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Development of brazing foils to join monocrystalline tungsten alloys with ODS-EUROFER steel

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Abstract

Results on rapidly solidified filler metals for brazing W with W and monocrystalline W with EUROFER steel (FS) are presented. Rapidly quenched powder-type filler metals based on Ti_{bal}–V–Cr–Be were developed to braze polycrystalline W with monocrystalline W. In addition, Fe_{bal}–Ta–Ge–Si–B–Pd alloys were developed to braze monocrystalline W with FS for helium gas cooled divertors and plasma-facing components. The W to FS brazed joints were fabricated under vacuum at 1150 °C, using a Ta spacer of 0.1 mm in thickness to account for the different thermal expansions. The monocrystalline tungsten as well as the related brazed joints withstood 30 cycles between 750 °C/20 min and air cooling/3–5 min.

1. Introduction

The targeted development of helium cooled high performance divertors for fusion DEMO reactors requires in present design concepts the selection and joining of refractory materials and advanced oxide-dispersion-strengthened, reduced activation ferritic steels. The development of tungsten armor for plasma-interactive components is thoroughly considered in [1–3]. However, tungsten and ferritic steel have significant differences in their coefficients of thermal expansion ($\alpha_W = (4.3–6.0)\ 10^{-6}\,K^{-1}$, $\alpha_{FS} = (10.5–12.3)\ 10^{-6}\,K^{-1}$). Thus the joining of such different materials is a serious problem, as the joint is exposed to mechanical and thermal loading not only during the long term operation of the divertor but also during the manufacturing. Various concepts to overcome this problem have been already published [4–6].

For a number of applications, a suitable brazing has demonstrated significant advantages in comparison with fusion and pressure welding [3,7,8]. Rapidly solidified amorphous and microcrystalline foil-type filler metals represent a promising approach for the joining of dissimilar materials [4,6–8]. For example, the brazing of powder metallurgy tungsten and monocrystalline tungsten to a Cr13Mo2NbVB ferritic/martensitic steel was carried out using a rapidly solidified 35–40 μm thick foil-type filler metal (Ni_{bal}–15Cr–4Mo–4Fe–(0.5–1.0)V–7.5Si–1.5B) in vacuum at 1150 °C, using a 0.5 mm thick spacer made of a 50Fe–50Ni alloy to

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decrease the residual thermal stresses in the brazed joint [6]. Thermocycling tests (100 cycles under the regime of heating to 700 °C for 20 min – air and water cooling for 3–5 min) have resulted in the formation of slightly branching cracks in powder metallurgy tungsten along the whole sample length at about 100 μm from the seam, i.e. in the zone of the highest tensile stresses. It is necessary to point out that the monocrystalline W/FeNi/EP-450 brazed joint microstructure has high stability during the thermocycling.

However, the use of a nickel spacer in the brazing zone is not desirable, as it can promote helium accumulation by (n,α) reactions under irradiation and a subsequent embrittlement enhancement of the brazed joint. It is not desirable to use a tungsten powder either. Therefore, an attempt has been made within the framework of the present work to join tungsten with EUROFER steel using newly developed brazing alloys based on radiologically more acceptable elements, to select a different spacer material, and to develop other powder filler metals for W–W joints.

2. Experimental procedure

2.1. Materials

To study the possibilities of making a non-detachable joint between ODS-EUROFER97 steel (FS) with tungsten of two grades, polycrystalline tungsten plates of technical purity (99.95), obtained by powder technology and rolling, and monocrystalline tungsten with [1 1 1] orientation along the contact surface of a 99.98 purity were chosen as materials for brazing. Ta was chosen as a spacer material for the compensation of residual thermal stresses ($\alpha_{Ta} = (6.6–8.0)\ 10^{-6}\,K^{-1}$). ODS-EUROFER97 is a reduced activation ferritic martensitic 9Cr–WMnVTa steel used as reference material for ITER test blanket modules and for DEMO in vessel components.

Filler metals were developed in two directions [9]: Ti-based alloys were used for brazing in the W||W and W||Ta systems; and Fe-based alloys for brazing Ta||FS. After the analysis of the related phase diagrams, the Ti-based alloys were prepared with a variable composition of Be as follows: 1 = Ti_{bal}–22.5Cr–7.5V–1Be; 2 = Ti_{bal}–22.5Cr–7.5V–1.5Be; 3 = Ti_{bal}–22.5Cr–7.5V–2.0Be; 4 = Ti_{bal}–22.5Cr–7.5V–2.5Be; 5 = Ti_{bal}–22.5Cr–7.5V–3Be. The Fe–Ta based alloys were fabricated with a variable composition of Ge, Si, and B: 1 = Fe_{bal}–18Ta–10Ge–2.0B; 2 = Fe_{bal}–18Ta–6Ge–4Si–2Pd–2.0B; 3 = Fe_{bal}–18Ta–6Ge–4Si–2Pd–2.5B; 4 = Fe_{bal}–18Ta–8Ge–2Si–2Pd–1.75B; 5 = Fe_{bal}–18Ta–8Ge–2Si–2Pd–2.0B; 6 = Fe_{bal}–18Ta–8Ge–2Si–2Pd–3.0B. To determine the melting points of these alloys and brazing temperatures, a differential calorimetric analysis, the results of which are shown in Fig. 1, was carried out. As seen from Fig. 1, the melting points of Ti-alloys have fixed values in the 950–1050 °C temperature range. Melting of the Fe–18Ta system alloys has a complicated two-phase character. It starts in the 890 (alloy 1) −960 °C (alloy 6) range and ends at about 1100 °C. The completion of melting for alloy 6 was not found by that point. Therefore, to decrease the melting point of the alloy, its B-concentration was

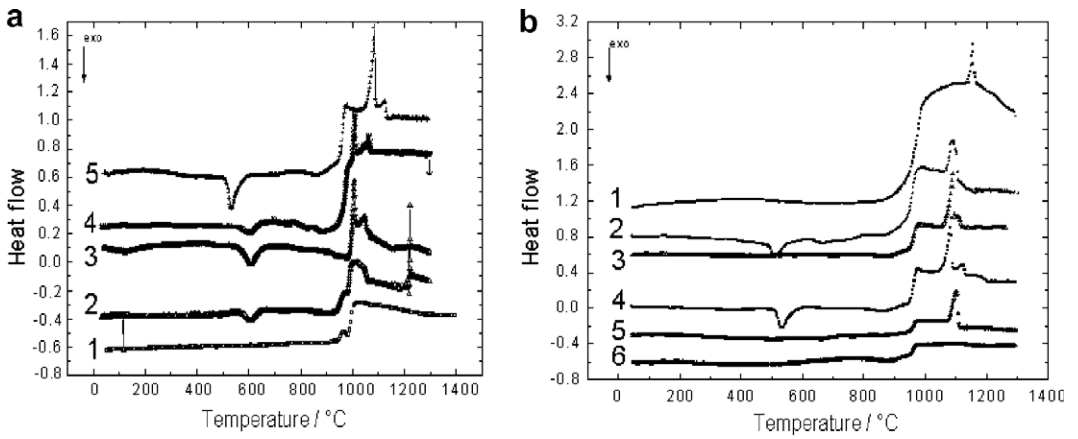

Fig. 1. Curves of differential calorimetric analysis: (a) 1 – Ti_{bal}–22.5Cr–7.5V–1Be; 2 – Ti_{bal}–22.5Cr–7.5V–1.5Be; 3 – Ti_{bal}–22.5Cr–7.5V–2.0Be; 4 – Ti_{bal}–22.5Cr–7.5V–2.5Be; 5 – Ti_{bal}–22.5Cr–7.5V–3Be. (b) 1– Fe–18Ta–10Ge–2.0B; 2 – Fe–18Ta–6Ge–2.0B–4Si–2Pd; 3 – Fe–18Ta–6Ge–2.5B–4Si–2Pd; 4 – Fe–18Ta–8Ge–1.75B–2Si–2Pd; 5 – Fe–18Ta–8Ge–2.0B–2Si–2Pd; 6 – Fe–18Ta–8Ge–3.0B–2Si–2Pd.

increased up to 3.5 wt%. The peaks frequently appearing in Fig. 1 in the temperature range 500–600 °C are not material specific but are correlated with the packing of the brazing foils in the calorimetry device.

Taking these results into consideration, as well as the experimental results on wetting by melted W, Ta and FS filler metals, the Ti based filler metal Ti_{bal}–22.5Cr–7.5V–3Be (FM-Ti) was selected for brazing W‖Ta, and the Fe based filler metals Fe_{bal}–18Ta–8Ge–2Si–2Pd–3.5B (FM-FeTa) was used for brazing FS‖Ta.

Fragments of brazing foils in the form of 30–50 μm thick ribbons were obtained by 'melt spinning' (Forschungszentrum Karlsruhe, Germany) and 'planar flow casting' (MEPhI, Russia) methods in controlled atmosphere (Ar or He, or vacuum) [6]. After fabrication, the fragments were powdered in a planar ball mill up to 300 μm in size. After sieving, the powders were used for making polyvinyl alcohol based pastes.

2.2. Brazing technique

The assemblies for brazing are shown in Fig. 2. The samples for brazing had the following geometry: FS, $25 \times 5 \times 5$ mm^3; W, $25 \times 1.5 \times 5$ mm^3; and the thickness of a Ta-spacer was 0.1 mm. The FS, Ta and W samples were collected in a special conductor. A 0.5 mm thick paste was applied onto surfaces to be brazed. The Ta-spacer (Fig. 2) was placed between the W and the FS at a pressure of 0.5 MPa. The applied heat treatment was the following: heating to 1150 °C with a rate of 20 °C/min – holding for 15–20 min (W_M‖W_P), 30 and 60 min (W_M‖Ta‖FS) – furnace cooling to 600 °C – holding 60 min – furnace cooling to room temperature in vacuum of 6.6×10^{-3} Pa.

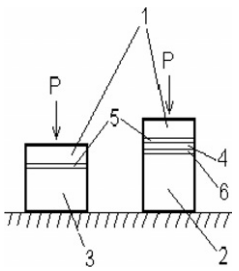

Fig. 2. Assemblies for brazing 1 – monocrystalline W; 2 – EUROFER steel; 3 – polycrystalline W; 4 – Ta-spacer; 5 – Ti–22.5Cr–7.5V–3.0Be; 6 – Fe–18Ta–8Ge–2Si–2Pd–3.5B.

2.3. Thermocycling tests of samples and investigation of the brazing zone structure

After grinding and polishing, microstructural analyses of the brazed joint samples have been performed using metallographic and electron-microscopic methods, as well as micro X-ray spectrum analysis of the chemical element distribution in the brazing zone [6,7].

Thermocycling tests of brazed samples were performed using a hermetic, argon filled stainless steel capsule. The capsule was argon arc welding and exposed to thermocycling tests as follows: 30 cycles of heating to 750 °C for 20 min followed by air and cooling for 3–5 min. Microstructural investigations and X-ray spectroscopy of brazed joints were carried out after the thermocycling tests.

3. Results and discussion

An overview of a brazed 'monocrystalline W‖polycrystalline W' (W_M‖W_P) joint after thermocycling is shown in Fig. 3. An analysis of metallography and the distribution of chemical elements has shown that the Ti–22.5Cr–7.5V–3Be filler metal is compatible with monocrystalline W and has a flat contact surface. The brazed joint demonstrates that there is a strong erosion of the surface and the diffusion zone in polycrystalline powder W. The penetration of Ti, V, and Cr from the filler metal into the powder W occurs along grain boundaries and defects up to 10–15 μm in depth. Estimated diffusion coefficients are of the order of 5×10^{-8} m^2 s^{-1} (1150 °C). W-penetration into the melted zone of the filler metal has also been established. X-ray spectrum analysis reveals a light seam zone (W in Fig. 3) with W impregnated with the filler metal. The W-particles in the filler metal arise from physicochemical interaction between the filler metal and the rough W-surface formed as a result of the surface grinding. An increase of the brazing time from 15 to 20 min and the thermocycling practically do not change the brazing zone microstructure.

An overview of the brazed 'W_M‖Ta‖FS' joint obtained with the use of filler metals Fe_{bal}–18Ta–8Ge–2Si–2Pd–3.5B before thermocycling is shown in Fig. 4. The brazed joint consists of two brazed zones and a Ta-spacer. The FS‖FM-FeTa‖Ta (interface 1) brazed zone has a complicated structure depending on the composition of the filler metal. With regard to Fe_{bal}–18Ta–8Ge–2Si–2Pd–3.5B filler metal (Fig. 4), its intensive interaction

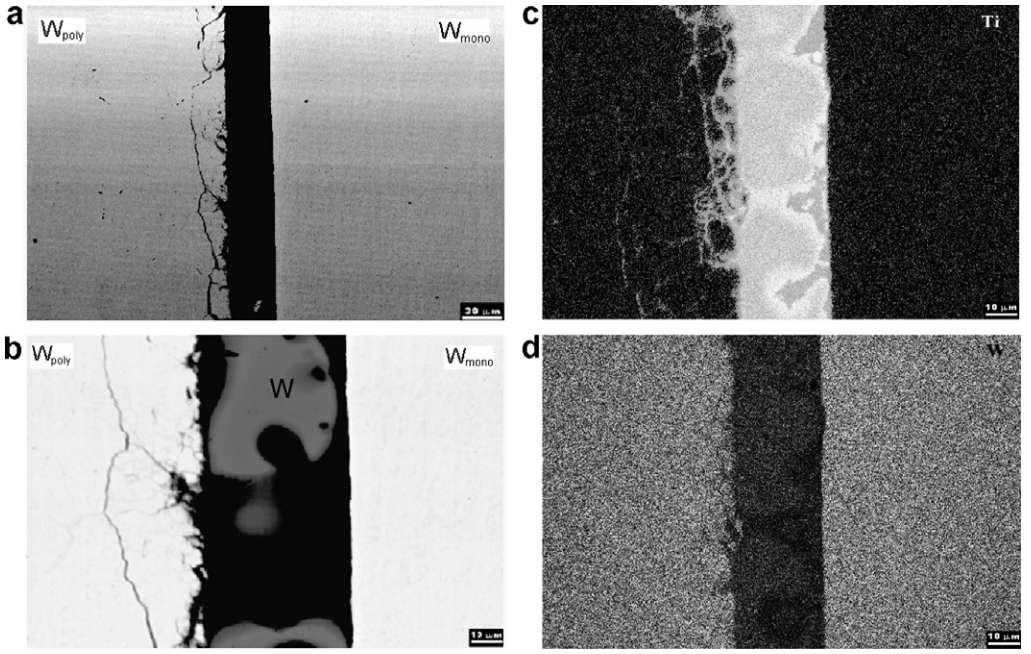

Fig. 3. Structure of the $W_P \| W_M$-brazing zone after thermocycling: (a) Microstructure of a W_P(left)$|W_M$(right) joint, (b) microstructure of a W_P(left)$|W_M$(right), (c) Cr-distribution in the brazing zone section and (d) W-distribution in the brazing zone section.

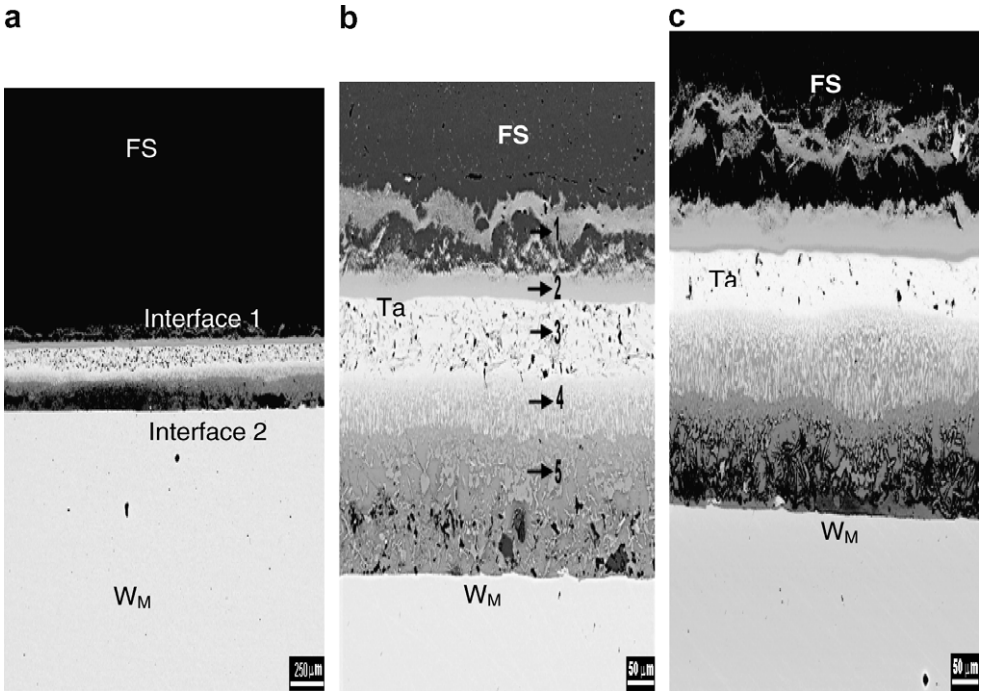

Fig. 4. Microstructure of the FS$\|$Ta$\|W_M$ joint (a) structure of the FS$\|$Ta$\|W_M$ joint, (b) microstructure of a brazed joint (the brazing time is 30 min, 1150 °C) and (c) microstructure of a brazed joint (the brazing time is 60 min, 1150 °C).

with EUROFER steel resulted from the high B-concentration [6]. As seen from Fig. 4(b) and (c), a wide interaction zone of the filler metal with the steel (area 1) and dissolution of the Ta-spacer in the filler

metal with formation of a solid solution of Ta in Fe (area 2), based on FeTa and Fe_2Ta are observed. In this case, a stratification of the filler metal into two parts (areas 1 and 2 in Fig. 4(b)) and transfer of Ge, Si and Pd in FS have been found. An increasing brazing time from 30 to 60 min (Fig. 4(c)) results in a further dissolution of a stratified part of the filler metal in the steel and in a broadening of the interaction zone of Ta with Fe.

The W‖FM-Ti‖Ta (interface 2) brazed seam shows a weak interaction of the filler metal with monocrystalline W and an intensive Ta-dissolution (up to 49 wt% of Ta) in the Ti–22.5Cr–7.5V–3Be filler metal. This results in formation of a transition area consisting of two areas (4 and 5). Area 4 contains a high concentration of Ta (to 84–89 wt%) and the following elements of the filler metal: Ti (8%), Cr (4.2%) and V (1.15%). Area 5 is an area of the filler metal enriched with Ta having the composition 48%Ta–36%Ti–12%Cr–3.6%V–0.5%W.

Microstructure investigations after the thermocycling tests showed that the brazed joint did not have any cracks. Its microstructure changes depend on brazing time. There are practically no significant microstructure changes in the brazed joint after a 30 min brazing. During the $20 \leftrightarrow 750$ °C thermocycling, the porosity of non-brazed areas in the $Ta‖FM‖W_M$ seam became enlarged after the brazing for 60 min. No microcracks were observed in this case. Therefore, the $FS‖Ta‖W_M$ brazed joint demonstrated its working capacity in conditions of the thermocycling used.

During the irradiation in a Demo-reactor, helium can be accumulated in the brazing zone by (n,α), $(n,n^I\alpha)$ and $(n\gamma)$ reactions taking place for a number of elements of the filler metal.

As the neutron induced production rates for helium accumulation are higher in Be and B, reliable neutronics calculations are even more important for filler metals containing these elements. At a given irradiation temperature, the sensitivity to helium embrittlement depends besides the helium production rate and on the local helium bubble morphology which in turn depends on the initial Be or B distribution as well as on the local microstructure. That is, for a reliable evaluation of helium effects in the brazing zones of promising composites, neutron irradiation of materials in test reactors with subsequent post irradiation experiments need to be done in the future.

4. Conclusion

To braze monocrystalline W with powder metallurgy W and with ODS-EUROFER97 steel, a new complex of rapidly solidified powder filler alloys based on Ti and (Fe–Ta), as well as a technology for brazing in vacuum at 1150 °C, using a 0.1 mm thick Ta-spacer for decreasing the residual thermal stresses in a brazed joint, have been developed.

No brazed joints with monocrystalline W have been broken during the thermocycling tests in an inert atmosphere (30 cycles under the regime of heating to 750 °C for 20 min – air cooling for 3–5 min). It is necessary to point out that the monocrystalline W‖Ta‖FS brazed joint microstructure has a high stability during the thermocycling.

A significant physicochemical interaction of the Fe_{bal}–18Ta–8Ge–2Si–2Pd–3.5B filler alloys with ODS-EUROFER97 steel by a high activity of B has been established. Therefore, in the future it is planned to decrease the brazing time and carry out reactor irradiation of the brazed assemblies.

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