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Gas bubble network formation in irradiated beryllium pebbles monitored by X-ray microtomography

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ABSTRACT

The effective and safe operation of helium cooled ceramic breeder blankets with beryllium as a neutron multiplier requires among others an efficient tritium release. A micrometric resolution computer aided microtomography (CMT) setup located at the European Synchrotron Radiation Facility made possible the 3D reconstruction of interconnected channel networks of helium bubbles in beryllium pebbles, thus enabling the identification of open porosities in the micrometer range. Beryllium pebbles of 2 mm diameter were neutron irradiated at 770 K to a fluence of $1.24 \times 10^{25}\ \mathrm{nm}^{-2}$, resulting in 480 appm helium and 12 appm tritium. After annealing at 1500 K, CMT was performed on the pebbles at 4.9 and 1.4 μm spatial resolution, respectively, followed by the post-processing of the reconstructed pebble volumes. Besides a bimodal pore distribution with a smaller population around 10 μm diameter and a high density of partly interconnected pores around 40 μm diameter, a swelling of 17% was found. The spatial distribution of the void fraction network will be discussed together with implications on tritium release behaviour.

1. Introduction

A key issue of Helium Cooled Pebble Bed (HCPB) blankets is its behaviour under fusion neutron irradiation. In the present European HCPB blanket concept and within a 40000 h lifetime, an integral neutron dose of typically $3 \times 10^{26}\ \mathrm{nm}^{-2}\,\mathrm{s}^{-1}$ ($E_n > 1$ MeV) results in the production of about 80 dpa, 25700 appm helium, and typically 640 appm tritium in beryllium [1]. Depending on the local neutron spectrum, the helium/tritium total yield ratio can vary between 10 and 100. According to present HCPB blanket designs an amount of about 310 tons metallic beryllium is foreseen as a neutron multiplier in form of layers of small pebbles. The knowledge of tritium and helium accumulation as well as the gas release kinetics from the pebbles is crucial for the reliable and safe operation of fusion DEMO reactors.

While even large amounts of about 20000 appm helium accumulated in beryllium used as a reflector material in the mixed spectrum reactor BR2 (Mol, Belgium) do not produce bubbles visible via TEM analysis after low temperature neutron irradiation, experiments encompassing post-irradiation temperature ramps revealed pronounced ^{4}He and ^{3}H release peaks [2,3], accompanied by a substantial increase of swelling. Significant swelling and creep were also found by other authors after high dose neutron irradiation at high temperatures or after low temperature irradiation followed by higher temperature annealing [4–7]. A combination of microstructural analysis and gas release measurements supports the evidence that at high temperatures the tritium inventory is concentrated either in helium bubbles or trapped in strain fields in the bubbles' vicinity and can be substantially released only together with helium, i.e. by the formation of open porosity networks often along grain interfaces followed by bubble venting. A holistic 3D view on bubble formation, growth, coalescence and network formation is critically important for the experimental validation of recently started multiscale modelling activities aiming to understand helium and tritium kinetics and predict tritium release characteristics at different irradiation temperatures and neutron doses.

While classical 2D microstructural analysis techniques do not actually suffice to carry out morphologic analyses of extended bubble networks and complete gas percolation paths, X-ray based tomography techniques have substantially enhanced their resolution limit to the micrometer and even sub-micrometer range during the past years. 3D computer aided microtomography (CMT) has already shown its capability in the non-destructive analysis of the packing factor of beryllium beds [8,9] with a spatial resolution of 10 μm, and its potential in analysing 3D porosity networks in irradiated beryllium [10]. In the present study, the initial results reported in Ref. [10] are extended. The analysis includes now (i) the whole reconstructed volume of irradiated and unirradiated beryllium pebbles, (ii) 3D rendering of surface and porosity channels, and (iii) analysis of the open-to-total-porosity ratio.

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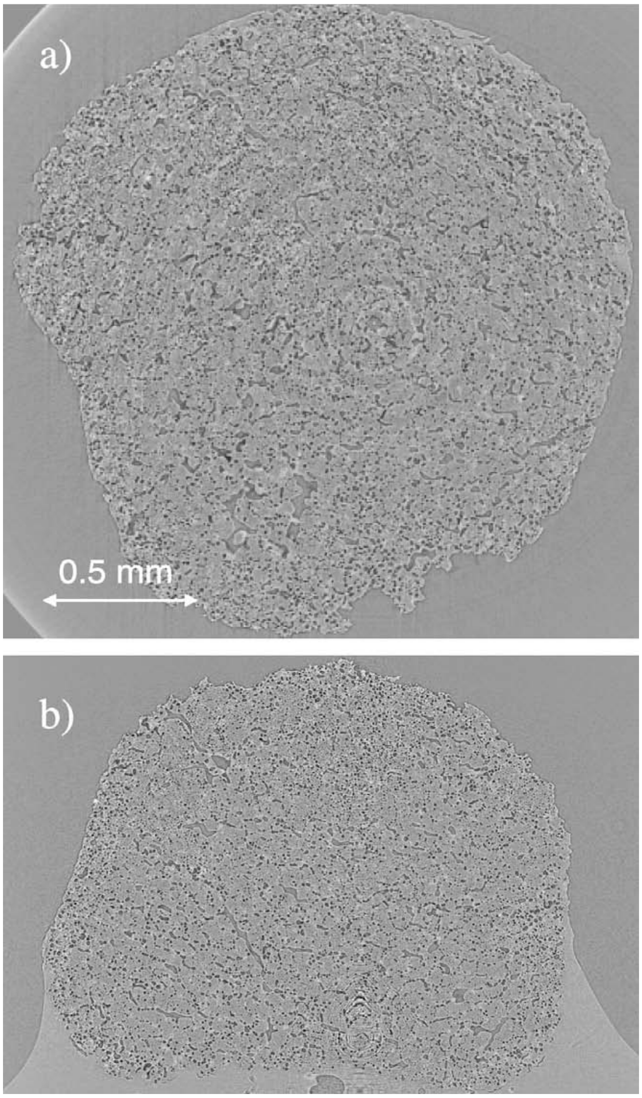

Fig. 1. Horizontal (a) and vertical (b) CMT cross sections of a Be pebble after neutron irradiation at 770 K and annealing at 1500 K. The spatial resolution is 1.4 μm.

2. Experimental

Beryllium pebbles with a diameter of 2 mm and grain sizes between 40 and 150 μm were produced by Brush-Wellmann, USA, and were irradiated in the BERYLLIUM experiment at the High Flux Reactor (Petten, The Netherlands) at 770 K to a fast neutron fluence of 1.24×10^{25} nm^{-2} ($E_n > 0.1$ MeV), producing 480 appm ^{4}He and 12 appm ^{3}H. After irradiation, the pebbles were tempered at ~1500 K, just below the beryllium melting point (1556 K), since a pronounced gas release peak was found to start at about this temperature in thermal ramping experiments [2]. For safety reasons both the non-irradiated and the irradiated samples were embedded in a double Plexiglas cylindrical capsule of about 6 mm external diameter and glued on the very bottom of it to avoid sample jiggling during the CMT rotational scans.

The high resolution microtomography setup at the ID19 beamline of the European Synchrotron Radiation Facility, Grenoble, France, was used with a monochromatic X-ray beam of 7 keV. The entire beryllium pebbles were scanned with two different spatial resolutions: 1.4 μm to reveal also single bubbles and 4.9 μm to focus on bubble network formation and percolation paths. For the post-processing of the as acquired microtomography data, a filtered back projection reconstruction program written at the ESRF was deployed.

3. Results and discussion

The CMT analyses were performed (i) on six unirradiated Be pebbles at 4.9 μm spatial resolution, and (ii) on two irradiated specimens at 4.9 and 1.4 μm resolution, respectively. Fig. 1 shows a horizontal (a) and a vertical (b) cross section of an irradiated specimen, recorded at 1.4 μm 'voxel' resolution. While still a quite high fraction of smaller, partly isolated bubbles can be observed, the post-processing reveals a high density of large bubbles which are in a vast majority interconnected through percolation paths to the pebble surface. Due to insufficient contrast between grey levels, the smaller bubbles are sometimes hardly visible. The observation that even after high temperature annealing at 1500 K still closed porosities are visible supports the main outcome of the temperature ramping experiments [2], that a small fraction of helium is not released before the melting

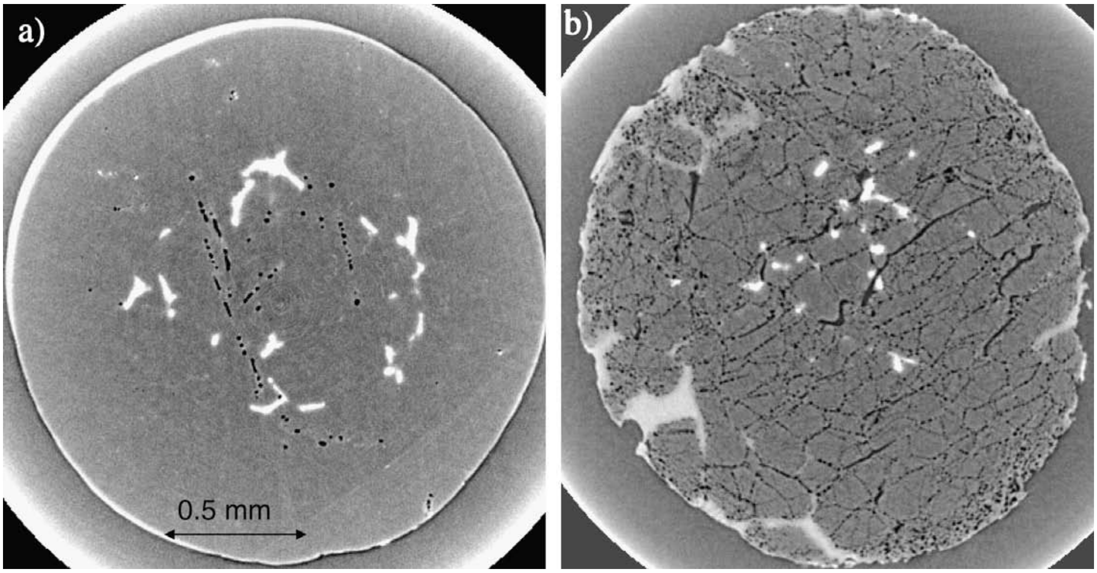

Fig. 2. Horizontal CMT cross sections of a Be pebble before (a) and after (b) neutron irradiation at 770 K and annealing at 1500 K. The spatial resolution is 4.9 μm.

temperature is reached. Actually, a moderate gas concentration of only 480 appm helium and 12 appm tritium results after 1500 K annealing in an advanced stage of bubble coalescence and network formation with extended percolation paths. It can be expected that after high neutron dose irradiation, i.e. end-of-lifetime conditions, similar bubble coalescence and network formation kinetics might be observed not only after high temperature annealing but already at blanket relevant irradiation temperatures, assuming comparable grain sizes, impurity contents and dislocation densities. Fig. 2(a) shows a 4.9 μm resolution horizontal cross section of a non-irradiated beryllium pebble before irradiation, revealing typical pores arising from the rapid condensation of the pebble during its fabrication process. The condensation induced voids having diameters ranging between 9 and 18 μm and a total volume fraction of 0.17%. The CMT image of an irradiated and annealed beryllium pebble is shown in Fig. 2(b), highlighting pronounced bubble networks with chain-like percolation paths. The typical distances between the percolation paths as well as their pattern suggest that they have been formed primarily along the grain interfaces. According to the CMT analysis the average diameter of the bubbles in the networks corresponds to the average width of the percolation channels, which is in the vast majority of the cases comprised in a narrow range around 39 μm. The mean volume fraction of the irradiation and annealing induced voids is 14.0%, and the void fraction surface within an entire pebble amounts to typically $2.1 \times 10^{-4}\,\mathrm{m}^{-2}$. Thermal annealing after a relatively short neutron irradiation (480 appm helium and 12 appm tritium) causes swelling which is almost two orders of magnitude above the fabrication induced porosity. The white and bright grey areas in Fig 2(a) and (b) are the signature of impurities present in the beryllium matrix and are featuring different material densities. The 3D rendering of an entire irradiated and annealed pebble is shown in Fig. 3 as a superposition of the surface contour and the internal complex bubble network. The surface transparency was chosen to be 30%. Finally, Fig. 4 shows a sub-volume (32 × 32 × 64 voxel) extracted from the centre of an irradiated and annealed pebble to illustrate the complex, interconnected bubble network. The technique used to visualise this volume element is the 'marching cube', applied directly to the binary data. The relatively high fraction of open-to-total-porosity confirms earlier results from temperature annealing experiments. In particular, it appears that most of the helium and consequently also of the tritium content has already escaped from the beryllium after annealing around 1500 K.

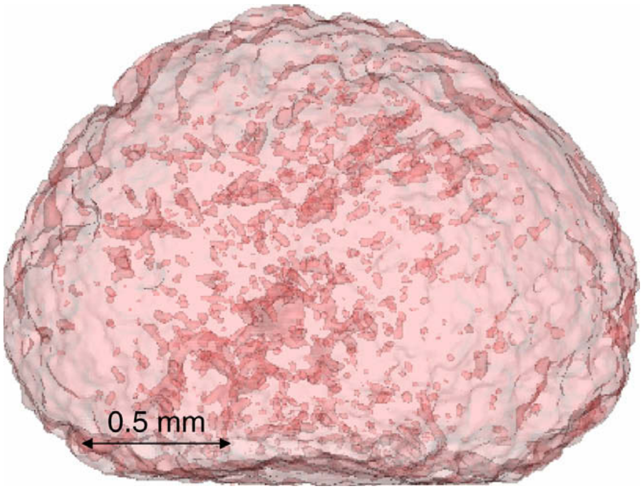

Fig. 3. Neutron irradiated (770 K, 480 appm He, 12 appm ^{3}H) and annealed (1500 K) Be pebble showing a superposition of the pebble surface and the volume porosity.

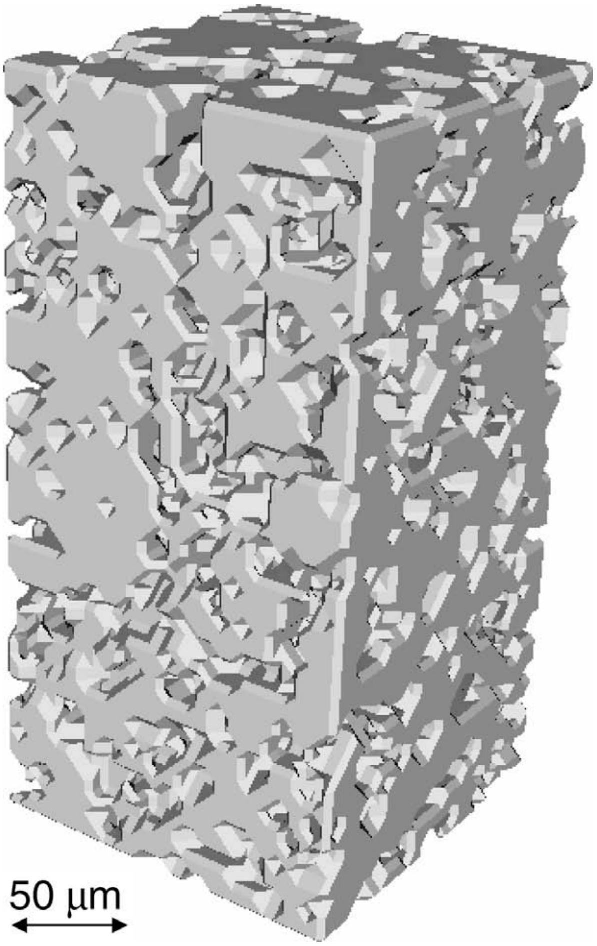

Fig. 4. A rectangular sub-volume extracted from an irradiated Be pebble showing the reconstruction of the complex bubble network (32 × 32 × 64 voxel).

4. Conclusion

High resolution synchrotron radiation CMT has been applied successfully to analysing the complex mechanisms of bubble network formation and gas percolation in irradiated ($T_{\mathrm{irr}} = 770$ K, 480 appm He, 12 appm ^{3}H) and annealed (1500 K) beryllium pebbles. The CMT post-processing capability represents a powerful tool to analyse in a holistic manner not only the bubble sizes and densities or the channel network topology but allows also the extraction of structural parameters that are not accessible from TEM, like the 3D reconstruction of the fraction of interconnected bubbles, or the determination of the open-to-closed-porosity ratio. This non-destructive imaging technique has proven to be an essential instrument not only for the experimental validation of multiscale modelling of gas release kinetics but also for the evaluation of the structural integrity of entire pebble bed assemblies. For a given microstructure it is reasonable to expect that bubble network formation and percolation follow similar kinetics in Helium Cooled Pebble Bed blankets at higher neutron doses but lower irradiation temperatures.

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