

Oxidation resistance of bulk plasma-facing tungsten alloys

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ABSTRACT

Tungsten (W) currently is the main candidate as plasma-facing armour material for the first wall of future fusion reactors, like DEMO. Advantages of W include a high melting point, high thermal conductivity, low tritium retention, and low erosion yield. However, in case of an accident, air ingress into the vacuum vessel can occur and the temperature of the first wall can reach 1200 K to 1450 K due to nuclear decay heat. In the absence of cooling, the temperature will remain in that range for several weeks. At these temperatures the radioactive tungsten oxide volatilizes. Therefore, ‘smart’ W alloys are developed that aim to preserve the properties of W during plasma operation coupled with suppressed tungsten oxide formation in case of an accident.

This study focusses on oxidation studies at 1273 K of samples produced by mechanical alloying followed by field assisted sintering. In a first step the sintering is optimized for tungsten (W) – chromium (Cr) – yttrium (Y) alloys. It is shown that the best oxidation resistance is achieved with submicron grain sizes. This yields a closed, protective oxide layer. In a second step the influence of the grinding process during sample preparation is analysed. It is shown that scratches initiate failure of the protective oxide. In a third step the oxidation and sublimation is measured for weeks – for the first time the sublimation is directly measured in order to determine the potential hazard in comparison to pure W. It is shown that the oxidation is suppressed in comparison to pure W. However, sublimation at a rate of $1 \times 10^{-6} \text{ mg cm}^{-2} \text{ s}^{-1}$ starts after a few days. Nevertheless, the progress in smart alloys is evident: sublimation is delayed by about two days and complete mechanical destruction of the first wall is avoided.

1. Introduction

The future DEMONstration power plant (DEMO) aims to demonstrate the technological and economical feasibility of fusion as a future energy source. This implies that the power plant must be safe. This is very challenging as the plasma facing components will have to withstand an unprecedented fluence of particles, radiation, and neutrons. These conditions make tungsten (W) a prime candidate as plasma facing material. Advantages include a low sputtering yield by plasma particles, a good thermal conductivity, a low tritium retention, and a high melting temperature [1–3]. Further, the radioactive isotopes, which are generated by neutron irradiation, decay to shielded hands-on level within 100 years [4].

One important safety criterion is that there should be full passive safety in a Loss-of-Coolant-Accident (LOCA) with simultaneous air ingress. During operation neutrons activate the W, as soon as the cooling system fails it heats up due to its nuclear decay heat. Modelling predicts

a temperature rise to 1200 K to 1450 K where it remains for around 100 days [5]. In the presence of air, which may penetrate the vacuum vessel through leaks, the W oxidises rapidly at such temperatures. It forms radioactive WO_3 which then sublimates. Several hundred kg week⁻¹ sublimate at 1273 K and above [6]. On top of that, W, WO_3 have a Pilling-Bedworth ratio ($V_{\text{oxide}}/V_{\text{metal}}$) of 3.4 [7]. This can cause strong tensions, breaking the W and detaching it from the first wall.

The aim is to overcome these problems by developing a new W alloy. On the one hand it must retain the beneficial properties of W for operation of the power plant. On the other hand it must form a protective layer in case of a loss-of-coolant accident coupled with air ingress. This protective layer shall fulfill two goals: firstly, the formation of WO_3 must be suppressed so that no radioactive W is released to the environment. Secondly, the first wall shall not disassemble in an uncontrolled way and shall not pose additional challenges to potential emergency measures.

It was shown that alloys containing tungsten (W), 11.4 weight (

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wt.%) chromium (Cr), and 0.6 wt. % yttrium (Y) are a promising candidate [8–10]. In the following this is written as W-11.4Cr-0.6Y. The Cr was found to form a protective oxide scale on the surface suppressing the formation of WO_3 . The Y was found to be crucial to support the formation of this protective oxide scale. However, these studies were conducted with thin films on the timescale of a few hours. This study aims at investigating the oxidation and sublimation behaviour of these alloys over several weeks – a timescale relevant for DEMO. This requires a large material reservoir. One option for compacting bulk samples is Field Assisted Sintering Technology (FAST). In a first step, different FAST parameters are tested to sinter the novel W–Cr–Y alloys. In a second step, the influence of the grinding process in the surface of the sintered samples is studied. In a third step, the oxidation and sublimation characteristics for weeks in synthetic air at 1273 K are studied.

2. Experimental

To start bulk sample production, W, Cr, and Y powders are mixed and milled in a planetary ball mill, Retsch PM400. After 60 h of milling a homogeneous powder is achieved – more details on the mechanical alloying procedure can be found in ref. [9]. Consolidation is achieved by Field Assisted Sintering Technology (FAST), FCT HPD5, FCT Systeme GmbH [11]. The heating rate is chosen to be 100 K min^{-1} or 200 K min^{-1} , and a maximum temperature in the range of 1700 K to 1850 K is selected. The powder is poured in a standard graphite tool of 20 mm inner diameter. Previously, a graphite foil is placed in the inner face of the die to ensure a good electrical, thermal, and mechanical contact. Sintering is achieved by applying an uniaxial pressure (50 MPa), and heating using a current generated by a DC pulse generator.

The obtained sample is cut using a diamond blade saw and ground using a planar grinding and polishing machine. Unless stated differently, the final grinding step is performed using a P1200-grade silicon carbide abrasive paper. Grinding creates a reproducible surface finish by removing the first 0.5 mm where carbon from the foil diffused during sintering.

The density of the samples is measured using Archimedes principle. The mass is measured using the Sartorius MSA225P micro-balance (resolution $10 \mu\text{g}$), whereas the volume is determined by the mass of the displaced ethanol using the same micro-balance.

Oxidation is performed in a thermogravimetric analyser, the TAG-16/18 from Setaram. It allows to measure the in-situ mass change of a sample in a temperature range of 300 K to 2000 K. The drift of the scale for isothermal oxidations with the duration of several days is $2 \times 10^{-8} \text{ mg s}^{-1}$. This is possible due to a symmetrical, dual furnace design: one furnace contains the sample, the other furnace contains an inert product with the same volume to compensate buoyancy effects. The mass increase of the sample is measured by placing the sample into an alumina crucible which is attached to the balance by a platinum wire. The oxidation rate is determined by the mass increase of the sample as oxygen is added to the material.

The same device can also be used to measure sublimation. The sublimation rate provides an estimate for the radiological hazard. It is measured as illustrated in Fig. 1: an alumina tube is attached to the balance using a platinum wire. The sample hangs inside the tube attached to a second platinum wire. Thus, there is no direct contact between the sample and the tube at the balance. As soon as sublimation of

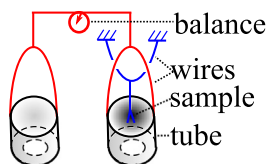


Fig. 1. Schematic of the sublimation measurement setup.

the sample starts, part of the material will deposit onto the alumina tube and cause a mass increase which is measured. The present work assumes that all material deposits onto the alumina tube and that no material sublimates from the tube due to its high porosity. Thus, the given sublimation rates are a lower boundary.

Electron microscopy provides insights into the grain structure and the structure and composition of the oxide layers. The combined Scanning Electron Microscopy (SEM) – Focussed Ion Beam (FIB) system Carl Zeiss CrossBeam XB540 equipped with an Energy Dispersive X-ray analysis (EDX) is used. The FIB allows to cut cross sections providing insights into the microstructure of a sample. During cutting the surface of the sample is damaged due to sputtering, therefore a layer of platinum (Pt) is deposited onto the sample before cutting.

The surface roughness is measured using the Dektak 6M from Bruker. A diamond-tipped stylus with a diameter of $2 \mu\text{m}$ is moved across the sample. The mechanical movement of the stylus yields a 2D surface profile.

3. Results

3.1. Material optimisation

There are four main methods to produce W–Cr–Y samples: Hot Isostatic Pressing (HIP) [12], pressureless sintering [13], Field Assisted Sintering Technology (FAST), and magnetron sputtering [6,8]. Magnetron sputtering allows quick production of a large number of samples. One obtains a thin film with a thickness of around $3.5 \mu\text{m}$ and a homogeneous distribution of W, Cr, Y, see Fig. 2a. The grain size is of around $0.1 \mu\text{m}$. Fig. 2e shows that this alloy forms a protective, closed Cr_2O_3 layer with some internal Cr_2O_3 when exposed to 1273 K at 20 kPa O_2 and 80 kPa Ar partial pressure. This protective oxide layer meets the main requirement – preventing the formation of WO_3 .

However, due to the small thickness the material reservoir of thin films is small: bulk samples are required for long-term studies. This study employs FAST where sintering happens within minutes and grain growth can be controlled. The main parameters that can be tuned are the heating rate h , the maximum sintering temperature T_{max} , and the holding time t_{hold} .

The first samples are produced with $h = 200 \text{ K min}^{-1}$, $T_{\text{max}} = 1823 \text{ K}$, and $t_{\text{hold}} = 1 \text{ min}$. A relative density of 98.5 % and large grains with a size of $1 \mu\text{m}$ is achieved. The oxidation performance is shown in Fig. 3: breakaway oxidation is found – breakaway oxidation is a short, temporal increase of the mass gain rate. The oxidation rate is still 1000 times slower than for pure W. However, further improvement is desired. Based on the experience with thin films, the following production steps aim at achieving a smaller grain size.

A smaller grain size can be achieved by reducing the sintering temperature T_{max} as grain growth slows down at lower temperatures. Different samples are prepared with fixed $T_{\text{max}} = 1733 \text{ K}$ and the following combinations of the heating rate h and the holding time t_{hold} :

- ($h = 100 \text{ K min}^{-1}$, $t_{\text{hold}} = 1 \text{ min}$) yields a relative density of 98 % and large grains with a size of $1.0 \pm 0.1 \mu\text{m}$, see Fig. 2b.
- ($h = 200 \text{ K min}^{-1}$, $t_{\text{hold}} = 1 \text{ min}$) yields a relative density of 99 % and medium grains with a size of $0.33 \pm 0.03 \mu\text{m}$, see Fig. 2c.
- ($h = 200 \text{ K min}^{-1}$, $t_{\text{hold}} = 0 \text{ min}$) yields a relative density of 99 % and small grains with a size of $0.23 \pm 0.03 \mu\text{m}$, see Fig. 2d.

The oxide structure after oxidation for 44 h is shown in Fig. 2f,g. The sample with large grains forms an oxide layer with a thickness of $8 \mu\text{m}$, see Fig. 2f. On top there is a $0.5 \mu\text{m}$ thick W–Y–O layer, almost fully covering the $0.3 \mu\text{m}$ thick Cr_2O_3 layer. Underneath, there is a thick, porous W–Cr–O layer. In contrast, the sample with small grains formed a closed, protective Cr_2O_3 layer with an average thickness $d = 1.3 \pm 0.3 \mu\text{m}$, as shown in Fig. 2g. The surface roughness (R_a), measured with the profilometer Dektak 6M (see section 2), is

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