited solubility in beryllium, and have strong tendencies to create precipitates and to segregate to defects such a grain boundaries or dislocations [5]. This may have significant deleterious effects on mechanical properties. For example:

1. Hard BeO particles play a role of stress concentrator sites and fracture origins, and the quantity, size and distribution of BeO particles can strongly affect the mechanical properties of beryllium [5]. Webster [6] also demonstrated that dispersed BeO

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particles control grain boundary migration and, consequently, grain size stability of beryllium.
2. Phases containing Fe and Al also have significant influence on mechanical properties of beryllium. X-ray diffraction studies of Jones and Weiner [7] of commercial beryllium after different heat treatments showed a link between the strain induced precipitation of the $Be_5(FeAl)$ phase and the decrease in tensile ductility and creep strength of the samples. Punni and Cox [8] studied corrosion properties of several beryllium grades and demonstrated that the Fe/Al/Be inclusions were the preferential sites for pit corrosion initiations.
3. Fe-Be-rich precipitates can lock dislocations and increase hardness of beryllium alloyed by iron, as was demonstrated by Morozumi and co-workers [9].

It is well-known that radiation damage, introduced by high-energy particles can change the quantity, size, spatial distribution and chemistry of phases present in the microstructure and, thus, the properties of exposed materials [10]. Although it is established that radiation generally leads to evolution of the properties of beryllium [11–13], the mechanisms underlying these changes are not well understood; for example, data about the stability of precipitates under radiation is very limited. No experimental data are currently available on phase stability under irradiation conditions relevant to beryllium components of particle accelerators (e.g. windows, targets).

Interactions of high-energy neutrons or protons with matter induce transmutation reactions. In the case of beryllium, most attention is currently paid to the gas transmutation products hydrogen and helium. Helium accumulation from either direct ion implantation or as a result of neutron-induced transmutation has been shown to result in significant changes in the properties of beryllium over a wide range of temperatures [14–17]. Such effects include those from the formation of nanosized helium bubbles, which is believed to be one of the main hardening mechanisms of beryllium irradiated at low temperatures [17]; additionally, helium assisted anisotropic swelling at low temperatures and helium bubble formation at grain boundaries at high temperatures may lead to grain boundary weakening and drastic loss of ductility [15,18]. The production rate of helium in beryllium under fission and fusion neutron irradiation conditions can be as high as 600 appm/dpa (displacements per atom) [19]. However, as presented here in section 3, in a high energy proton irradiation environment it can reach 4000 appm/dpa with thus a potential for even greater degradation of mechanical properties.

Non-gaseous transmutants can also alter the microstructure of irradiated materials and potentially lead to property degradation. This has been studied in a wide range of metallic alloy systems. For example, Ohnuki and co-workers observed that formation of Cr by transmutation reaction in V-alloys during irradiation in the High Flux Isotope Reactor (HFIR) caused very high embrittlement of the exposed alloys [20–22]. Works of Garner, Greenwood and Edwards have demonstrated that Mo-Re alloys strongly transmute to Mo-Re-Os-Ru-Tc system in a fast neutron spectra of the Fast Flux Test Facility (FFTF) that had serious consequences on the alloys phase stability. The transmutant osmium shows a strong tendency to co-segregate with Re under irradiation [23,24] potentially contributing to radiation induced embrittlement of the Mo-Re alloys [25]. Studies of ferritic-martensitic (FM) steel after mixed spectrum irradiation of high energy protons and spallation neutrons of the Swiss spallation neutron source (SINQ) showed that transmutants Ca, Sc and Ti extensively participate in the evolution of the microstructure through formation of radiation-induced clusters, segregation at the dislocation loops and alteration of the microchemistry of carbides [26], resulting in hardening and loss of ductility of the irradiated materials [27]; however, it was not possible to separate the influence of displacement damage, helium production and solid transmutant effects. To our knowledge, there are no data regarding the behaviour of solid transmutation products formed in beryllium. This work reports on the microstructure of industrial purity beryllium and changes induced by high-energy proton irradiation in the “Neutrinos at the Main Injector” (NuMI) beamline [28] at Fermilab, Batavia, Illinois.

## 2. Materials and techniques

### 2.1. Materials

The material used in this work is the industrial PF-60 beryllium grade. Its nominal chemical composition as specified by the manufacturer (Materion Electrofusion Corporation) is specified in Table 1 [29]; the main specified impurities are O (up to 2900appm), C (up to 450appm), N (up to 195 appm), Al (up to 165 appm), Fe (up to 130 appm) Si (up to 130 appm) and Mg (up to 81 appm).

A disk of PF-60 of 0.25 mm thickness served as a primary beam window in the NuMI beam line for 7 years from May 2005 till April 2012. It was exposed to a 120 GeV pulsed proton beam. The pulse frequency was 0.5 Hz, with a total pulse length of 9.78 µsec or 8.14 µsec for different operational modes, and with each pulse consisting of 6 or 5 batches respectively. Each batch was separated in time from the other by about 56 nsec, and consisted of 84 bunches with a spacing of 18.8 nsec. Each pulse delivers about $3 \times 10^{13}$ protons. The final integrated exposure of the window reached $1.57 \times 10^{21}$ protons. The beam had a Gaussian profile with the approximate size (as standard deviation of the distribution from the centre) $\sigma_x = \sigma_y = 1.1$ mm. Over the 7 years of operation, the position of the beam was changed several times for better adjustment to the position of targets and horns. These shifts were less than half of the beam sigma away from the central position for about 95% of total beam exposure. Although, the working temperature cycled with the pulsing of the proton beam, the average working temperature of the window was estimated to be approximately 50 °C over the last half of its operating period.

The displacement damage levels and concentrations of transmutation products were assessed using the Monte-Carlo MARS15 code [30–32]. The code calculates the number of displacements based on the NRT model [33] using an energy-dependent damage efficiency function (see Ref. [34] for details). The predicted maximum radiation damage level in the central part of the window was about 0.48 dpa, which, considering the beam position changes, decreases to the periphery following an almost Gaussian profile with $\sigma \approx 1.2$mm, giving a wide spectrum of radiation doses at almost the same temperature. The exposed area was experimentally localised using Gafchromic dosimetry film (type HD-V2) as shown in Fig. 1(b).

After irradiation, the central part of the beam window was punched out from its mounting flange. The applied stresses during this procedure caused deformation of the central part of the window and development of radial cracks as shown on Fig. 1(a). The sample remained in one piece overall, with no obvious fragmentation. The cracks were used for investigation of fracture behaviour of beryllium in the unirradiated areas and those exposed to varying levels of radiation damage.

For the investigation of non-irradiated beryllium, two complementary studies were made:

1. Material from outer areas of the NuMI window, at least 8 mm from the centre of the exposed area i.e. not exposed to proton irradiation, was investigated by atom probe mapping, S (TEM)/EDS and study of the fracture paths;