Available online at www.sciencedirect.com

ScienceDirect

Acta Materialia 88 (2015) 293–301

Acta MATERIALIA

www.elsevier.com/locate/actamat

Microstructure of out-of-pile annealed neutron irradiated beryllium studied by X-ray tomography

P. Vladimirov,[a,*] C. Ferrero,[b] V. Chakin,[a] P. Kurinskiy,[a] A. Moeslang,[a] R. Pieritz,[c] T. Weitkamp[d] and E. Brun[b,e]

[a]*Karlsruhe Institute of Technology, Karlsruhe, Germany*
[b]*European Synchrotron Radiation Facility, Grenoble, France*
[c]*ARS – Applied Research Solutions, Chirens, France*
[d]*Synchrotron SOLEIL, Gif-sur-Yvette, France*
[e]*Ludwig Maximilian University, Munich, Germany*

Received 5 August 2014; revised 11 January 2015; accepted 12 January 2015

Abstract—In fusion reactors, hydrogen isotopes, i.e., tritium and deuterium, will be used as fuel producing energetic neutrons and helium atoms. While deuterium is a stable nuclide frequent in nature, tritium is beta-radioactive. Its half-life time is about 12 years, requiring constant renewal via tritium generation in nuclear reactions. Neutrons produced in fusion reactions should therefore be effectively multiplied. To this end, beryllium is planned to be used in the form of pebble beds in the blanket of a fusion reactor. Unfortunately, helium and tritium are created under neutron irradiation and accumulated in the beryllium matrix, resulting in the formation of gas bubbles, swelling, and, under some circumstances, even in the loss of pebbles' structural integrity.

In this work, beryllium reflector fragments irradiated for 15 years in the research reactor BR2 (SCK-CEN, Mol, Belgium) 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 after annealing was investigated using synchrotron X-ray micro-tomography. This technique enables a non-destructive and quantitative analysis of the 3D morphology of the gas-induced porosity providing important insights into the kinetics of the gas bubble growth.

Using advanced post-processing of the micro-tomography data it has been possible to determine the volume fraction of gas bubbles and their size distribution, and reveal the formation of 3D bubble clusters. The importance of the data obtained for tritium release and relation between microstructure and tritium retention properties are discussed.

Keywords: Beryllium; Swelling; Neutron irradiation; Porosity; X-ray tomography

1. Introduction

The forthcoming generation of fusion reactors will use hydrogen isotopes, namely deuterium and tritium, as a fuel. Although deuterium is abundant in nature, the tritium isotope is beta-radioactive. It decays to ^{3}He with a half-life time of 12.3 years and is typically obtained in neutron-induced nuclear reactions. Therefore, fusion reactors are being designed to achieve closed fuel cycles: at least one tritium nucleus should be generated per each tritium burnt in a fusion reaction. Accounting for unavoidable neutron losses and instant tritium decay, the tritium breeding ratio should be about 10–12% higher than unity. In the Helium Cooled Pebble Bed (HCPB) design concept of a fusion reactor, beryllium in the form of pebble beds plays the role of a neutron multiplier which increases the tritium production per fusion neutron, thus closing the fuel cycle of the fusion reactor. Beryllium is known to be very effective in neutron multiplication, since, being hit by a high energy neutron, its nucleus often decays with production of two neutrons (and two helium atoms). The excess neutrons can be used for tritium generation on lithium ceramic pebbles, thus closing the fuel cycle.

Both tritium and helium are produced in beryllium under neutron irradiation by transmutation. Being practically insoluble in the beryllium matrix, helium atoms precipitate into the bulk as well as onto the grain boundaries and dislocations, resulting in helium bubble growth and substantial volume increase, i.e., swelling. Swelling depends both on the amount of helium accumulated in beryllium and on irradiation temperature. At temperatures below ~400 °C only very small (7–8 nm) facetted bubbles (if any) can be observed in a transmission electron microscope. At higher temperatures, the size of helium bubbles increases with the irradiation temperature, reaching, e.g., 140 nm at 695 °C [1]. Interconnected chains of bubbles are often

* Corresponding author.; e-mail: pavel.vladimirov@kit.edu

http://dx.doi.org/10.1016/j.actamat.2015.01.045
1359-6462/

observed on the grain boundaries and sometimes within the grain body.

Beryllium irradiated at low temperatures exhibits low swelling, but it can swell more than 30% after post-irradiation annealing [2]. Moreover, the degree of swelling can be controlled by selecting annealing temperature and dwell time. Therefore, to study the kinetics of beryllium swelling we used in this work annealing at two temperatures and various dwell times.

Tritium accumulated in helium bubbles determines mainly the radioactive inventory of the spent pebble beds, bearing the risk of abrupt release under accidental conditions. On the other hand, helium bubbles abundantly formed along grain boundaries trigger intergranular brittle fracture, destroying the structural integrity of pebbles. As it was shown by the gas thermal desorption experiments, the burst release of tritium and helium from the samples irradiated above 500 °C occurs simultaneously, which might be explained by the formation of pore channels connected with the sample surface [3,4]. As a consequence, it is important to study the process of helium bubble formation and evolution of complicated pore networks to substantiate this hypothesis and to obtain important insights into the mechanisms of tritium release from beryllium.

This evolution can be investigated using optical microscopy [5] or scanning electron microscopy performed on sample cuts. However, these methods provide a 2D cut through the 3D porosity network. Even the pore size distribution obtained on the sample cuts needs to be corrected (usually based on the assumption that pores are spherical) to reproduce their real radius distribution.

X-ray micro-tomography is nowadays a standard analytical method for the quantification of the porous structure of bulk samples [6–8]. For the investigation of porous materials composed of light elements, at length scales down to 1 μm, X-ray phase contrast as accessible at synchrotron radiation facilities presents substantial advantages [9–11]. Therefore, X-ray absorption micro-tomography provides a unique opportunity for non-destructive study of the 3D porosity network.

Since X-ray tomography cannot distinguish between gas-filled bubbles and voids, we will not explicitly distinguish them in the following sections and call them "pores".

2. Experimental details

2.1. Samples

The samples used in this study were made of commercial beryllium grade S200E (Materion Brush Beryllium & Composites, USA) used for about 15 years as a neutron reflector in the material testing nuclear reactor BR2, SCK-CEN, Mol, Belgium. Outer parts of the beryllium reflector were exposed to the temperature of cooling water (~50 °C), while the temperature inside the reflector block could be somewhat higher (<120 °C). This long-term irradiation resulted in a fission neutron fluence of 5.32×10^{22} n/cm^2 which corresponds to the generation of approximately 22,500 appm He [2]. Tritium was also produced under neutron irradiation, however, due to a complex operation history it is difficult to calculate exactly its amount.

The S200E material, which utilizes attritioned powder (<200 μm), was consolidated by vacuum hot pressing (VHP). Typically, VHP is performed at temperatures in the range 1050–1150 °C and under a pressure of about 14 MPa during 36 h. Like all other beryllium powder products, S200E contains coarsened BeO particles (0.1–0.5 μm) heterogeneously distributed along grain boundaries [12]. The oxide particles originate from the oxidation of raw powder [13]. The average grain size is about 10–13 μm. Unfortunately, this material was not available in the unirradiated state, therefore we used similar grade S200F as a reference.

For X-ray micro-tomography measurements, the irregularly shaped pieces with a size of about 0.5 mm were produced by crashing neutron reflector fragments. The samples were vacuum-annealed at 850 °C and 1000 °C, respectively, for 0.5, 1, 5 and 10 h.

2.2. X-ray measurements

The beam line ID19 [14] at the European Synchrotron (ESRF), Grenoble, France was used to acquire X-ray tomography data, which were subsequently processed to produce 3D images of the investigated samples.

Ten beryllium samples were scanned using the full-field parallel-beam in-line phase contrast micro-tomography setup at the ESRF with variable sample-to-detector distances (15, 10 and 8 mm) and three effective voxel sizes (see Table 1). The energy was set to 17.4 keV. The imaging detector used at the beam line was the chip of a CCD sensor based on the FReLoN design developed at the ESRF [15], with an array of 2048 × 2048 pixels. The physical pixel size of the CCD is 14 μm. Different detector optics are available covering a range of effective pixel sizes from 0.18 to 30 μm. In our experiment the samples were imaged at effective pixel sizes of 1.4, ~0.3 and ~0.2 μm. The detector system consists of a thin single-crystal scintillator screen converting the X-ray intensity distribution to a visible-light image which is then magnified and projected onto the CCD via commercially-available microscope lenses. The sample stage is equipped with motors allowing horizontal translation of the sample in both dimensions, in addition to the tomographic rotation around the vertical axis.

For safety reasons, because of the radioactivity of the sample, each specimen was enclosed in a Plexiglas cylinder with double walls during measurements. To prevent unwanted motion of the samples during rotation we used a spring-loaded piston placed in the inner cylinder which pushed onto the sample. The glue used for this purpose in the previous experiments [4] can penetrate a specimen through a pore network and should be avoided. Double confinement appears to be very useful, as after several consequent measurements the inner container broke under simultaneous action of irradiation and stress. Thus, only a subset of scheduled measurements could be accomplished. Table 1 summarizes the successful measurements performed during the experiment.

2.3. Post processing and topological analyses

A substantial amount of data (~1 TB) was collected during the experiments requiring advanced processing according to the following major steps: raw data assembling, pre-processing of raw data, volume decomposition, tomographic reconstruction, and post-processing [16]. The processed data are in the form of 2D gray scale images,