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High-temperature annealing of proton irradiated beryllium – A dilatometry-based study☆

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ABSTRACT

S–200 F grade beryllium has been irradiated with 160 MeV protons up to 1.2 10^{20} cm^{-2} peak fluence and irradiation temperatures in the range of 100–200 °C. To address the effect of proton irradiation on dimensional stability, an important parameter in its consideration in fusion reactor applications, and to simulate high temperature irradiation conditions, multi-stage annealing using high precision dilatometry to temperatures up to 740 °C were conducted in air. X-ray diffraction studies were also performed to compliment the macroscopic thermal study and offer a microscopic view of the irradiation effects on the crystal lattice. The primary objective was to qualify the competing dimensional change processes occurring at elevated temperatures namely manufacturing defect annealing, lattice parameter recovery, transmutation ^{4}He and ^{3}H diffusion and swelling and oxidation kinetics. Further, quantification of the effect of irradiation dose and annealing temperature and duration on dimensional changes is sought. The study revealed the presence of manufacturing porosity in the beryllium grade, the oxidation acceleration effect of irradiation including the discontinuous character of oxidation advancement, the effect of annealing duration on the recovery of lattice parameters recovery and the triggering temperature for transmutation gas diffusion leading to swelling.

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. While the relatively low thermal neutron absorption cross section makes beryllium the material of choice for reactor reflectors long term exposure to neutrons results in gradual buildup of transmutation gases such as ^{4}He and ^{3}H and whose buildup is linked to swelling and degradation in its mechanical properties. The irradiation generated transmutation gases exhibit low solubility leading to super-saturation of the beryllium matrix and eventual precipitation into helium bubbles that coalesce inducing swelling. In addition to its consideration as plasma facing material in fusion reactors and as reflector in fission reactors beryllium has been also considered and evaluated as spallation neutron source target and as pion source target in a number of high power accelerator initiatives. A comprehensive summary of physical, mechanical and nuclear properties unirradiated beryllium properties are presented by K. A. Walsh [1] where the physical, elastic and crystallographic properties as functions of temperature are described.

To understand the effect of proton irradiation on dimensional stability, a crucial parameter in its consideration in fusion reactor applications, and to simulate high temperature irradiation conditions, multi-stage annealing using high precision dilatometry to temperatures up to 740 °C were conducted in air.

To that end S–200 F grade beryllium has been irradiated with energetic protons (160 MeV) from the Brookhaven National Laboratory Linac to 1.2 10^{20} cm^{-2} peak fluence and irradiation temperatures in the range of 100–200 °C followed by post-irradiation examination studies. X-ray diffraction studies were also carried out to compliment the macroscopic thermal study and offer a microscopic view of the irradiation effects on the beryllium crystal lattice.

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The overall objective was to qualify and quantify the competing dimensional change processes taking place at elevated temperatures namely manufacturing defect annealing, lattice parameter recovery, transmutation ^{4}He and ^{3}H diffusion and swelling and oxidation kinetics. The study revealed the presence of manufacturing porosity in the beryllium grade, the oxidation acceleration effect of irradiation including the discontinuous character of oxidation advancement, the effect of annealing duration on the recovery of lattice parameters recovery and the triggering temperature for transmutation gas diffusion leading to swelling.

High sensitivity dilatometry is a powerful technique to capture the dilatation of a sample as a function of temperature and time in a very precise manner. High resolution capabilities of the employed instrument can reveal phase transformations and other kinetics that are triggered by temperature. In this study a high precision LINSEIS dual rod dilatometer (sub-nm resolution) was used to study the response of beryllium samples, both irradiated and unirradiated for up to 740 °C temperatures in a multi-stage annealing thermal cycles with isotherms up to 10 h.

The objective was that annealing of beryllium samples irradiated at low temperature will provide an interesting method to mimic the effects of high temperature irradiation and also offer the means to study the kinetics of transmutation gas diffusion, swelling, lattice parameter recovery and oxidation. The effects of proton irradiation on a beryllium reflector on microstructural changes and evolution have been explored in other studies [2] as part of the effort to emulate the effects of neutron irradiation on beryllium. In Ref. [2] S200–F beryllium was irradiated with low energy protons (120 KeV) and fluence of 2.0×10^{18} ions/cm^2 at room temperature. It was assessed in Ref. [2] that due to proton irradiation voids preferentially developed at the interface between the beryllium matrix and the BeO and that the existence of BeO will accelerate the embrittlement and swelling of beryllium.

A number of studies have been conducted with interest in the dimensional stability affected by manufacturing defects, neutron irradiation and thermal annealing, oxidation as well as diffusion kinetics of transmutation gases.

The impact of impurities and in particular the role of manufacturing defects of manufacturing process of beryllium on the volumetric stability or the porosity state has captured the attention of studies [3] where S200–F beryllium grade was studied in its unirradiated state along with neutron irradiated S200-E and confirmed the presence of fabrication porosity and its evolution with temperature.

Leenaers et al. [4] performed post-neutron irradiation microstructural studies on S-200-E grade Be manufactured by vacuum hot-pressing that has been subjected to a temperature of approximately 50 °C and to a fission neutron fluence of $5.32 \cdot 10^{22}$ n/cm^2. Annealing was performed on the irradiated beryllium samples at 500, 750 and 900 °C for periods ranging between 50 h and three months. Swelling and porosity were evaluated and were correlated to the diffusion kinetics of ^{4}He and ^{3}H. Their microstructural analyses showed a clear evolution in bubble growth and He diffusion towards the grain boundaries where the bubbles coalesce. They assessed complete release of ^{4}He is at temperatures >750 °C resulting in a plateau of beryllium swelling. They further deduced that the microstructural changes are mainly temperature driven and observed that only at high annealing temperature the influence of the annealing time is important.

P. Vladimirov and co-workers [5] investigated beryllium reflector fragments irradiated for 15 years in the BR2 research reactor at temperatures below 120 °C and containing about 2 at. % helium was vacuum annealed at two temperatures and various annealing times. Gas-induced porosity developed as a result of annealing was investigated using synchrotron X-ray microtomography and important insights into the kinetics of the gas bubble growth. As pointed out above and in establishing the unirradiated reference for beryllium S200–F grade was used.

A. Khomutov et al. [6] summarized results on neutron-irradiated beryllium under consideration for fusion reactors (ITER). They conclude that after irradiation at 70 and 200 °C up to (1.4–3.9) 10^{22} cm^{-2}, swelling ranged from 0.2% to 1.5% and gradually increased with the neutron dose. The important conclusion reached in this study was that no strict dependence of swelling on irradiation temperature was observed. An important point with direct relevance to this study is made in their summary regarding the mobilization of transmutation gases that will be generated during proton irradiation (expected to be higher due to highly energetic protons as compared to neutrons). Mobilization of implanted tritium occurs in two ways. One is diffusion, which is slow; the other is a non-diffusive burst release. The potential for tritium “burst” mobilization from beryllium depends on temperature and microstructure, which itself depends on temperature, time at temperature, neutron damage/swelling and porosity. Normally, burst releases are not seen below 600 °C and occur following an annealing time of several hours. They note that at ITER operating temperatures, implanted tritium never diffuses deep into beryllium during a thermal excursion event, its mobility increases and some of the implanted tritium diffuses into the bulk of the material and becomes trapped. From the mobilization point of view, the neutron-bred tritium behaves as trapped tritium as well. At temperatures <500 °C trapped tritium is not mobilized from beryllium in any significant amount.

V.P. Chakin and co-workers [7] investigated neutron irradiation influence on the dimensional stability of beryllium by studying the radiation damage of the TE-56 beryllium grade manufactured by hot extrusion and irradiated in the SM reactor at the temperatures of 70 °C and 200 °C up to neutron fluence of (1.3–14.2) 10^{22} n/cm^2 ($E > 0.1$ MeV). Dimensional change measurements and X-ray analyses were conducted revealing that dimensional changes occurs a result of the superposition of two processes – radiation growth and anisotropic swelling. In addition, beryllium crystal lattice parameter changes as a function of neutron fluence were reported.

The oxidation kinetics of beryllium and its connection with temperature have been explored through a number of studies. In Ref. [1] a compiled summary of beryllium in gaseous atmospheres is presented that includes oxygen and air with experimental results from high-temperature oxidation of beryllium in air up to the melting temperature. It is reported that at a given temperature there exists a threshold time above which the initial low oxidation rate is followed by much higher rate. The threshold time reduces with increasing temperature. Also reported that epitaxial growth is observed between 300 and 400 °C while up to 600 °C randomly oriented beryllium oxide is observed followed by random growth between 600 and 800 °C. The type of beryllium plays a significant role in its resilience to oxidation.

C. Tomastik et al. [8] investigated the interaction of beryllium with air at elevated temperatures up to 600 °C on a microscopic level, using a high resolution Auger electron microscope. They assessed, based on their experimental observations, that diffusion of oxygen from the surface into the bulk has not detected up to 390 °C noting that a thin protective oxide film forms at 390 °C, while at 500 °C oxidation starts to enter into the grain boundaries. In their study they established that growth of the oxide thickness at 390 °C follows the square root of time while at the higher temperatures 500 °C and 600 °C the oxide growth is accelerated. In the present study, prompted by these threshold temperatures we evaluated the kinetics at 390 °C but also explored the 320 °C isotherm to investigate the volumetric change (if any) that might be noticeable at even lower temperatures in conjunction with