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Manufacturing and Characterization of PIM-W Materials as Plasma Facing Materials

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Abstract. Powder Injection Molding (PIM) was used to produce pure and particle reinforced W materials to be qualified for the use as plasma facing material (PFM). As alloying elements La_2O_3, Y_2O_3, TiC, and TaC were chosen with a particle size between 50 nm and 2.5 µm, depending on the alloying element. The fabrication of alloyed materials was done for different compositions using powder mixtures. Final sintering was performed in H_2 atmosphere at 2400 °C resulting in plates of $55 \times 22 \times 4$ mm^3 with ~98 % theoretical density.

The qualification of the materials was done via high heat flux testing in the electron beam facility JUDITH-1. Thereby, ELM-like 1000 thermal shock loads of 0.38 GW/m^2 for 1 ms and 100 disruption like loads of 1.13 GW/m^2 for 1 ms at a base temperature of 1000 °C were applied. The obtained damage characteristics, i.e. surface roughening and crack formation, were qualified versus an industrially manufactured pure reference tungsten material and linked to the materials microstructure and mechanical properties.

1. Introduction

Within the framework of the EUROfusion Consortium, high heat flux materials development for the first wall of a DEMO reactor and in particular for the high heat flux regions of the divertor is one key topic. Thereby, the focus for the plasma facing materials was in the past [1] and still is strongly set on tungsten and tungsten-based materials. Tungsten-based materials are attractive to be used in various applications for fusion power plants. Besides its advantages, the main drawbacks are its brittleness and correlated difficult machinability as well as the narrow range of possible alloys using the commercial fabrication route. In addition, the semi-finished products are limited to plates and rods. These issues are addressed by the development and fabrication of complex near-net shaped tungsten parts via Powder

Injection Molding (PIM), which also allows the joining of different materials without brazing and a rapid development of new tungsten materials [2, 3].

The R&D on new tungsten materials aims to improve its resistance to operational loads, i.e. thermal shock and thermal shock induced thermal fatigue loads mainly due to ELMs [4-8] and pure thermal fatigue loads during steady state heat loading [9, 10]. While the resistance to ELMs is a purely material dependent issue, the resistance to steady state heat loads also strongly depends on the component design. In this work the focus is set on the development of materials with an increased resistance to ELMs. Therefore, various particle reinforced PIM-W materials with the alloying elements La_2O_3, Y_2O_3, TiC, and TaC were manufactured using powder mixtures of commercially available powders.

Characterization of the materials was performed determining the materials microstructure and hardness and by performing high heat flux testing in the electron beam facility JUDITH-1 [11]. Thereby, ELM-like 1000 thermal shock loads of 0.38 GW/m^2 for 1 ms at a base temperature of 1000 °C were applied. These application relevant conditions are slightly above the damage threshold for commercial reference tungsten grades, allowing the qualification of the individual materials towards this reference material. Based on this pre-qualification, disruption like tests with 100 pulses at 1.13 GW/m^2 for 1 ms and at 1000 °C base temperature were performed on selected materials (pure W, W-2Y_2O_3, W-1TiC, W-2TaC). In all cases, the damage quantification was done by post-mortem examinations using metallographic means.

2. Materials - manufacturing and microstructure

2.1. PIM: used powders, powder mixtures and feedstock preparation

The average particle size distribution of the used tungsten powder was in the range of 1.0 to 2.0 µm Fisher Sub-Sieve Size (FSSS). As doping materials La_2O_3 (FSSS < 2.50 µm), Y_2O_3 (FSSS < 1.50 µm), TiC (FSSS < 50 nm), and TaC powders (FSSS < 1.0 µm) were used. These individual powders were mixed with pure tungsten to obtain various powder compositions defined by wt.-%: pure W with two different powder mixtures (W-170, W-5050), W-2La_2O_3, W-0.5/1/2Y_2O_3, W-1/1.5/2/3TiC, and W-0.5/1/2TaC.

After a heat treatment at 80 °C for moisture removal, the powders were dry mixed to form the "feedstock" with a 50 vol.-% binder system in a kneader. The finished granulated feedstock was homogeneous and free from agglomeration.

2.2. PIM: production of plates and microstructure

The injection molding of the "green parts" (consisting of powder and binder) was carried out on an ELEKTRA injection molding machine. After injection molding the green parts were debound. During the debinding step not only binder and impurities (mainly O and C) were removed, but also the high residual stresses generated during injection molding were released. Finishing of the plates was done in a subsequent sintering process in dry H_2 for 2 h. The finished plates (Figure 1) are characterized by a shrinkage of nearly 20 % during the heat-treatment process and achieve ~ 98 % T.D after sintering at 2400 °C.

The materials microstructure is in all cases isotropic, but varies among the different material compositions strongly with regard to their average grain size (figure 1). The smallest grains were found for Y_2O_3 and TiC doped materials, for which the average grains size is, depending on the composition, only by a factor 2 to 7 larger than the original size (average: 1.5 µm) of the W-powder. This suggests that these materials should be also highly recrystallization resistant. All other materials exhibit an increase in grain size by more than a factor 16. In particular, for the TaC doped materials this result is surprising as a similar performance to TiC additions would have been expected, even if larger particle sizes were used (see section 2.1). For the La_2O_3 doped material it has to be stated that the sintering temperature was above the melting temperature of La_2O_3 and in the microstructural analysis no La_2O_3 was found, but very clean grain boundaries with regard to impurities.

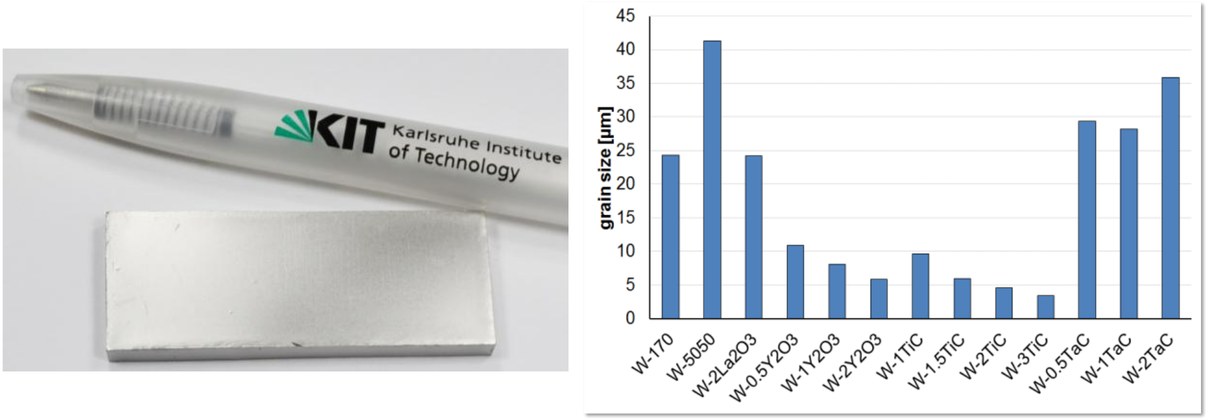

Figure 1: Finished plate with dimensions of 55 × 22 × 4 mm^3 and a weight of ~75 g; average tungsten grain size of the produced PIM-W materials.

2.3. Reference material

The reference material is pure tungsten grade (IGP W) with a purity of 99.97 wt% manufactured by the Plansee AG, Austria, according to the ITER material specification. The forged rod was produced with the dimensions 36 × 36 × 480 mm³ and was exposed to a stress relieving treatment by thermal annealing after the production process. Due to the production process the grain structure is strongly anisotropic with needle like grains parallel to the forging direction (∅ = 5 – 10 µm, length ca. 25 µm). In its recrystallised state (1600 °C for 1 h) the materials loses its pronounced elongated grain structure and experiences grain growth with an average grain size of about 64 µm × 102 µm.

3. Experiments

The simulation of transient thermal shocks was done with the electron beam device JUDITH-1 at Forschungszentrum Jülich [JUDI] on small-scale samples. These samples were prepared via electrical discharge machining (EDM) from the PIM-materials with dimensions of 10 × 10 × 4 mm³. In comparison, due to the anisotropic microstructure, the tests on the reference W-grade were performed on specimens from the stress-relieved material in two different orthogonal orientations (elongated grains parallel and perpendicular to the loaded surface), both with dimensions of12 × 12 × 5 mm³. Similar sized specimens were tested for the recrystallized material only in one orientation. The final preparation of the specimens was done by polishing the surface to a mirror finish. This allows obtaining well defined starting conditions, in particular in view of expected surface damage, i.e. roughening and crack formation.

The chosen testing conditions were based on experience with other tungsten materials and in view of relevant operational scenarios in a fusion reactor. Therefore, for the ELM-like loading an absorbed power density of 0.38 GW/m² (taking an electron absorption coefficient of 0.55 into account) a pulse duration of 1 ms, a pulse number of 1000, and a base temperature of 1000 °C was chosen. These conditions are expected to be above the expected damage threshold of the materials to allow quantification of the damage and qualification of the material. Furthermore, applying 1000 pulses addresses the influence of thermal fatigue on the damage behavior. Based on this qualification, disruption like tests with 100 pulses at 1.13 GW/m^2 for 1 ms and at 1000 °C base temperature were performed on selected materials (pure W, W-2Y_2O_3, W-1TiC, W-2TaC). The exposed area for all tests is 4 × 4 mm² with a focused electron beam (∅ ~1 mm) at very high frequencies (40 kHz in x-direction and 31 kHz in y-direction). The repetition frequency was < 1 Hz and defined by requirement to allow a cool down back to base temperature.

After the exposure in JUDITH-1, the induced surface damage was investigated by SEM, determining the amount of crack formation, and laser profilometry for quantification of the surface roughness. Subsequently, the cross sections of the samples were investigated by metallographic means in order to analyse particularly crack propagation into the bulk material. Supporting experiments were done by measuring the materials Vickers hardness (HV1; max. load of the indenter: 2 N) and for a few materials also the stress-strain behavior via 4-pt. bending tests between RT and 400 °C. The latter were done with a strain rate of 0.033 mm/min on 12 × 1 × 1 mm³ specimens.

4. Results

When dealing with thermal fatigue induced surface damage due to ELM-like loading conditions, a clear indicator of the amount of plastic deformation is the surface roughness. Based on an original value of R_a = 0.08-0.1 µm for the mirror like surface of the specimens, the surface roughness increased for all specimens (figure 2a). In comparison to the reference material, characterized by a roughness of ~0.6 µm in its stress relieved state, part of the Y_2O_3 doped and the TiC-doped materials showed a better performance, i.e. lower surface roughness. Taking the performance of the recrystallized reference material into account (Ra = 1.33 µm), all PIM-W grades showed an improved performance. One factor thereby is the material's grain size, as indicated in figure 2b. By correlating grain size and obtained surface roughness a clear tendency is found that, within certain variations, smaller grains also result in a reduced amount of surface roughening.

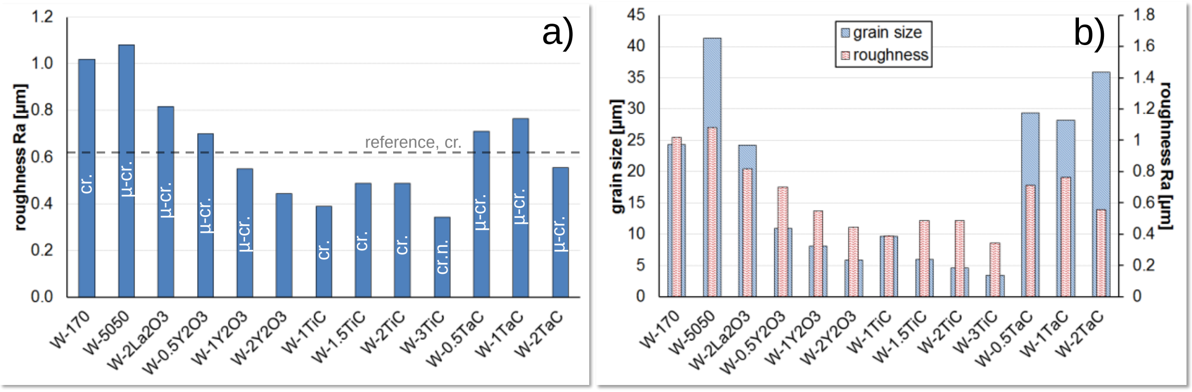

Figure 2: Determined roughness of the PIM-W materials a) in relation to the reference material (stress relieved; recrystallized: R_a = 1.33 µm) including observed crack formation (µ-cracks, cracks and crack network) and b) in relation to the materials grain size

In addition to surface roughening, for almost all tested PIM materials, except W-2Y_2O_3, the loaded surface was characterized by thermal fatigue induced crack formation. This was ranging from µ-cracks in areas of local inhomogeneity or at grain boundary triple junctions to crack formation along grain boundaries and formation of crack networks (figure 2a). The penetration of the cracks towards the bulk is limited to a few tens µm in the worst case, which is based on the lack of materials brittleness at the chosen base temperature and the still comparably low number of pulses [7, 8]. In correlation, for the reference material shallow cracking along grain boundaries but no crack network formation was found, which is shown in figure 3 in relation to the PIM materials with the best performance for a certain material composition / particle reinforcement.

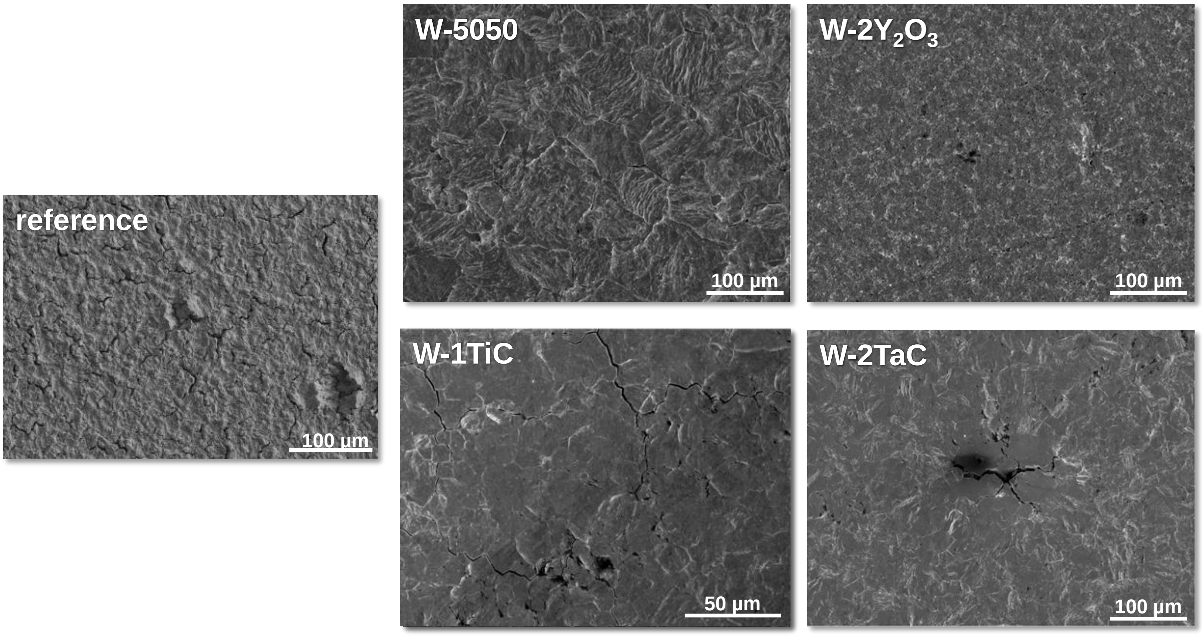

Figure 3: SEM images of the loaded surface: comparison between the reference tungsten grade with grains perpendicular to the loaded surface (stress relieved) to the materials with the best performance for a particular particle reinforcement

Besides the above mentioned correlation between grain size and surface roughening, there seems to be also a link between the former and the mechanical properties of a material. By measuring the material's hardness as an indicator for the material's strength, figure 4 shows that a decreasing grain size and accordingly a decreasing surface roughness is directly related to an increase in hardness. By comparison to the reference material (stress relieved, 445 HV1), only the TiC-doped materials with particle additions of ≥ 1.5 wt.% exhibit a significant increase in hardness while the hardness for W-1TiC and the best performing PIM-W grade W-2Y_2O_3 is in the same range as the one for the reference material. For all other W-grades, i.e. the pure W and W-TaC doped materials, hardness values were obtained in the proximity of those for the recrystallized reference material (355 HV1).

In related 4pt.-bending tests of the PIM-W grades shown in figure 3, pure W and W-2TaC performed similar with regard to yield and bending strength and in particular pure W showed already some ductility at 100 °C. In contrast, W-1TiC and W-2Y_2O_3 showed ductile behavior first at 200 °C and 300 °C, respectively, but with a significantly higher yield and bending strength, which is at 300 °C for W-1TiC and W-2Y_2O_3 about 3.5 and 5.5 times larger, respectively, than those for pure W (~200 MPa). In comparison to reference tungsten, on which tensile tests were performed with a deformation speed of 0.2 mm/min, the yield strength was about 550 MPa [12].

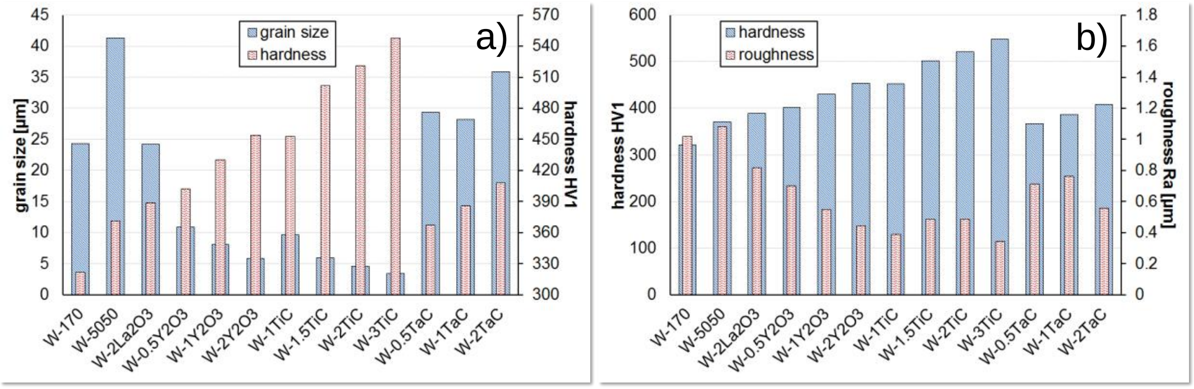

Figure 4: Hardness of the PIM-W materials in relation to a) the grain size and b) the surface roughness after thermal shock; hardness of reference material: 445 HV1 and 355 HV1 in the stress relieved and recrystallized state, respectively

In contrast to ELM-like loading, during disruption like testing the melting threshold of the material is exceeded. This results for the four investigated materials in either homogeneous (pure W, W-1TiC and W-2TaC) or inhomogeneous (W-2Y_2O_3) molten and resolidified structures (figure 5). The reason for the inhomogeneous melting might be related to the comparably low melting temperature and binding energy for Y_2O_3 compared to TiC and TaC and the increased probability for the formation of tungsten oxides. Furthermore, large and, despite of the high base temperature, kind of brittle crack formation was found for those materials exhibiting homogeneous melting with a crack depth of up to several mm. In case of W-1TiC and W-2TaC, these cracks were not limited to the molten area but also protruded towards the non-loaded but, due to lateral heat transfer, heat affected zone surrounding the loaded spot.

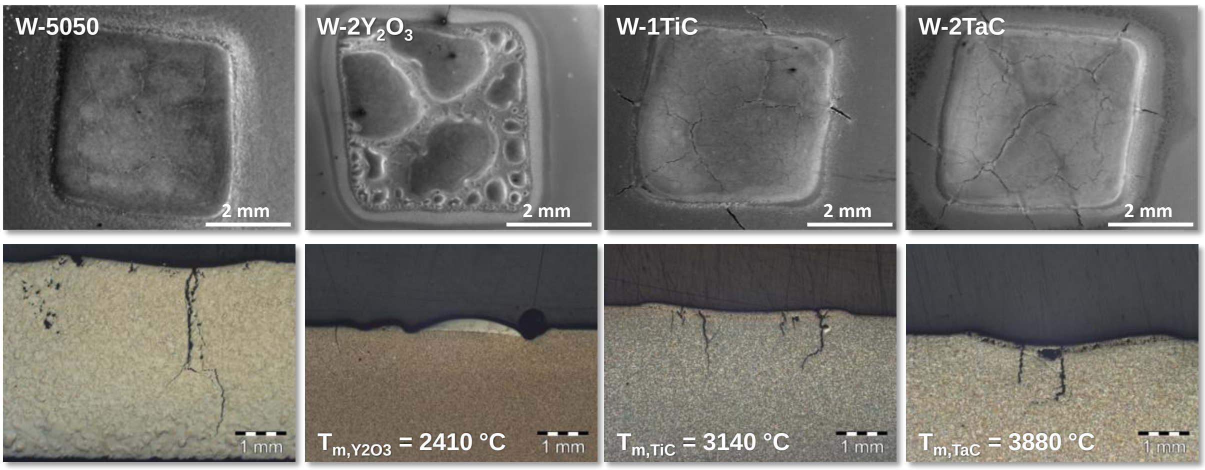

Figure 5: SEM images of the surface (top) and LM images of the cross sections (bottom) after disruption like loads (P_{abs} = 1.13 GW/m^2, Δt = 5 ms, n = 100, T = 1000 °C)

5. Summary

For the qualification of new W-grades manufactured via PIM, the materials were subjected to 1000 and 100 pulses of ELM and disruption like loading conditions, respectively, in the electron beam facility JUDITH-1. The results for ELM-like tests have shown that the occurring thermal fatigue damage, i.e. roughening and crack formation, is depending on the grain size of the material and the correlated mechanical properties. Lower grain sizes and therefore higher hardness / mechanical strength results in decreased roughening and therefore an increased resistance to thermal fatigue damage in view of the expected number of ELM pulses in ITER and beyond. Accordingly, economically viable W-materials with Y_2O_3 (< 1.50 µm) and TiC (< 50 nm) content that have been produced by using standard industrially available powders, have shown an improved performance compared to a reference industrially manufactured tungsten grade fulfilling the ITER requirements. In contrast, W reinforced with TaC (< 1.0 µm) particles performed similar to the reference while pure W exhibited similar properties and results as the reference material after recrystallization.

The results for the disruption like tests have shown, that melting, melt motion and crack formation is related to the added particle reinforcement. Threreby, Y_2O_3 doped materials have shown inhomgeneous melting and resolidification but hardly any crack formation while all other materials are characterized by homogeneous melting with strong crack formation up to several mm depth. A decisive role seems to play the melting temperature and binding energy. In view of the performance in a tokamak and with regard to the qualification of Y_2O_3 and TiC doped W-materials further investigations are needed to determine if extensive crack formation or inhomogeneous melting poses the higher risk for plasma operation and component lifetime.

6. Acknowledgement

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