Fusion Engineering and Design 133 (2018) 51–58

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

On the relation between microstructure and elastic constants of tungsten/steel composites fabricated by spark plasma sintering

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ARTICLE INFO

Keywords:
Plasma-facing materials
Tungsten/steel composites
Spark plasma sintering
Elastic constants

ABSTRACT

Tungsten/steel composites might play an important role in plasma-facing components, especially in joining of tungsten armor to structural parts made of steel. In this work, Young's moduli and shear moduli of a set of tungsten/P91 steel composites fabricated by spark plasma sintering were determined by resonant ultrasound spectroscopy. It was observed that the sintering temperature and the volume fractions of the individual phases have strong effects on the macroscopic elastic constants. The results were interpreted by means of a finite elements numerical model, showing that the regions of imperfectly bonded tungsten particles appearing in the microstructure of some of the composites act effectively as inclusions with very low elastic stiffness. A good correlation between the number of these regions in the microstructure and the elastic constants was observed.

1. Introduction

Extreme conditions in fusion reactors require the use of advanced materials with enhanced materials properties. Tungsten is considered as a primary candidate plasma-facing material due to its high resistance to sputtering, high melting point, and high thermal conductivity [1–3]. On the other hand, pure bulk tungsten cannot be usually utilized as a structural material due to its brittle nature with a high temperature of brittle-to-ductile transition, and due to difficult processing and machining. Therefore, to fulfill high demands put on the components of the fusion reactors, novel advanced materials have to be developed.

Plasma-facing components, primarily based on tungsten, have to be joined with the supporting structure. Due to the high amount of energy absorbed from the plasma, the heat has to be efficiently removed from the exposed surfaces of plasma-facing components, in order to minimize armor erosion [1]. In the current ITER design, the adjacent water-cooled heat sink is made of CuCrZr pipes, and joined to the tungsten divertor by a copper interlayer [4–8]. Similar divertor design with the water-cooled copper-based heat sink is also one of the options for future fusion devices, such as DEMO [8–10]. However, cracking and delamination can occur between the tungsten components and the copper-based heat sink due to a large difference in thermal expansion coefficients [11–14]. Alternatively, a helium gas-cooled modular divertor is considered, in which the tungsten components are joined with a steel heat sink [9,15–17]. Nevertheless, the difference in thermal expansion coefficients of tungsten and steel is also significant, which results in high stresses at the interface [15–18]. Therefore, in order to assure the best interconnection and to reduce stress concentration at tungsten/steel interfaces, several joining techniques of steel and tungsten have been presented [19]. Tungsten may be deposited on steel substrates as a plasma-sprayed coating [11,20–23]. Kruszewski et al. [18] designed possible bonding of tungsten and P91 steel elements through WL10 (W – 1 wt.% La_2O_3) interlayer and successfully fabricated W/WL10 and WL10/steel joints by pulse plasma sintering. Weber et al. [17] produced functionally graded tungsten/steel coatings by vacuum plasma spraying and magnetron sputtering. Zhong et al. [24] produced tungsten/ferritic steel joints with a Ni interlayer by diffusion bonding. Rasinski et al. [25] prepared Fe/W binary coatings by magnetron sputtering deposition technique. Powder metallurgy techniques have also been used. Qu et al. [26] used resistance sintering at ultra-high pressure. Despite very short processing times, intermetallic layer formation at the steel/tungsten interfaces was observed. Tan et al. [27] used mechanical alloying and spark plasma sintering. It was found that increased milling times lead to particle refinement and more homogeneous mixing, but increased amount of intermetallics upon sintering. In both cases, only very basic characterization was performed.

In this paper, tungsten/P91 steel composites fabricated by spark plasma sintering (SPS) [28–30] are addressed. In a fusion reactor (such as DEMO) with significant neutron fluxes, Ni-containing stainless steel cannot be used due to activation. Therefore, reduced activation ferritic-

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https://doi.org/10.1016/j.fusengdes.2018.05.056
Received 26 September 2017; Received in revised form 12 May 2018; Accepted 13 May 2018

martensitic steels such as Eurofer 97 [31,32] are foreseen as structural materials. P91 steel was chosen because of a wide availability and similar composition (~9% Cr) to Eurofer.

The aim of this paper is to show how the different processing conditions (different volume fractions of the constituents, sintering times and temperatures) affect the microstructure and, consequently, the macroscopic elastic constants of the composites, and to analyze the microstructural origin of these effects. In principle, the macro-scale mechanical performance of the composite results not only from the volume fractions of P91 and tungsten, but also from several microstructural features, such as micro-cracking, imperfect bondings of the particles, and formation of intermetallic phases. We show that all these features are sensitively reflected by the elastic constants, and use finite elements modelling (FEM) to identify the contribution of these individual features to the macroscopic elastic behavior.

2. Materials and methods

2.1. Materials fabrication and microstructural characterization

One P91 steel sample and seven types of tungsten/steel composites were consolidated by spark plasma sintering (SPS), using a 10-4 SPS device from Thermal Technology, USA (see [29] for more details). Powders of tungsten (Osram Sylvania, USA) and P91 steel (Karlsruhe Institute of Technology, Germany) were sieved to < 20 μm size. The powders were mechanically mixed at various volume ratios (ranging from 0/100 to 69/31, as summarized in the first column of Table 1), put into a graphite die lined with a graphite foil, and sintered in a pulsed current regime in He atmosphere of 50 Pa under a uniaxial pressure of 80 MPa. The sintering was controlled by a pyrometer sensing the process temperature in a hollow cavity made in the graphite die only about 1 mm from the sintered material. This characteristic processing temperature ranged from 1100 to 1400 °C, and the samples are always referred in this paper with the respect to this temperature. The sintering times were 2 min for all composites, except for the 53/47 composite sintered for 30 min, as seen in Table 1. The heating rate up to the maximum temperature and cooling rate down to 800 °C were 100 °C/min, followed by free cooling. The sintered compacts had a diameter of 19 mm and thickness of 3 mm; the surface affected by a contact with graphite was ground away.

The W/P91 volume ratio as well as the long sintering time for the last sample was chosen with the aim to maximize the formation of the Fe_7W_6 intermetallic compound in order to facilitate the characterization of its properties. This intermetallic phase typically appears in tungsten/steel composites between steel grains and tungsten grains due to diffusion [26–29], and it has much higher yield strength than steel and tungsten, but its thermal conductivity is low, which is unfortunately not desirable for plasma-facing components [29]. Moreover, as a typical intermetallic compound, Fe_7W_6 is brittle, and thus, its formation embrittles the resulting material [15].

The rapid growth of an intermetallic phase is one the features of spark plasma sintering of a mixture of materials. As shown by Anselmi-Tamburini et al. [33] for the case of sintering of Si and Mo layers, the electric current taking place during SPS enhances the growth rate of an intermetallic $MoSi_2$ phase. Similarly, in the papers by Kondo et al. [34] and Li et al. [35], the diffusion growth of thick intermetallic layers is observed after the SPS of samples consisting of two plates of different materials (Nb and C, or Fe and Al, respectively), with the strong dependence on sintering temperature and time. Moreover, the growth rate of an interphase layer is not linear, as the thickness of the layer follows square-root dependence in time [33–35]; therefore, the SPS of a powder mixture can lead to a formation of thick inter-granular layers only after a few minutes when utilizing a sufficient temperature. Such behavior was observed e.g. for the SPS of Ni and Ti powders [36], where thick layers surrounding the Ni grains were observed after 10 min of sintering; or for the SPS of CoNiAl and Ti powders [37], where intermetallic layers with the thickness of several μm were formed after the sintering for only 1 min.

The microstructures of the composites were studied by scanning electron microscopy (SEM, EVO MA15, Carl Zeiss SMT, Germany) at the polished cross-sections. The phase identification was performed using X-ray diffraction (D8 Discover, Bruker AXS, Germany); their volume fractions were determined by image analysis of the SEM micrographs.

Table 1
Volume ratios of W and P91 steel powders prior to sintering, sintering temperatures *T*, sintering times *t*, and phase compositions of sintered tungsten/steel composites.

Sintering parameters			Sintered compact		
vol. % W/P91	*T* [°C]	*t* [min]	vol. % W	vol. % P91	vol. % Fe_7W_6
0/100	1100	2	0	100	0
20/80	1100	2	11	68	21
43/57	1100	2	37	51	12
69/31	1100	2	61	26	13
69/31	1200	2	52	23	25
69/31	1300	2	47	9	44
69/31	1400	2	41	0	59
53/47	1400	30	15	0	85

2.2. Resonant ultrasound spectroscopy measurements

From the sintered pellets of tungsten/steel composites, samples with dimensions of approx. 2 mm × 3 mm × 4 mm were cut. Density of the composites was determined by weighting and measuring the volume of the samples. Room-temperature elastic properties were determined by the combination of pulse-echo method and resonant ultrasound spectroscopy. By the pulse-echo method, velocities of longitudinal and shear ultrasonic waves propagating through the samples were measured.

Laser-based contactless resonant ultrasound spectroscopy [38] was utilized for determining elastic constants of the tungsten/steel composites. In this method [39,40], elastic constants are determined by measuring resonant spectra of free elastic vibrations of small samples. The vibrations of the samples are generated by a pulsing infrared laser beam focused on one side of the sample, and they are detected on the other side by a scanning laser vibrometer. The resonant spectrum is then obtained by Fourier transform of the time-amplitude signals. The elastic constants are determined inversely, i.e. by an iterative comparison of the measured resonant spectrum with a resonant spectrum calculated from given elastic coefficients, density and dimensions of the sample. The final set of elastic constants is obtained after the sum of squared differences between measured and calculated resonant frequencies is iteratively minimized.

For all examined SPSed samples, more than 20 resonant frequencies were taken into account for the inverse calculation. It was assumed that the examined materials are macroscopically isotropic, i.e. they can be described by two independent elastic constants only: the Young's modulus *E* and the shear modulus *G*. This assumption was verified by measuring the longitudinal wave propagation velocity in directions perpendicular to the individual faces of the samples using the pulse-echo method. For all samples, the differences between velocity values obtained in different direction were smaller than the experimental accuracy of the pulse-echo method (~ $0.05\,mm{\cdot}\mu s^{-1}$), which confirms that the materials do not exhibit any measurable anisotropy. The velocities of ultrasonic waves measured by the pulse-echo method were also used as initial values for the RUS iteration and as a complementary information for the inverse procedure [38].