

## **In-situ TEM investigation of nano-scale helium bubble evolution in tantalum-doped tungsten at 800 °C**

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# In-situ TEM investigation of nano-scale helium bubble evolution in tantalum-doped tungsten at 800 °C

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## Abstract

The aim of this work is to probe the helium induced defect production and accumulation in 40 keV He<sup>+</sup> irradiated polycrystalline W and its alternative alloy W-5wt.%Ta using transmission electron microscopy (TEM) combined with in-situ helium irradiation at 800°C. A maximum damage level of 1 dpa with a maximum He-to-dpa ratio of 5.5 at%/dpa has been reached in this work for both materials, which corresponds to an ion fluence of  $7.33 \times 10^{16}$  He<sup>+</sup>/cm<sup>2</sup>. The presence of radiation-induced dislocation loops was not observed at this temperature. The low density of the incipient bubbles in W has been already detected at 0.004 dpa, which corresponds to a fluence of  $3.3 \times 10^{14}$  He<sup>+</sup>/cm<sup>2</sup>. The experiments conducted at 800 °C have shown that the addition of 5wt.% of tantalum into tungsten may diminish the binding of He ions with vacancies into complexes, which serve as the core of the bubble, thus hindering helium bubble formation below 0.02 dpa and their further growth and population at higher damage levels. By exceeding the damage dose  $\geq 0.3$  dpa, a progressive transition from a spherical to a faceted shape of the bubbles has been observed in W but not in the W-5Ta alloy. At 1 dpa, >80% of the bubbles in W were of the faceted type with the facet planes of {110}.

**Keywords:** W/W-5Ta; fusion materials; in-situ helium exposure; faceted helium defects; transmission electron microscopy; bubble detection.

## Introduction

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Tungsten and its engineered alloys are the preferred choice as armour material for plasma-facing components (PFCs) by the nuclear fusion community, and therefore lie at the heart of the international roadmap for the realisation and deployment of fusion reactor technology [1, 2]. In ITER, the divertor design includes W monoblocks in the inner and outer vertical targets in each water-cooled cassette assembly [3]. The divertor itself is intended to serve as the plasma power exhaust, particle control (deuterium/tritium (DT) and He repumping), also functioning as a screen for impurities, reducing contamination [4]. The peak power flux density values for the ITER design range from  $8 \text{ MW m}^{-2}$  at most in the non-nuclear phases (H/He discharges) to  $10 \text{ MWm}^{-2}$  in the nuclear phases (DT burning discharges), corresponding to W surface temperatures of  $\sim 800 \text{ }^\circ\text{C}$  and  $\sim 1100 \text{ }^\circ\text{C}$  respectively [5]. This may increase to as much as  $20 \text{ MWm}^{-2}$  during transient events at the monoblock surface [6]. Radiation-induced damage levels in ITER divertor monoblocks are predicted to be  $\sim 0.1 \text{ dpa}$  after a four-year ITER nuclear operation phase, prior to cassette replacement of the first divertor, and  $\sim 0.5 \text{ dpa}$  for permanent divertor components during ITER's end-of-life operation [6].

An ITER-like divertor design for the future fusion demonstration power plant (DEMO) is being considered [7], alongside alternative configurations able to handle the more demanding radiation environment in the inner target of the divertor [8]. Besides this, DEMO is envisaged to comprise a first wall with a W armour joint to a reduced-activation ferritic/martensitic (RAFM) steel [9, 10], with predicted surface heat fluxes of  $0.5\text{-}1.2 \text{ MWm}^{-2}$  [10, 11]. The expected lowest shield temperatures for W armour materials range from  $\sim 500 \text{ }^\circ\text{C}$  in the first wall up to  $>800\text{-}900 \text{ }^\circ\text{C}$  in proposed He-cooled divertor designs and  $>1700 \text{ }^\circ\text{C}$  in the divertor armour surface [12], whereas neutron bombardment is

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estimated to cause ~4 dpa in the W armour at the end-of-life of W-Cu plasma-facing components in DEMO [13].

Tungsten is characterised by a relatively low ductile-to-brittle-temperature (DBTT), namely ~200-400 °C [14-17], which can increase progressively with damage level due to the radiation fields inside the fusion reactor core up to ~800-1000 °C [18, 19]. Thermal cycling across the DBTT from temperatures between 70-120 °C up to the maximum surface temperature in W mock-ups during High-Heat-Flux (HHF) tests does not induce the formation of macro-cracks. However, macro-cracks are observed at a heat load of 20 MWm<sup>-2</sup> with the surface temperature approaching 2000 °C [3]. The highest temperature for safe operation of W components is limited by the resistance to creep, low-cycle fatigue, helium embrittlement and recrystallization [20, 21]. Mechanical test data at 800 °C constitute a representative property database for W monoblocks that is compared to their performance under HHF tests and is also used as key input for the universal slope method aimed at correlating tensile properties and fatigue performance of W monoblocks [3]. Plasma exposure of W-based PFCs causes radiation-induced lattice damage, surface sputtering, hydrogen isotope retention and He accumulation and diffusion into the bulk structure [22]. The latter effect can lead to bubble formation and potential blistering and, as a consequence, embrittlement at low temperatures and ductility loss at high temperatures [22]. Additionally, W exposure to He ions in the eV energy regime (>20-30 eV), where physical sputtering does not play a significant role in changes to surface morphology, at temperatures >700 °C and a minimal He fluence of ~10<sup>25</sup> m<sup>-2</sup> induces the formation of ‘nanostructured or fuzzy W’ on the material surface [5, 23, 24].

In this work we have applied extensive in-situ damage and microstructural analysis techniques to observe W and W-5wt.%Ta (W-5Ta) materials at 800 °C and exposed to 40 keV He<sup>+</sup> irradiations at incremental damage levels from only 0.004 dpa up to 1 dpa with a

1 maximum He-to-dpa ratio of 5.5 at%/dpa. Tantalum alloying is one of the proposed  
2 engineering solutions to overcome the inherent drawbacks of W as a structural material, since  
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4 Ta is reported to shift the onset of W recrystallization [25-28]. It has also been shown to  
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6 reduce the vacancy mobility and to consequently delay void formation at 800 °C under  
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8 proton irradiation [29]. In W-5Ta, voids were not found up to a damage level of 0.3 dpa,  
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10 however, the excess of free vacancies present in the W-5Ta irradiated at 800°C led to the  
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12 formation of visible voids in TEM study after post-irradiation annealing of the sample at  
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14 1000 °C. At lower temperatures the presence of Ta also retards the mobility of self-interstitial  
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16 atoms (SIAs) and interstitial  $a/2\langle 111 \rangle$  dislocation loops under irradiation at lower  
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18 temperatures, and consequently the loop growth and coalescence, attaining saturation in loop  
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20 dimensions at relatively low damage levels [30, 31]. Ta is also observed to hinder the surface  
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22 blistering in W under high-fluence deuterium plasma irradiation [32]. In addition, dual beam  
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24 and sequential irradiation experiments have shown to shift the fluence threshold for fuzz  
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26 formation higher in W-Ta alloys at 950°C as the ratio of  $D^+/He^+$  ions increases due to a  
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28 significant D de-trapping at that temperature which led to the fast diffusion and escape of the  
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30 implanted species [33]. The main research objective is to demonstrate the effect of the  
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32 presence of Ta solute atoms in W-Ta binary system on response to radiation under  
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34 cumulative helium beam exposure at an elevated temperature of 800 °C which remains  
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36 largely unexplored. The results obtained in this work are important to the fusion community  
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38 helping to expand the pallet of candidate structural materials for future fusion devices that are  
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40 more resistant to  $He^+$  induced surface damage. This data will also help to validate predictive  
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42 models of the expected in-service degradation of fusion construction materials, so that more  
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44 advanced alloy compositions with optimised properties can be designed and developed for  
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46 the fusion reactors.  
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## Materials

The initial 1 mm-thick W sheet (99.95%) was provided by Goodfellow Cambridge Ltd. The as-received W material was annealed in vacuum at 1400 °C for 2 h for recrystallization. The W-5Ta alloy was instead produced by Plansee AG via powder metallurgy. W-5Ta was double forged and then annealed at 1600 °C for 1 h [34]. After delivery, the material was annealed for 1 h at 1000 °C for degassing [35]. Afterwards, smaller samples of  $2 \times 2 \text{ cm}^2$  were cut and also annealed at 1400 °C for 2 h to remove defects introduced during machining. After annealing, both W and W-5Ta materials were pre-thinned down to 80-100  $\mu\text{m}$  in thickness using SiC abrasive papers from grit 220 up to 4000. TEM discs were then punched and electropolished at -5 °C using a Struers Tenupol-5 unit and an electrolyte comprising an aqueous solution of 0.5 wt.%  $\text{Na}_2\text{S}$  in the case of W, whereas a mixture of 15 vol.%  $\text{H}_2\text{SO}_4$  (95%) and 85 vol.%  $\text{CH}_3\text{OH}$  was employed for W-5Ta. The average grain size was  $3.9 \pm 0.8\text{mm}$  (W) and  $2.3 \pm 0.7\text{mm}$  (W-5Ta) [29, 31].

## Experimental

The samples were exposed sequentially to a 40 keV  $\text{He}^+$  beam at the temperature of 800 °C, using the MIAMI-2 TEM/ion accelerator system located at the University of Huddersfield [36]. The in-situ TEM facility under ion irradiation is composed of a 300 kV Hitachi H-9500 Transmission Electron Microscope (TEM), coupled to a 350 kV NEC ion accelerator incorporating a Danfysik 921A ion source. The ion beam is incident on the sample at an angle of  $18.7^\circ$  to the electron beam in the TEM. Sample heating was achieved by a double-tilt heating holder (Gatan Model 652) that uses a current flow through a Ta furnace surrounding the sample. The temperature was measured via a thermocouple attached to the furnace. The sample was held in place using a Hexring® clamping mechanism to ensure good thermal contact between the sample and the furnace. On reaching the target temperature, the sample was irradiated by the  $\text{He}^+$  beam in a series of steps from the lowest

1 damage level of 0.004 dpa up to 1 dpa at a rate of  $4.5 \times 10^{-4}$  dpa/s, corresponding to a He<sup>+</sup>  
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3 fluence of  $3.3 \times 10^{14}$  and  $7.3 \times 10^{16}$  He<sup>+</sup>/cm<sup>2</sup> respectively.  
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5 The simulated damage profile and the Helium concentration profile (Fig.1) were  
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7 calculated using the *Stopping and Range of Ions in Matter* (SRIM-13) software with the  
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9 quick Kinchin-Pease approach [37-39], using an average displacement energy of 90 eV [40]  
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11 and default values for other software settings. The total current deposited on the sample was  
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13 0.09 nA, with a He<sup>+</sup> flux of  $3.3 \times 10^{13}$  ions/cm<sup>2</sup>/s. It was predicted that over 50% of helium  
14  
15 ions was transmitted through the foil, and 32% was implanted. The damage level was  
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17 estimated as the average value in the foil thickness of  $\sim 100 \pm 10\%$  nm, as determined by  
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19 Convergent Electron Beam Diffraction (CBED) prior to the experiment. Micrographs were  
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21 recorded along the zone axes <001> and <111> during the stepwise increase in He fluence,  
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23 starting at the lowest damage level of only 0.004 dpa. At damage steps representative of the  
24  
25 microstructure evolution, a through-focal series of micrographs was taken with a defocus  
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27 value  $\Delta f \leq 1 \mu\text{m}$  in order to assess the presence of He bubbles  $\geq 1$  nm in diameter, based on the  
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29 “out-of-focus” Fresnel imaging technique [41].  
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### 37 **Helium bubble detection**

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39 In order to determine the number and size of the bubbles in the image, the  
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41 topologically structured contour detection algorithm was applied from [42] as implemented in  
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43 the OpenCV Library [43]. Furthermore, a graphical user interface was developed in C++  
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45 using Qt Creator as a software development kit. The methodology for detection consisted of  
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47 three steps: generation of a binary image, contour estimation and area calculation.  
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51 The original TEM micrograph taken in under-focus condition was processed first  
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53 using Adobe Photoshop to enhance the bubbles out of the background via adjusting the  
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55 brightness and shadows and making small adjustments to individual colours with the hue and  
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57 saturation layers. Then determination of relative occupations of the surface by the helium  
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1 bubbles was specified after choosing suitable thresholds in ImageJ, version 1.53a [44]. In  
2 order to generate the binary image, the thresholded image was converted to grayscale (single-  
3 channel conversion). This followed by a fixed-level thresholding to determine whether a  
4 pixel is black or white, thus resulting in a binary representation of the image. The contour  
5 estimation algorithm provided a set of connecting pixels, contours, at the edges of the  
6 bubbles. The area of each contour was calculated using the Green's Theorem. Additionally,  
7 the centroid of each bubble was obtained by computing the image moment of its contour, this  
8 allowed to extract the area and centroid of each bubble, giving a full description of the  
9 location and size distribution of the bubbles in the micrograph. The images obtained at each  
10 of the main steps of bubble detection are shown in Fig. 2.

## 23 **Results**

24 Fig. 3 shows the evolution of the W microstructure at 800 °C as a function of damage  
25 level in the range of 0.01 up to 1 dpa, whereas the early stages of damage between 0.004 and  
26 0.016 dpa are shown in Fig. 4. Comparatively, Fig. 5 shows the radiation-induced damage in  
27 W-5Ta in the same damage range as in W in Fig. 3. In both materials, the damaged structure  
28 is characterised by a population of helium bubbles that change in density, size and  
29 morphology with increasing damage level up to the maximum value of 1 dpa in this work.  
30 Fig. 6 displays the evolution of the average bubble size and the number density as it relates to  
31 damage level in both materials. The radiation-induced dislocation loops were not detected in  
32 any of these materials at 800 °C. A significant number of spherical bubbles of >1 nm in  
33 diameter are already present in W at the relatively low damage level of 0.004 dpa, which  
34 corresponds to a He<sup>+</sup> fluence of  $3.3 \times 10^{14}$  He<sup>+</sup>/cm<sup>2</sup>. Both the average bubble size and density  
35 increase continuously up to a damage level of 0.2 dpa, where the average size reaches ~3nm  
36 and the density amounts to  $\sim 26 \times 10^{22}$ /m<sup>3</sup>. At higher damage levels, both the average size and  
37 density continue increasing but at a lower rate, and do not reach saturation at the end of the  
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1 experiment. At the maximum damage level of 1 dpa, the average size of the bubble  
2 population is 3.9(3) nm and the density is  $\sim 32 \times 10^{22}/\text{m}^3$ . Additionally, at damage levels  $\geq 0.3$   
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4 dpa, the He bubbles gradually change their morphology from spherical to faceted (see insets  
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6 in Fig. 3 and Fig. 7a). In contrast, there are no visible bubbles in W-5Ta below 0.02 dpa, and  
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8 the bubble density remains low up to 0.1 dpa. Between 0.1 and 0.2 dpa, there is a sharp  
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10 increase in bubble density. At  $>0.2$  dpa the density continues to rise at a lower rate than  
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12 before but still higher than in W and reaches a value of  $\sim 31 \times 10^{22}/\text{m}^3$ , which is close to the  
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14 bubble density in W at the same damage level. Additionally, the average bubble size in  
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16 W-5Ta undergoes a smaller increase with damage level and seems to saturate at 2.2(3) nm at  
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18 1 dpa. That size value is  $\sim 1.7$  times smaller than the value measured in W also at 1 dpa. At  
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20 such damage level, most of the He bubbles in W-5Ta still remain largely spherical (see Fig.  
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22 7b). Figure 8 plots the size frequency distribution of helium bubbles showing an increase in  
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24 size as a function of a damage level up to 1 dpa. The histograms reveal a shift in bubble sizes  
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26 limited to a maximum of  $<3.5$  nm and a peak frequency at  $<2.5$  nm in W-5Ta and a  
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28 maximum of  $<5$  nm and a peak frequency at  $<4$  nm in W at 1 dpa.  
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## 37 Discussion

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39 The helium irradiated microstructure of W at 800 °C is characterised by the presence  
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41 of a relatively large number of bubbles, whose average size and bubble density change with  
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43 an increase of a damage level, and addition of Ta content. There was no evidence of  
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45 dislocation loops at 800 °C. This is supported with data on tungsten-rhenium alloys irradiated  
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47 with neutrons and tungsten-tantalum alloys irradiated with protons at temperatures up to  
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49 800 °C [29, 45]. The interstitial dislocation loops would be unstable at this elevated  
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51 temperature and emit self-interstitial atoms, or glide to the free sample surface. Whereas,  
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53 tungsten sample irradiated with neutrons at  $T < 750$  °C and a damage level of  $<1$  dpa are  
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55 characterised by the presence of dislocation loops, whereas at higher irradiation temperatures  
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1 the irradiated microstructure is dominated by the presence of nm-sized voids and the absence  
2 of loops [46]. Generally, both W and W-Ta exhibit microstructures with  $\leq 20\%$  of the area  
3 occupied by the radiation-induced bubbles under 40 keV He<sup>+</sup> exposure up to 1 dpa. The  
4 helium fluence was reported to exhibit a very limited impact on bubble size in W at  
5 T < 1200°C. In tungsten irradiated at 800°C by 10 keV helium ions, the average bubble  
6 density was found to be around  $(3.0-4.0) \times 10^{16} \text{m}^{-2}$ , and the mean bubble size was reported to  
7 be between 2.0 nm and 3.5 nm which is similar to results obtained in this work ( $\sim 2 \times 10^{16} \text{m}^{-2}$ )  
8 [47].  
9

10 Helium atom diffusion is the principal factor affecting the helium bubble nucleation  
11 and growth processes. Helium has an extremely low solubility and low migration energy in  
12 metals [48]. The irradiation temperature 800°C in this work corresponds to the annealing  
13 stage IV reported to occur in W between 650-1000°C, when the radiation-induced vacancies  
14 become mobile [49]. In tungsten, substitutional helium atoms tend to form various strong  
15 complexes with vacancies, which are considered as the nuclei of the helium bubbles, which  
16 preferentially grow by further helium and helium-vacancy absorption [50]. DFT calculations  
17 showed that helium will migrate to vacancies rather than remain in interstitial or  
18 substitutional sites [51]. Helium atoms are strongly bound with the vacancies because they  
19 compensate its negative volume dilatation [52]. The energy barrier was estimated and  
20 reported to be quite high (2.8 eV) for a possibility of a direct jump of the He atom from one  
21 vacancy to another in W, supposing a great trapping effect between He and the vacancy in W  
22 [53]. At temperatures > 600°C mobility of the He vacancy complexes is reported to increase,  
23 which leads to the development of larger (greater than several nanometres) bubbles [54].  
24 Ultimately these implanted He atoms that are trapped at vacancies may cause helium  
25 embrittlement of the material.  
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Diffusion in an alloy is more complex than in a single element component where  
vacancy-helium atom clustering might be retarded due to the existence of solute dopants. The  
first-principle calculations revealed a repulsive Ta solute-vacancy interaction in W with a  
binding energy between Ta and a monovacancy from  $-0.1$  to  $-0.3\text{eV}$  for the first-to-third  
nearest-neighbour positions. However, a weak attractive interaction was reported for Ta atom  
on the neighbouring position perpendicular to the  $\langle 111 \rangle$  crowdion, resulting in the impeded  
diffusion of the SIA [55]. Therefore, the solute Ta partly increases the recombination of  
vacancy with interstitial, and decreases the concentration of point defects, thereby, leading to  
a reduced vacancy concentration within the matrix. Moreover, the binding energy between a  
Ta interstitial atom and a He atom in W, when He was initially situated in the third to first  
nearest-neighbour sites relatively to a Ta atom, was reported to be  $0.1\text{-}0.3\text{ eV}$ , and the  
binding energy between the second introduced He atom and existing He-Ta complex was  
declared to be  $1.1\text{ eV}$ . This indicates a strong favourable binding effect and consequent  
trapping of He by substitutional Ta or already formed He-Ta complexes preventing He atom  
migration in the W lattice [56]. The consequence of the described above events is a lower  
probability of interaction between vacancies and He atoms in the vicinity of tantalum atoms.

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In addition, the formation energy of a mono-vacancy for the first-to-fourth nearest-  
neighbour positions in W-5Ta alloy was reported to be higher compared to the pure tungsten,  
suggesting that mono-vacancies are not that easily formed around the Ta atom in W [57].  
delaying vacancy diffusion via a solute drag effect [58], which may retard the vacancy  
mobility and further binding of a vacancy with He in W. The formation energy of a He  
interstitial in the W-5Ta alloy, with the most stable configuration for He in the substitution  
site, was reported to be lower ( $4.56\text{ eV}$ ) and therefore more favourable compared to the pure  
tungsten system ( $4.92\text{ eV}$ ) [59]. The formation energies of the He atom in the tetrahedral  
interstitial site and octahedral interstitial site were also calculated in pure W and were

1 reported to be 6.18 and 6.43 eV which is also higher than in W-Ta alloy (5.50 and 5.84 eV  
2 respectively). The helium-vacancy clustering and the ability of bubble formation and their  
3 further growth is therefore retarded in the W-Ta system below 0.02 dpa. Whereas in contrast,  
4 already at a damage level of 0.004 dpa, small bubbles of >1 nm have been detected in W  
5 yielding the density of  $4.2 \times 10^{20} \text{ m}^{-3}$ . Hence, the presence of Ta interferes with  
6 helium/vacancy clustering and subsequently delays bubble nucleation causing deceleration of  
7 the bubble transition into faceted shape at higher damage levels.  
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17 A similar trend in the radiation-induced defect retention was observed in W-5Ta  
18 irradiated in-situ by 40 keV protons at 800°C [29, 60]. In comparison with the growing  
19 number of voids in W, the voids in W-5Ta were not detected up to a damage level of  
20 0.3 dpa, however, the proton irradiation-induced voids became visible when the same sample  
21 was post-irradiation annealed at 1000 °C, with a relatively low void density of  $4.9 \pm 1.4 \times$   
22  $10^{21} \text{ m}^{-3}$  and an average size of  $4.9 \pm 1.4 \text{ nm}$ , occurring as a result of the agglomeration of  
23 voids by a thermally activated process [29]. And the exposure of W-5Ta to a lower energy of  
24 100 eV He<sup>+</sup> ions showed similar behaviour with surface fuzz, a result of defect mobility  
25 being suppressed in comparison to unalloyed W due to the bigger lattice spacing forced by  
26 doping of W with Ta [33, 61]. The higher fluence threshold for the formation of surface  
27 nano-structuring implies that the W-Ta alloy's surface thermal conductivity will degrade at a  
28 slower rate compared to the pure W system during the lifespan of a reactor [62].  
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46 The surface effect needs to be taken into account during the in-situ irradiations,  
47 especially when the TEM foil thickness is <100 nm [63]. The free surface effect was  
48 previously observed on the dislocation structure due to the thin foil in the in-situ experiment  
49 [31]. The ex-situ TEM analysis revealed the coexistence of dislocation tangles and loops in  
50 both materials, as opposed to the lack of dislocation tangles in the in-situ specimens.  
51 Furthermore there was no change in the nature (i.e. interstitial to vacancy type) of the  
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1 dislocation loops and their Burgers vector, and a significant number of vacancy clusters was  
2 also not present, but we did observe the appearance of dislocation tangles in the ex-situ  
3 samples. The ex-situ irradiation of bulk tungsten with He ions of 400 keV showed the  
4 formation of larger bubble diameters ( $>10$  nm) but lower areal bubble density  
5 ( $\sim 10^{16}$  bubbles/m<sup>2</sup>) compared to the in-situ experiments. This was due to higher amounts of  
6 He retained in the irradiated bulk materials while in the in-situ experiments on thin foils some  
7 may escape due to the proximity of surfaces [64]. In addition, at higher irradiation  
8 temperature the diffusivity ratio of point defect is greater [63]. Hence, considering the  
9 influence of the surface effect, the population of the helium bubbles measured in the thin  
10 TEM foils is lower than the actual value in bulk would be.

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At a damage level  $\geq 0.3$  dpa, the bubbles present in W develop energetically favourable facets with the facet planes of  $\{110\}$ , as interpreted by the anisotropy of the surface energy and by the preferential adsorption of diffusing helium ions on specific plane orientations with respect to the bubble surface. The descending order of surface energies in bcc is for low-index planes  $\{111\}$ ,  $\{100\}$ , and  $\{110\}$  with the lowest surface energy for the planes  $\{110\}$  due to the greater planar density [65]. To minimize the total free energy, the facets with the lowest surface energy occupy most of the bubble surface along  $\{110\}$ . The fraction of the faceted bubbles in W rises and amounts to  $>80\%$  at a maximum of 1 dpa. However, the spherical-to-faceted transition of bubble shape is clearly suppressed in W-5Ta alloy.

## 66 67 68 69 70 71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 87 88 89 90 91 92 93 94 95 96 97 98 99 100

In-situ analysis of helium bubble formation has been performed in W and W-5Ta alloy, caused by 40 keV He<sup>+</sup> irradiation at 800°C up to 1 dpa with a maximum He-to-dpa ratio of 5.5 at%/dpa. The extensive use of advanced in-situ TEM characterizations showed the effect of helium fluence and alloying component on the structural morphology of the

1 helium bubbles. The spherical bubbles of >1 nm were observed in W at the relatively low  
2 damage level of 0.004 dpa, which corresponds to a He<sup>+</sup> fluence of 3.3×10<sup>14</sup> He<sup>+</sup>/cm<sup>2</sup>. The  
3 presence of only 5wt.%Ta in solid solution impedes the diffusion of helium ions at 800 °C  
4 and interferes with the binding of He ions with vacancies into complexes, which serve as the  
5 core of a helium bubble, and therefore hinders the nucleation of the bubbles <0.02 dpa.  
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7 Moreover, at 1 dpa, >80% of the bubbles in W steadily develops facets, whereas in contrast  
8 in W-5Ta bubbles still present a spherical shape.  
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### 10 11 12 13 14 15 16 17 **Figure captions**

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19 **Fig. 1.** Simulated damage and He<sup>+</sup> concentration profiles in W at a damage level of 1 dpa to a  
20 He-to-dpa ratio of 5.5 at%/dpa, using the SRIM software with the quick Kinchin-Pease  
21 approach and the total current deposited on the sample in the experiment of 0.09 nA. The  
22 damage level was estimated at each step in the microstructure evolution as the average value  
23 in the foil thickness of ~100 nm. According to SRIM calculations, 32% of the 40keV He ions  
24 was implanted and 56% was transmitted through the foil.  
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34 **Fig. 2.** A series of images of helium bubbles observed in W at 800 °C and a damage level of  
35 1 dpa through the main steps of the bubble detection: (a) bright-field image in under-focus  
36 condition, (b) thresholded image with enhanced bubbles, (c) grayscale binary image, (d)  
37 estimated contoured areas of detected bubbles  
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44 **Fig. 3.** In-situ observation of the bubble evolution W at 800 °C at selected damage levels up  
45 to 1 dpa induced by a 40 keV He<sup>+</sup> ion beam: (a) 0.01 dpa, (b) 0.02dpa, (c) 0.05 dpa, (d) 0.075  
46 dpa, (e) 0.1 dpa, (f) 0.2dpa, (g) 0.3 dpa, (h) 0.4 dpa, (i) 0.75 dpa, (j) 1 dpa. The insets show  
47 the bubble transition from spherical to faceted at  $\geq 0.3$  dpa. The TEM foil thickness was  
48 derived by way of the graphical method from the spacing of the fringes of the CBED patterns  
49 and was measured to be  $\sim 100$  nm  $\pm$  10%  
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1 **Fig. 4.** In-situ observation of bubble formation in W at 800 °C caused by a 40 keV He<sup>+</sup> ion  
2 beam at the relatively low damage levels of: (a) 0.004 dpa, (b) 0.008 dpa, (c) 0.016 dpa. The  
3 TEM foil thickness was derived by way of the graphical method from the spacing of the  
4 fringes of the CBED patterns and was measured to be ~100 nm ± 10%  
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9 **Fig. 5.** In-situ observation of the bubble evolution W-5Ta at 800 °C at selected damage levels  
10 up to 1dpa induced by a 40 keV He<sup>+</sup> ion beam: (a) 0.01 dpa, (b) 0.02dpa, (c) 0.04 dpa, (d)  
11 0.05 dpa, (e) 0.1 dpa, (f) 0.2 dpa, (g) 0.3 dpa, (h) 0.4 dpa, (i) 0.75 dpa, (j) 1 dpa  
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16 **Fig. 6.** Average bubble size and bubble density in W and W-5Ta alloy at 800 °C as a function  
17 of damage level  
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21 **Fig. 7.** Bright field TEM image of spherical bubbles in W-5Ta and faceted bubbles in W, at  
22 800 °C and the damage level of 1 dpa induced by a 40 keV He<sup>+</sup> ion beam. The insets show  
23 the electron diffraction patterns taken along the zone axis <001> and <111>  
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28 **Fig. 8** Frequency distribution histograms of helium bubble size showing an increase in size as  
29 a function of a damage level in W and W-5Ta irradiated with 40 keV He ions to a He-  
30 appm/dpa ratio of 55,000 and dose of 1 dpa  
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