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Development of yttrium-containing self-passivating tungsten alloys for future fusion power plants

T. Wegener^{a,*}, F. Klein^a, A. Litnovsky^a, M. Rasinski^a, J. Brinkmann^b, F. Koch^b, Ch. Linsmeier^a

^a Forschungszentrum Jülich GmbH, Institut für Energie- und Klimaforschung – Plasmaphysik, 52425 Jülich, Germany ^b Max-Planck-Institut für Plasmaphysik, Boltzmannstr. 2, 85748 Garching, Germany

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ABSTRACT

Tungsten is a prime material candidate for the first wall of a future fusion reactor. In the case of a lossof-coolant accident (LOCA) wall temperatures of about 1450 K could be reached lasting about 30-60 days due to nuclear decay heat. In the worst case scenario combining LOCA with air ingress, the formation and release of highly volatile and radioactive tungsten trioxide (WO₃) into the environment can occur. Smart self-passivating tungsten alloys preventing the formation of WO₃ can be a way to mitigate this release.

In this contribution we present the studies of a new yttrium-containing W-Cr-Y alloys. The extent up to which yttrium acts as an active element improving the adherence and stability of the protective Cr₂O₃ layer formed during oxidation is assessed. The approach is similar to the one taken for high-temperature steels where active elements stabilize the oxide layers at a substantially reduced thickness by changing the oxygen diffusion and improving the adherence of the protective oxide layer by e.g. avoiding of pores. Further, simulations on mobilized material for the case of a LOCA are developed. In addition, the loss of alloying elements during normal operation of a reactor is estimated. This is done by modelling a thermally activated diffusion, using a diffusion coefficient which is extrapolated from experimental data at higher values.

The oxidation behaviour of magnetron sputtered and therefore alloyed at the atomic level W-Cr-Y alloys is tested in a thermo-gravimetric facility. The isothermal oxidations are performed in a gas mixture, containing 20 kPa oxygen and 80 kPa argon under ambient pressure at temperatures of 1273 K and 1473 K, respectively. Experiments with W-Cr-Y show a parabolic oxidation rate of $k_p = 3 \cdot 10^{-6} \text{ mg}^2 \text{ cm}^{-4} \text{ s}^{-1}$ which is more than five orders of magnitude lower than that of pure tungsten at 1273 K. Investigations using X-ray diffraction analysis and focused ion beam cross-sections in combination with scanning electron microscopy and energy dispersive X-ray spectroscopy are conducted. A protective Cr₂O₃ layer is detected on the surface with a thickness between 100 and 300 nm.

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1. Introduction

Tungsten is one of the most promising candidate materials for the first wall in future fusion reactors having a high melting point, high thermal conductivity, low tritium retention and low erosion yield [1–3]. However, in case of a loss-of-coolant accident (LOCA), the temperature of the first wall armour made of tungsten could rise up to 1450 K within 20-40 days as shown in Fig. 1 [3]. In combination with a break of the vacuum vessel followed by air ingress, significant amounts of tungsten oxidise forming WO₃, which is mobilized by sublimation at temperatures of about 1170 K. At the highest predicted temperatures of around 1450 K an evaporation rate of $300 \text{ m} \text{ h}^{-1}$ is calculated assuming a 1000 m² of first wall surface [4], as shown in Fig. 2. In case of a reactor LOCA normally <50% of the elements typically found in the aerosols (like Ag, Re, W,...) are actually released into the environment [5].

Assuming a similar behaviour for a future fusion reactor, this would result in a mobilization rate of radioactive tungsten of the order of \sim 150 kg h⁻¹, which in turn equals an escaping activity of $\sim 7.5 \cdot 10^{16}$ Bq h⁻¹. This is an unacceptable value.

To prevent this release, self-passivating tungsten alloys with a new composition of about 12 wt.% chromium and 0.6 wt.% yttrium

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^{*} Corresponding author. E-mail address: t.wegener@fz-juelich.de (T. Wegener).

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Fig. 1. Calculated temperature profiles after an accident with a total loss of all coolant [3].



Fig. 2. Mass of tungsten lost by evaporating WO_3 in dependence of the temperature. This calculation assumes a LOCA combined with air ingress in a fusion reactor with a 1000 m² first wall, vapour pressure data is from ref. [6].

Fig. 3. Self-passivating tungsten alloys on the left side (a) during normal operation with a depleted zone of nearly pure tungsten on the plasma-facing surface and on the right side (b) in accidental condition with a protective passivating layer on the surface which prevents further oxidation of tungsten.

(W-12Cr-0.6Y) could be used. In case of a LOCA the chromium of the alloy forms a protective barrier layer of Cr_2O_3 on the surface and prevents further oxidation of the tungsten [7] as depicted on the right side in Fig. 3b. In regular plasma operation the alloyed chromium and yttrium are depleted in the first few nm of the plasma-facing surface by selective sputtering of alloying elements leading to a to nearly pure tungsten plasma-facing surface [8] as shown on the left side in Fig. 3a.

In the novel alloy, the active/reactive element yttrium is supposed to stabilize the passivating oxide layer similar as reported for the W-Cr-Y₂O₃ system [9]. In literature three main possible ways on how active elements work are discussed. Firstly, they are thought to form pegs, which act as a connection between the oxide and the alloy as shown in the Fig. 4a [10–12]. Secondly active elements alter the diffusion, so that the oxide layer grows from the metal surface, avoiding pores as shown in Fig. 4b [10]. Thirdly they react with impurities like S, C or P as illustrated in Fig. 4c. Otherwise, these impurities destabilize the oxide layer [11]. It is stated that yttrium is reducing the thickness of the protective oxide layer [13], leading to more efficient self-passivation. Apart from

Fig. 4. Ways of how yttrium effects the adhesion and stability of the oxide layer. (a) Pegs are improving the adhesion. (b) yttrium alters transport mechanism through the oxide, avoiding of pores. (c) Reaction of yttrium with impurity's, improves the purity of the Cr_2O_3 scale.

these studies, mostly concentrating on steels and nickel based alloys, several studies were already published investigating the possibility to improve the oxidation behaviour of tungsten by adding various elements like Si, Cr, Ti, Ta, Nb and Zr to name a few [9,14–20]. Also, Y was part of the elements which were screened in one of the studies. In this study on thin films W-14Cr-2Ti and W-18Cr-2Ti showed the best results on oxidation behaviour [7]. Nevertheless, this could be related to the near to optimum chromium content (as shown in Fig. 8 of \geq 12 wt%) of the investigated W-Cr-Ti alloys in combination with the low chromium and high yttrium content in the investigated W-7Y-3Cr alloy. These studies and others already indicate the beneficial effects of Cr as main passivating agent for W which is also used in this study.

For the oxidation behaviour two different models are used:

$$\left(\frac{\Delta m}{A}\right) \propto k_l \cdot t \tag{1}$$

Where Δm is the change of mass while oxidation, *A* the surface area of the specimen, and *t* the time.

Linear oxidation behaviour (Eq. (1)) takes place where the oxygen adsorption is the rate determining step, normally this is the case where the metal builds a porous oxide layer. Furthermore, the velocity remains constant over time and is given as k_l the linear oxidation constant for a certain temperature [10]. Pure tungsten exhibits such a linear oxidation behaviour for the temperature ranges and time intervals under consideration.

$$\left(\frac{\Delta m}{A}\right)^2 \propto k_p \cdot t \tag{2}$$

Parabolic oxidation behaviour (Eq. (2)) occurs when a metal forms a stable protective oxide layer on the surface, which decelerates the oxidation. After formation of a continuous oxide layer, the diffusion of reactants (cations and/or anions) through the growing oxide layer gets the rate determining step [10]. The resulting parabolic oxidation constant is given as k_p . In this study the beneficial effects of yttrium addition to the binary W-Cr alloys are further investigated.

2. Theoretical assessment

A key question on the feasibility of self-passivating tungsten alloys is being how the material composition of the first wall is changed during regular plasma operation and if the beneficial properties are preserved till the LOCA. To approach this issue a one-dimensional diffusion model is used with the boundary condition.

$$c(x=0) = c(x=d) = 0$$

Hence, the concentration c of the considered element on the surface of a layer is zero [21]. The concentration of the observed element depends on the time t and the position relative to the surface x, d is the thickness of a layer and c_0 represents the initial

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