Deuterium release from deuterium plasma-exposed neutron-irradiated and non-neutron-irradiated tungsten samples during annealing

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Abstract
We examine the effect of neutron irradiation on the release of deuterium from tungsten at 573 K to understand the efficiency of tritium removal by baking out at moderate temperatures. Tungsten samples, undamaged and neutron-irradiated to a damage level of approximately 0.016 displacements per atom, are exposed to low-energy (108 eV), high-flux (3.0 × 10²¹ to 9.4 × 10²¹ m⁻² s⁻¹) deuterium plasma at temperatures ranging from 573 to 773 K to an ion fluence of 1.1 × 10²⁵ m⁻². At each exposure temperature, two undamaged and two neutron-irradiated tungsten samples are exposed to plasma. The deuterium content in the tungsten samples is measured by thermal desorption spectrometry soon after the plasma exposure and after post-plasma annealing at 573 K for 30 h. It is found that: (i) the deuterium retention in the neutron-irradiated tungsten samples is significantly higher than that in the undamaged tungsten samples; (ii) annealing at 573 K of undamaged tungsten samples pre-exposed to deuterium plasma at 573–773 K leads to an almost complete (60%–99%) release of deuterium from the samples; (iii) annealing at 573 K of neutron-irradiated tungsten samples pre-exposed to deuterium plasma at 573–773 K leads to a significant (8%–20%) release of deuterium from the samples.

Keywords: tungsten, neutron-induced defects, deuterium retention, deuterium release under annealing

(Some figures may appear in color only in the online journal)
quickly and efficiently. Methods for removing hydrogen isotopes from tokamaks have already been the subject of discussion [1–5]. Those used to measure the effectiveness of tritium removal from the vacuum vessel components of the Joint European Torus and the Tokamak Fusion Test Reactor were (i) a deuterium gas soak; (ii) glow discharge cleaning with working gasses of helium, deuterium, and a mixture of helium–oxygen gases; (iii) electron cyclotron resonance discharges with deuterium as a working gas; (iv) a vessel vent with nitrogen and air; and (v) bake-out of in-vessel surfaces at temperatures up to approximately 410 K [3–5]. Tritium removal by laser desorption and annealing at 623 K were proposed for use in ITER [6, 7].

As a potential material for PFCs in future fusion reactors, tungsten will be subjected to intensive fluxes of energetic deuterium and tritium and 14 MeV neutrons. Neutron irradiation generates displacements in the tungsten bulk and thus creates defects at which hydrogen isotopes are trapped [8–18]. One of the possible tritium removal methods is to heat the PFCs after the deuterium–tritium operation using the decay heat generated by the radioisotopes that appear in the PFCs due to neutron irradiation. It was assumed that during servicing, the temperature of the elements of the vacuum chamber would be reduced to 573 K, and these elements would remain at this temperature for a month.

This work aimed to estimate the fraction of released deuterium from neutron-irradiated and non-neutron-irradiated (undamaged) tungsten samples previously irradiated with deuterium plasma at elevated temperatures, followed by annealing at 573 K for 30 h.

2. Experiments

Two types of tungsten samples (diameter 6 mm, thickness 0.5 mm, and polished to a mirror shine) were used in this work:

(i) Polycrystalline tungsten samples (A.L.M.T. Co, Japan) recrystallized after polishing by annealing in dry hydrogen at 2073 K. The purity of the recrystallized samples was 99.99 mass%, and the grain size was in a scale of mm. In the fusion reactor BR2 at the Belgian Nuclear Research Center [19], the recrystallized tungsten samples were irradiated with fast neutrons ($E > 1$ MeV) at a temperature of approximately 563 K to a fluence of $3.0 \times 10^{23}$ n m$^{-2}$, which corresponded to a damage level of approximately 0.016 displacements per atom (dpa). Before the plasma exposure, the surface of each neutron-irradiated tungsten sample was electrochemically polished with a 1 M NaOH solution at 5 V for 5 min to remove impurities and oxide layers formed during neutron irradiation at an elevated temperature for 22 d and post-irradiation handling. If, after neutron irradiation, contaminants were observed on the tungsten sample surfaces, then, after electropolishing, the surfaces acquired a characteristic metallic luster. Thin surface layers ($10–20 \mu$m for each surface) were removed by the electrochemical polishing.

(ii) Polycrystalline hot-rolled tungsten samples (Goodfellow Cambridge Ltd, UK) with a purity of 99.95 mass% were annealed at 1273 K after polishing in vacuum for 1 h to relieve the internal stress induced during the fabrication process and polishing. The microstructure of the hot-rolled tungsten samples consisted of two-dimensional flat grains, 2–5 $\mu$m in size and around 1 $\mu$m in thickness, positioned in parallel to the surface [20]. These samples are referred to as ‘undamaged’ tungsten samples.

The choice of materials for these experiments is explained as follows. Polycrystalline hot-rolled tungsten with a microstructure of two-dimensional flat grains located parallel to the surface was selected as undamaged samples. Recrystallized tungsten samples with grain sizes in a scale of mm were chosen for irradiation with neutrons.

It was previously shown that the accumulation of deuterium in tungsten samples irradiated with heavy metal ions does not depend on the structure of the tungsten material because deuterium retention by ion-induced defects dominates deuterium retention by defects associated with the structure of tungsten [21, 22]. Therefore, in the case of tungsten irradiated with neutrons to the damage levels reachable in future fusion reactors (approximately 0.7 dpa in ITER for a whole campaign [23] and much higher in the DEMOnstration Power Station (DEMO), 20–40 dpa per operational year [24]), the effect of defects in the initial tungsten microstructure on the accumulation of hydrogen isotopes will be insignificant. Nevertheless, recrystallized tungsten was chosen for neutron irradiation to minimize the possible effect of the original intrinsic defects of the tungsten matrix on the accumulation of deuterium during plasma irradiation and the deuterium release during post-plasma annealing for the purity of the experiment.

However, recrystallization of tungsten results in embrittlement [25, 26], and hence worked tungsten with fine grains is preferable for fusion applications. This characteristic has led to the choice of material for our undamaged samples.

Each tungsten sample was mounted separately on the sample holder and exposed to deuterium plasma in a compact divertor plasma simulator (CDPS) [27] located in the radiation-controlled area of the International Research Center for Nuclear Materials Science, Institute for Materials Research, Tohoku University.

Before plasma exposure, low-density plasma was generated, and the sample holder was biased positively at $+5$ V to heat the tungsten sample to the required temperature by electron bombardment. As soon as the sample reached the required temperature, the deuterium plasma was turned on, and a negative bias of $–110$ V was applied to the sample holder. Of note, the temperature of the sample was determined by the ion flux, and therefore, the plasma exposure at a higher sample temperature occurred at a higher ion flux. The sample temperature was monitored with a tungsten–rhenium (W-5%Re/W-26%Re) sheath-type thermocouple. The thermocouple head was pressed against the back of the sample to ensure a good thermal connection between the thermocouple and sample. During the plasma exposure, the temperature of the sample was kept constant by adjusting the rate of air flow passing inside the holder using a mass flow controller and a feedback controller for the temperature of the sample measured by the thermocouple [27].
The undamaged and neutron-irradiated tungsten samples were exposed to deuterium plasma at the exposure temperature, $T_{\text{exp}}$, of 573, 673 and 773 K to an ion fluence of $1.1 \times 10^{23} \text{ m}^{-2}$. The temperature of the sample during deuterium plasma exposure varied within 1.6% of the required exposure temperature expressed in Kelvin. To measure the electron temperature and density and the plasma space potential, a fast reciprocating Langmuir probe was introduced into the plasma 9 mm away from the sample holder. The plasma potential depended weakly on the ion flux and was approximately $-2 \text{ V}$. Thus, the energy of the deuterium ions was estimated to be 108 eV.

The ion composition in the deuterium plasma generated in CDPS had not previously been measured. However, Hollmann et al. \cite{28} published their results after measuring the ion composition of hydrogen plasma generated in a linear plasma device NAGDIS-II. This measurement was carried out using an omegatron mass spectrometer. The CDPS and NAGDIS-II are similar devices with the same ion sources, generating plasmas of similar parameters at equal working gas pressures \cite{27, 28}. The omegatron mass spectra peak heights indicate that hydrogen (H) plasma generated in the NAGDIS-II typically consists of H$^+$ ions, with smaller (<10%) concentrations of H$_2^+$ and/or H$_3^+$ \cite{28}. The same ion composition was assumed for deuterium plasma generated in the CDPS.

For each deuterium plasma exposure, the ion flux was determined based on the electron temperature and density (table 1, figure 1). The ion flux was determined by averaging the data obtained by the Langmuir probe over the three central points above the surface of the sample (figure 1). The diameter of the plasma beam in the region of the sample holder was approximately 50 mm, and the diameter of the sample was 6 mm.

When irradiated with deuterium plasma at a certain fixed temperature, the ion flux depends on the thermal contact between the sample and holder, and it can be different in each case. Thermal contact is determined by the roughness of the back of the sample and the force of pressing the sample to the holder. The elasticity of the springs controlling the hooks holding the sample decreases with time, especially after irradiation at a temperature of 773 K. The roughness of the samples arises due to the electrochemical removal of surface contamination resulting from neutron irradiation.

The CDPS is equipped with a sample carrier system with a tray made of molybdenum that allows plasma-irradiated samples to be transferred from the sample holder to a tube made of quartz glass without exposing the sample to air \cite{27}. The quartz tube was heated by an external infrared heater for thermal desorption spectrometry (TDS). Linear heating of the sample was carried out using a programmable temperature controller (ULVAC-Riko Inc, Japan, TPC-5000). Thermocouples attached to the bottom of the molybdenum tray were used for temperature monitoring \cite{27}.

Two identical tungsten samples, undamaged and neutron-irradiated, were individually exposed to deuterium plasma at each sample temperature. The deuterium retention in the first plasma-exposed tungsten sample was immediately measured after the plasma exposure using TDS when heated to 1300 K with a heating rate of 0.5 K s$^{-1}$.

The second plasma-exposed tungsten sample was initially heated in the quartz tube by an infrared heater up to 573 K and was retained at this temperature for 30 h at a background pressure of $(3–4) \times 10^{-6} \text{ Pa}$. Then, the amount of deuterium remaining in the second tungsten sample was determined by TDS.

A quadrupole mass spectrometer (QMS) (MKS Instruments Inc, USA, Microvision-2 100D) was used to measure the mass 2 (H$_2$), mass 3 (HD), and mass 4 (D$_2$) signals to determine the deuterium desorption.

To calculate the absolute contributions of the recorded masses to the deuterium release rate, the partial intensities of the QMS signals of HD and D$_2$ were normalized as described by Franzen et al. \cite{29}. The normalization included the ionization probability, cracking pattern of each recorded species in the ionizer, and transmission factor of each mass through the quadrupole to the detector. By normalizing the product of these factors to 1 for D$_2$, a factor of 0.7 was obtained for HD. Taking into account the number of deuterium atoms per molecule and assuming that all deuterium was desorbed as D$_2$ and HD, we arrived at the deuterium release rate

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**Figure 1.** Distribution of the ion flux along the diameter of the sample holder in the case of low-energy (108 eV) deuterium plasma exposure of neutron-irradiated tungsten samples at sample temperatures, $T_{\text{exp}}$, of 573, 673 and 773 K in a CDPS. The center of the samples corresponds to 0 mm. The values of ion fluxes averaged over three central points at which the electron temperature and density were measured by the Langmuir probe are presented in table 1. The diameter of the plasma beam was approximately 50 mm.
defect production is not fulfilled. It could be explained by the mechanisms of fracture and/or plastic deformation due to deuterium supersaturation [30, 31].

The accumulation of deuterium in undamaged polycrystalline hot-rolled tungsten exposed to deuterium plasma with ion energies well below the displacement threshold can be explained by the mechanisms of fracture and/or plastic deformation due to deuterium supersaturation [32]. During exposure to low-energy, high-flux deuterium plasma, the transient deuterium concentration in the implantation zone greatly exceeds the solubility limit and stress the matrix lattice. When the compressive stress induced by the deuterium supersaturation exceeds the yield stress of the material, fracture deformation and/or plastic deformation occur [32], and intergranular/intragranular cracks (cavities) [33–35] and vacancy-type defects [36] are generated.

The low deuterium retention in the undamaged polycrystalline tungsten exposed to deuterium plasma at a temperature of 773 K (figure 2(c)) indicates that the requirement for extended defect production is not fulfilled. It could be explained by the strongly increased deuterium solubility, diffusivity and rate of surface recombination avoiding the transient deuterium concentration exceeding the solubility.

Table 1. Conditions of exposure of undamaged and neutron-irradiated tungsten samples with low-energy (108 eV) deuterium plasma to an ion fluence of 1.1 × 10^{25} m^{-2} (T_{exp} is a temperature of tungsten sample maintained during deuterium plasma exposure, \Gamma_i is the ion flux, and \Gamma is the exposure time).

| Samples exposed to D plasma | Samples exposed to D plasma and then annealed |
|----------------------------|---------------------------------------------|
| T_{exp}, K                 | \Gamma_i, m^{-2} s^{-1}/\Gamma, s           |
| Undamaged tungsten samples |                                             |
| 573                        | 3.0 × 10^{23}/3700                          |
| 673                        | 5.7 × 10^{23}/1900                          |
| 773                        | 7.9 × 10^{23}/1400                          |
| Neutron-irradiated samples |                                             |
| 573                        | 3.4 × 10^{23}/2860                          |
| 673                        | 5.8 × 10^{23}/2860                          |
| 773                        | 9.4 × 10^{23}/1150                          |

R_D = k (0.7 \times I_{HD} + 2 \times I_{D2})/S_{sample}, where I_{HD} and I_{D2} are the intensities of the HD and D_2 signals, respectively, S_{sample} is the surface area of the sample exposed to deuterium plasma, and k is the calibration factor obtained after each TDS analysis by measuring the I_{D2} when deuterium flowed from a calibrated D_2 leak. Two calibrated D_2 leaks with an accuracy of 7% were employed to calibrate the QMS after each TDS analysis.

3. Results and discussion

The TDS deuterium spectra for the undamaged polycrystalline hot-rolled tungsten samples exposed to low-energy (108 eV) deuterium plasma at T_{exp} of 573, 673 and 773 K to an ion fluence of 1.1 × 10^{25} m^{-2}, with and without subsequent post-plasma-exposure annealing at 573 K for 30 h, are shown in figure 2. According to the TDS measurements immediately performed after the plasma exposure, an increase in the exposure temperature from 573 to 773 K led to a decrease in the amount of released deuterium from 3.9 × 10^{20} to 4.7 × 10^{18} D m^{-2} (table 2). This decrease in the amount of deuterium correlated well with previously published data on the retention of deuterium in neutron-irradiated tungsten samples exposed to low-energy (76 eV), high-flux (about 5 × 10^{23} m^{-2} s^{-1}) deuterium plasma [30, 31].

R_{exp} = \Gamma_i \times S_{sample} \times (1 - e^{-S_{sample}/T_{exp}}), where \Gamma_i is the ion flux, and S_{sample} is the surface area of the sample exposed to deuterium plasma.

The TDS deuterium spectra for the neutron-irradiated tungsten samples at T_{exp} 573 K and 673 K were successfully employed to calibrate the QMS after each TDS analysis. The TDS deuterium spectra for the neutron-irradiated tungsten samples at T_{exp} 573 K and 673 K were successfully employed to calibrate the QMS after each TDS analysis. During plasma exposure, the deuterium retention in the neutron-irradiated tungsten samples at 573 K was higher than that in the undamaged samples by a factor of approximately 4 after plasma exposure at 573 K and a factor of approximately 100 after plasma exposure at 773 K (table 2).

Post-plasma annealing of the neutron-irradiated tungsten samples at 573 K for 30 h led to the release of 8%–20% of the deuterium previously accumulated (figure 3, table 2). Obviously, neutron irradiation of tungsten creates vacancy-type defects for which the binding energy with hydrogen isotopes is much higher than the binding energy for defects created only by plasma exposure.

The TDS spectra for the neutron-irradiated tungsten samples exposed to deuterium plasma at T_{exp} 573 K demonstrated an excess of the deuterium release rate after post-plasma annealing over the deuterium release rate immediately after exposure to deuterium plasma. This effect was observed in the high-temperature range of the TDS spectra (≥835 K for T_{exp} = 573 K and ≥1050 K for T_{exp} = 673 K).
Figure 2. Thermal desorption spectra of deuterium for undamaged tungsten samples exposed to low-energy (108 eV) deuterium plasma at sample temperatures, $T_{\text{exp}}$, of 573 K (a), 673 K (b) and 773 K (c) to ion fluence of $1.1 \times 10^{25}$ m$^{-2}$ with and without subsequent post-plasma-exposure annealing at 573 K for 30 h. The deuterium release rate scales in panels (a), (b) and (c) are different from one another.
Figure 3. Thermal desorption spectra of deuterium for neutron-irradiated tungsten samples (\(\sim 0.016\) dpa) exposed to low-energy (108 eV) deuterium plasma at sample temperatures, \(T_{\text{exp}}\), of 573 K (a), 673 K (b) and 773 K (c) to ion fluence of \(1.1 \times 10^{25}\) m\(^{-2}\) with and without subsequent post-plasma-exposure annealing at 573 K for 30 h. The deuterium release rate scales in panels (a), (b) and (c) are different from one another. \(Q_d\) is assumed to be the amount of deuterium detrapped from defects during annealing at 573 K for 30 h and trapped again by defects localized in deeper layers. The shaded area shows the difference between the deuterium release rates for the TDS spectra obtained after post-plasma annealing and after exposure only to deuterium plasma in the cases of integration from 835 to 1300 K for \(T_{\text{exp}} = 573\) K, and from 1050 to 1300 K for \(T_{\text{exp}} = 673\) K, as examples.
detrap/trap deuterium detrapped from the neutron-induced defects during the subsequent annealing and again trapped by defects localized in deeper layers of plasma-exposed tungsten sample during subsequent annealing (Tungsten sample after deuterium plasma exposure and subsequent annealing; tungsten sample after post-irradiation annealing at 573 K will be more efficient. Therefore, after annealing, deuterium could not fill all the traps. Indeed, Toyama et al. [37], who measured positron lifetime in tungsten after neutron irradiation at ~573 K for 48 d and after post-irradiation annealing at 573 K for 100 h, observed no noticeable change in positron lifetime after the post-irradiation annealing.

Second, a part of the deuterium detrapped during the post-plasma annealing at 573 K moved inward and was re-trapped in a deeper region instead of being diffused to the surface and released. Under the deuterium plasma exposure conditions used in this work, the deuterium could not fill all the neutron-induced traps created throughout the bulk of the tungsten samples. As shown by Yajima et al. [18], the exposure of a neutron-irradiated tungsten sample with 108 eV deuterium plasma at a temperature of 563 K to an ion fluence of $3 \times 10^{25}$ m$^{-2}$ in the CDPS (i.e. in the same plasma generator that was used in this work) leads to the trapping of deuterium at depths of up to 17 $\mu$m.

We can assume that if a neutron-irradiated tungsten sample is completely saturated with deuterium, then its removal during annealing at 573 K will be more efficient. Therefore, after post-plasma annealing at 573 K, the amount of deuterium, detrapped from defects during this annealing and trapped again by defects localized in deeper layers should be estimated.

For neutron-irradiated tungsten samples exposed to deuterium plasma at $T_{\text{exp}} = 573$ K, the amount $Q_D^{\text{detrap/trap}}$ was estimated as the difference in the integrals of the deuterium release rate over time at temperatures ranging from 835–920 K up to 1300 K for TDS spectra obtained after post-plasma annealing and after exposure only to deuterium plasma. For $T_{\text{exp}} = 673$ K, the amount $Q_D^{\text{detrap/trap}}$ was estimated using a temperature range from 1050 to 1300 K.

| $T_{\text{exp}}$, K | plasma $Q_D$, D/m$^2$ | plasma-ann $Q_D$, D/m$^2$ | released $F_D$ | detrapped $Q_D$, D/m$^2$ | released $F_D +$ detrapped $F_D$ |
|-------------------|-------------------|-------------------|---------------|------------------|-----------------------------|
| 573               | $3.9 \times 10^{20}$ | $5.7 \times 10^{18}$ | 0.99          | 0                | 0                           |
| 673               | $3.0 \times 10^{19}$ | $5.8 \times 10^{18}$ | 0.81          | 0                | 0.81                        |
| 773               | $4.7 \times 10^{18}$ | $1.9 \times 10^{18}$ | 0.60          | 0                | 0.60                        |
| 573               | $1.54 \times 10^{20}$ | $1.41 \times 10^{20}$ | 0.08          | (3.0–3.7) $\times 10^{20}$ | 0.19–0.24 |
| 673               | $1.42 \times 10^{20}$ | $1.14 \times 10^{20}$ | 0.20          | $4.2 \times 10^{19}$ | 0.03 |
| 773               | $4.8 \times 10^{20}$ | $4.1 \times 10^{20}$ | 0.15          | 0                | 0.15 |

(2020) 096025 V. K. Alimov

Table 2. Parameters characterizing deuterium retention in undamaged and neutron-irradiated tungsten samples after exposure with low-energy (108 eV) deuterium plasma at 573, 673 and 773 K to an ion fluence of $1.1 \times 10^{25}$ m$^{-2}$, and deuterium release during subsequent annealing at 573 K for 30 h. $T_{\text{exp}}$ is the temperature of the tungsten sample maintained during deuterium plasma exposure; plasma $Q_D$ is the deuterium retention in the tungsten sample after deuterium plasma exposure; plasma-ann $Q_D$ is the deuterium retention in the tungsten sample after deuterium plasma exposure and subsequent annealing; released $F_D$ is the fraction of deuterium released from the plasma-exposed tungsten sample during subsequent annealing (released $F_D = 1$ — plasma-ann $Q_D$/plasma $Q_D$); detrapped $Q_D$ is the amount of deuterium detrapped from the neutron-induced defects during the subsequent annealing and again trapped by defects localized in deeper layers; released $F_D$ is the fraction of deuterium detrapped from the neutron-induced defects during the subsequent annealing and trapped again by defects localized in deeper layers (released $F_D = \frac{\text{detrap/trap} Q_D}{\text{detrap/trap} Q_D + \text{released} F_D}$); and released $F_D +$ detrapped $F_D$ is the estimated fraction of released deuterium when all neutron-induced traps were filled with deuterium. For the exposure temperature, $T_{\text{exp}}$, of 573 K, the amount $Q_D^{\text{detrap/trap}}$ was estimated as the difference in the integrals of the deuterium release rate over time at temperatures ranging from 835–920 K to 1300 K for TDS spectra obtained after post-plasma annealing and after exposure only to deuterium plasma. For $T_{\text{exp}} = 673$ K, the amount $Q_D^{\text{detrap/trap}}$ was estimated using a temperature range from 1050 to 1300 K.

4. Summary

Tungsten samples, undamaged and neutron-irradiated to a damage level of 0.016 dpa, were exposed to low-energy (108 eV), high-flux ($3.0 \times 10^{21}$ to 9.4 $\times 10^{21}$ m$^{-2}$ s$^{-1}$)
deuterium plasma at sample temperatures in the range of 573 to 773 K to an ion fluence of $1.1 \times 10^{25} \text{m}^{-2}$. At each sample temperature, two identical tungsten samples were exposed to the plasma, and these samples were distributed on the first and second batches. The deuterium retention in the tungsten samples from the first batch was measured by TDS almost immediately after the completion of the plasma exposure. The tungsten samples from the second batch, after the deuterium plasma exposure, were annealed at 573 K for 30 h, and then, the amount of deuterium remaining in the samples after this annealing was analyzed by TDS. The following findings were formulated:

1. Hydrogen isotope retention in neutron-irradiated tungsten samples is significantly higher than that in undamaged tungsten samples.
2. Annealing at 573 K of undamaged tungsten samples pre-exposed to deuterium plasma at 573–773 K leads to a very significant and even almost complete (60%–99%) release of deuterium from the samples.
3. Annealing at 573 K of the neutron-irradiated tungsten samples pre-exposed to deuterium plasma at 573, 673, and 773 K leads to a significant (8%–20%) release of deuterium from the samples. For the neutron-irradiated tungsten samples exposed to deuterium plasma at 573 K, the fraction of deuterium released from traps and newly captured in deeper layers is estimated to be in the range from 0.19 to 0.24, whereas after deuterium plasma exposure at 673 K, this fraction is 0.03. After deuterium plasma exposure at $T_{\text{exp}} = 773$ K, capture of deuterium in deeper layers is not observed.

Thus, the heat treatment of PFCs of a fusion reactor at 573 K for one month is thought to be an effective method for removing a significant fraction of tritium from undamaged and neutron-irradiated tungsten components.

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