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Characterization of an explosively bonded aluminum proton beam window for the Spallation Neutron Source

David A. McClintock [a,*], Jim G. Janney [a], Chad M. Parish [b]

[a] Instrument and Source Design Division, Oak Ridge National Laboratory, P.O. Box 2008, Building 8600, MS 6466, Oak Ridge, TN 37831-6466, USA
[b] Materials Science and Technology Division, Oak Ridge National Laboratory, P.O. Box 2008, Building 4515, MS 6064, Oak Ridge, TN 37831-6064, USA

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ABSTRACT

An effort is underway at the Spallation Neutron Source (SNS) to redesign the 1st Generation high-nickel alloy proton beam window (PBW) to a 2nd Generation design that utilizes aluminum for the window material. One of the key challenges to implementation of an aluminum PBW at the SNS was selection of an appropriate joining method to bond an aluminum window to the stainless steel shielding structure of the PBW assembly. An explosively formed bond was selected as the most promising joining method for the aluminum PBW design and a testing campaign was conducted to evaluate the strength and efficacy of explosively formed bonds that were produced using two different interlayer materials: niobium and titanium. The characterization methods reported here include tensile testing, thermal-shock leak testing, optical microscopy, and advanced scanning electron microscopy. All tensile specimens examined failed in an aluminum interlayer and measured tensile strengths were all slightly greater than the native properties of the aluminum interlayer, while elongation values were all slightly lower. A leak developed in the test vessel with an niobium interlayer after repeated thermal-shock cycles, which was attributed to an extensive crack network that formed in an interfacial layer of a niobium-rich constituent phase located on the bond interfaces of the niobium interlayer; the test vessel with a titanium interlayer did not develop a leak under the conditions tested. Due to the experience gained from these characterizations, an explosively formed bond with a titanium interlayer was selected for the aluminum PBW design at the SNS.

1. Introduction

A proton beam window (PBW) is an integral component of an accelerator-based neutron source and serves an important function: to separate the ultra-high vacuum environment of the accelerator from the relatively low vacuum or atmospheric pressure environment of the target vessel region. Therefore, a PBW must be able to withstand (1) the thermal stresses caused by heating from the proton beam, (2) the mechanical stresses originating from the pressure imposed by the flow of cooling water, and (3) the pressure differential between ultra-high vacuum of the accelerator and the relatively low vacuum of the target core vessel. In addition to withstanding the stresses imposed during operation by the thermal and mechanical forces, the PBW material must be able to tolerate the radiation-induced changes in microstructure and mechanical properties, typically referred to as "radiation damage". Throughout the PBW lifetime the window material must maintain sufficient mechanical properties, including adequate ductility and fracture toughness, required to prevent failure during operation. The amount of radiation-induced change in mechanical properties of materials is frequently determined by characterizing the mechanical properties of material taken from components after removal from service in a radiation environment. In general, the selection of materials for service in high radiation environments is guided by mechanical properties data obtained from irradiated components. Therefore, a brief review of materials utilized in the past for PBW applications is prudent prior to a review of the aluminum SNS PBW design development.

A review of the current literature reveals that a variety of different materials have been utilized or considered for PBW applications in the past, including: martensitic steels T91 [1] and DIN 1.4926 [2]; precipitation-hardened nickel alloy Inconel® 718 (Alloy 718) [3,4]; and aluminum alloy $AlMg_3$ [5,6]. The experience with DIN 1.426 showed that it maintained high strength after irradiation but both uniform and total elongation values decreased to less than 50% of their initial values after less than 0.5 displacements per atom (dpa), and the failure mode transformed from ductile to brittle cleavage after 5.6 dpa [2]. Data obtained from tests on a precipitation-hardened (PH) Alloy 718 PBW irradiated at the Los Alamos

* Corresponding author. Tel.: +1 (865)576 5295; fax: +1 (865)576 2674.
E-mail address: mcclintockda@ornl.gov (D.A. McClintock).

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Neutron Science Center (LANSCE) indicate that the yield strength increased to a maximum around 2 dpa then steadily decreased with increasing dose, while total elongation values decreased from approximately 20% in the unirradiated condition to below 5% after less than 3 dpa [3,4]. Recent discussions with colleagues from the ISIS Spallation Neutron Source (Rutherford Laboratories, United Kingdom) indicate that the Alloy 718 beam entry window (BEW) that was recently removed from service at the ISIS First Target Station (FTS) received maximum doses greater than 34 dpa [7]. Despite reaching exceptionally high dpa dose levels, no operational issues were observed with the FTS BEW during 25 years of service, and replacement was done as a preventative measure during a larger performance upgrade project at ISIS [8]. Tensile data from the proton beam entrance region of the Swiss Spallation Neutron Source (SINQ) Target-3 $AlMg_3$ safety-hull showed that the solid-solution strengthened aluminum-magnesium alloy (similar to Al 5454) increased in strength, with yield strength increasing from ~75 MPa in the unirradiated condition to ~250 MPa at doses around 3 dpa, while maintaining 7–10% uniform elongation and failing in a ductile mode at doses greater than 3.5 dpa [6]. The successful use of aluminum in high radiation environments, including the SINQ safety-hulls, and the inherent neutronic performance benefits of aluminum relative to high-nickel alloys are the two primary reasons many accelerator-based neutron facilities now utilize aluminum PBWs, including Al 5083 windows now in operation at the ISIS Second Target Station (STS) [7] and the Japan Spallation Neutron Source (Japan Proton Accelerator Research Complex, Japan) [9].

During the engineering and design phase of the SNS facility aluminum was not widely used for PBW applications, rather corrosion-resistant high-nickel alloys were prevalent. Due to the successful use of Alloy 718 for PBW applications at the ISIS FTS [7] and the LANSCE accelerator facility (Los Alamos National Laboratory, USA) [3], the design of the 1st Generation PBW at the SNS utilized two curved windows composed of Alloy 718, shown in Fig. 1. The curved-window design permits the use of relatively thin window walls, relative to the thickness required for a flat-plate window, to counter the stresses caused by the differential pressure between the accelerator and target vessel, as well as the thermal stresses imposed by the proton-beam heating during operation. The window assembly in the 1st Generation PBW design is cooled by deionized water flowing between the two curved Alloy 718 walls; therefore maintaining the proper gap size between the two walls is critical to ensure adequate removal of the beam-induced heat generated in the window. The stringent dimensional requirements of the curved-window design necessitated complex fabrication techniques, including precise welds with minimal tolerance for thermal distortion, which increased both fabrication time and costs.

Despite the successful use of Alloy 718 as a beam window material at the LANCSE and ISIS facilities, engineers that designed the 1st Generation SNS PBW were concerned with the loss of ductility observed in precipitation-hardened Alloy 718 after irradiation [10,11,4]. Tensile characterization of PH Alloy 718 irradiated in a spallation environment revealed a dramatic loss of ductility occurs after only a few dpa and at 10 dpa the total elongation falls below 5% [11]. To address this concern, Alloy 718 in the solution-annealed (SA) condition was used for the 1st Generation SNS PBW, which has increased ductility relative to the precipitation-hardened condition and more than adequate strength for the 1st Generation PBW design. Tensile data from neutron irradiated Alloy 718 in the SA condition showed it experienced a decrease in ductility with increasing dpa dose, but maintained approximately 20% uniform elongation after 1.2 dpa, while PH Alloy 718 specimens irradiated to the same dose decreased to 1.5% [12]. Unfortunately, no data is available on the changes in mechanical properties of SA Alloy 718 after irradiation in a proton environment to dose levels prototypical the SNS PBW.

The uncertainty in the response of SA Alloy 718 to high dpa dose values led to establishment of an administrative maximum dose limit of 10 dpa for the 1st Generation PBW, which corresponds to ~7500 MW-h of total energy and has historically taken approximately 1½ years at current SNS operational power levels (850 kW–1 MW). At the time of publication, three 1st Generation PBW assemblies have been installed and removed at the SNS; all were operated without any issues and were removed from service upon reaching the 10 dpa administrative dose limit. But the high costs for fabrication and disposal of the Alloy 718 PBW assemblies and operational downtime required for PBW replacement motivated the development of a 2nd Generation SNS PBW that would utilize an aluminum window and have a longer lifetime.

The lifetime of the 2nd Generation SNS PBW will not be based on dpa level, but rather the gas production in the aluminum window. Helium (He) production is especially pronounced in aluminum alloys irradiated with high-energy protons and embrittlement from He has been attributed to a decrease of ductility observed in aluminum after irradiation in a spallation environment [6]. Since He embrittlement is expected to have the greatest detrimental effect on mechanical properties of aluminum, He gas production during operation was analyzed to establish a lifetime limit for the aluminum PBW window at the SNS. Operational experience with aluminum components at the SINQ Spallation Source (Paul Scherrer Institute, Switzerland) was used to determine an administrative limit for the SNS aluminum PBW. SINQ target safety-hull assemblies were fabricated from $AlMg_3$ and post irradiation measurements on material from the Target-3 safety-hull show He concentrations were approximately 1125 appm He [13], while estimates for later safety-hulls suggests as much as 2000 appm He was generated [14]. The SINQ safety-hulls performed as designed and no signs of component compromise were observed, therefore the 2000 appm He level was chosen as the basis for the aluminum SNS PBW lifetime limit. Based an administrative limit of 2000 appm and a He generation rate of 1220 appm He/year (2 MW operation for 5000 h/year) the SNS aluminum PBW will reach the limit after approximately 16,000 MW-h of operation, which corresponds to a maximum dose of 3.6 dpa. This limit may be too conservative and will likely be modified in the future as more information becomes available on He embrittlement effects in aluminum alloys and post irradiation examination of components removed from service are completed.

The specific alloy for the window was selected after a review of previous experience with neutron and proton irradiated aluminum alloys. The aluminum alloys most frequently used in radiation environments fall into two general alloy categories: solid-solution strengthened (Al-5xxx series) and precipitate strengthened (Al-6xxx series) alloys. Examinations of irradiated solid-solution strengthened $AlMg_3$ by Dai and Hamaguchi have shown that after irradiation in a mixed proton/spallation neutron environment small (~2 nm) and large helium (~6 nm) bubbles form in the interior of grains, while larger (15–30 nm) He bubbles align homogeneously along precipitate interfaces and grain boundaries [5]. The loss of ductility, observed as a decrease in total elongation, of $AlMg_3$ irradiated to greater than 2.7 dpa were believed to originate from the population of large He bubbles along the grain boundaries [6]. Early work on radiation-induced changes in microstructure of aluminum alloys showed that when irradiated with 800 MeV protons helium bubbles formed along grain boundaries of solid-solution strengthened alloy $AlMg_3$ (Al 5454), while no bubbles were observed in the precipitation hardened Al 6061 [15]. Additional transmission electron microscopy (TEM) examinations of Al 6061 after irradiation with 600 MeV protons by Victoria et al. showed that He bubbles formed along the Mg_2Si-type precipitates