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Development of rapidly quenched brazing foils to join tungsten alloys with ferritic steel

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Abstract

Results on rapidly solidified filler metals for tungsten brazing are presented. A rapidly quenched foil-type filler metal based on Ni_{bal}-15Cr-4Mo-4Fe-(0.5-1.0)V-7.5Si-1.5B was developed to braze tungsten to ferritic/martensitic Crl3Mo2NbVB steel (FS) for helium gas cooled divertors and plasma facing components. Polycrystalline W-2CeO₂ and monocrystalline pure tungsten were brazed to the steel under vacuum at 1150 °C, using a 0.5 mm thick foil spacer made of a 50Fe-50Ni alloy. As a result of thermocycling tests (100 cycles between 700 °C/20 min and air-water cooling/ 3-5 min) on brazed joints, tungsten powder metallurgically processed W-2CeO₂ failed due to residual stresses, whereas the brazed joint with zone-melted monocrystalline tungsten withstood the thermocycling tests. © 2004 Elsevier B.V. All rights reserved.

1. Introduction

The targeted development of a helium cooled high performance divertors requires the selection and combination of refractory materials and advanced oxidedispersion-strengthened, reduced activation ferritic steels. In ITER parts of the baffle, as well as the upper region of the inner and outer vertical targets of the divertor will be armored with tungsten. 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. Thus the joining of such different materials is a serious problem, as it is exposed to mechanical and thermal loading not only during the long term operation of the divertor but also during the manufacturing. It can be solved by various ways [4,5].

Brazing demonstrates the significant advantage in comparison with fusion and pressure welding [3,6]. Rapidly solidified amorphous and microcrystalline foiltype filler metals represent a promising approach for the

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joining of dissimilar materials [4,6,7]. They offer a number of advantages – extremely high chemical and phase uniformity, narrower melting and solidification ranges, 'instantaneous' melting across the whole width and thickness of the foil, and outstanding ductility. Amorphous foils bend 180° without fracturing and thus accommodate complex joint geometries. The rapidly solidified filler metal STEMET[®] has been used to join dissimilar PFC materials, including Cu and graphite, Mo and graphite, V and graphite, Be and Cu, V and Be, and SS and Be [6,7].

From these viewpoints, the present authors' studies on rapidly solidified brazing foils have been carried out to manufacture non-detachable tungsten-ferritic-martensitic steel joints for the gas-cooled divertor of a DEMO thermonuclear reactor, of which the simplest cell scheme is shown in Fig. 1.

2. Experimental procedure

2.1. Materials

To study the possibilities to make a non-detachable joint of a type EP-450 ferritic/martensitic



Fig. 1. Schematic of a divertor target for DEMO.

(Crl3Mo2NbVB) steel with tungsten of two grades, polycrystalline tungsten plates (W–2%CeO₂), obtained by powder metallurgy and rolling, and zone-melted monocrystalline tungsten of a 99.99 purity with [111] orientation along the contact surface were chosen as materials for brazing. A 50%Fe–50%Ni (FeNi) alloy was chosen as a spacer material to compensation of residual thermal stresses. Properties of these materials are presented in Table 1.

Based on Ni_{bal}-15Cr-4Mo-4Fe-(0.5-1.0)V-7.5Si-1.5B, Ti_{bal}-22.5Cr-7.5V and Ti_{bal}-28V-3.5Be alloys (in wt%), 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].

Based on preliminary tests of brazed joints and the results of a differential-thermal analysis, a STEMET 1309 Ni_{bal}-15Cr-4Mo-4Fe-(0.5-1.0)V-7.5Si-1.5B (ST-9) 35-40 µm thick filler metal was chosen and manufactured to braze tungsten to steel with an acceptable melting point of 1120 °C.

2.2. Brazing technique

The assemblies for brazing are shown in Fig. 2. In contrast to Fig. 2(a), a chromium coating of a



Fig. 2. Schematic of an assembled cross-section for brazing.

thickness $\leq 10 \ \mu\text{m}$ was applied onto the surface of monocrystalline tungsten in Fig. 2(b). The samples for brazing had the following geometry: ferritic/martensitic steel, $40 \times 7.5 \times 5 \ \text{mm}^3$; tungsten, $40 \times 7.5 \times 1.5 \ \text{mm}^3$; and the thickness of a 50Fe–50Ni spacer was 0.5 mm. The steel and tungsten samples were collected in a special conductor. The filler metal (ST-9) was adjusted to a spacer by point electro-welding from both sides. The spacer (Fig. 2) was placed between the tungsten and the steel at a pressure of 0.5 MPa. The applied heat treatment was: heating to 1150 °C with a rate of 20 °C/min – holding for 20 min; – cooling to 650 °C; – holding for 180 min; – furnace cooling to room temperature ($\gg 10 \ \text{h}$) in a vacuum of $6.6 \times 10^{-3} \ \text{Pa}$.

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

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

Thermo-cycling tests of brazed samples were performed using a hermetic, argon filled stainless steel capsule. The capsule was welded in an arc furnace in argon and exposed to thermocycling tests, as follows: 100 cycles of heating to 700 °C for 20 min followed by air- and water cooling for 3–5 min. Repeated metallography and X-ray spectroscopy of brazed joints were carried out after the thermocycling tests.

3. Results and discussion

An overview of a brazed 'polycrystalline W-2%CeO₂ – steel' joint before thermocycling is shown in Fig. 3(a).

Table 1 Mechanical and thermophysical properties of the different materials (300 $^{\circ}$ C)

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Alloy	$\alpha \ (10^{-6} \ \mathrm{K}^{-1})$	E (GPa)	$\sigma_{\rm B}~({\rm MPa})$	δ (%)	Poisson's ratio, v
W-2CeO ₂	4.3-5.0	342-400	600–900	0–5	0.26-0.35
\mathbf{W}^{a}	4.3	390-420	960	2-18	0.24-0.26
FeNi	6.0–9.4	160	450	35	_
EP-450	11.0	200	700-800	10-18	0.23-0.31

^a Monocrystalline.



Fig. 3. Cross-section of a polycrystalline W/FeNi/EP-450 joint: (a) general view, (b) polycrystalline W–2CeO₂/FeNi joint and (c) polycrystalline W–2CeO₂/FeNi/EP-450 joint after thermocycling (20–700 $^{\circ}$ C).

The brazed joint consists of two brazed seams and a FeNi spacer. The FeNi/EP-450 (interface 2) brazed seam is uniform and has no peculiarities. The W/FeNi brazed seam has a two-layer structure (Fig. 3(b)). X-ray spectrum analysis reveals a light seam zone (zone 1) with tungsten impregnated with a filler metal, and the dark zone (zone 2) consists of the filler metal components with particles of pure tungsten. The pure tungsten particles in the filler metal arise from physicochemical interaction between the filler metal and the rough tungsten surface formed as a result of the surface grinding. Scanning electron microscopy has shown that the technical powder rolled tungsten used has a significant part of the second phase particles CeO_2 (Fig. 3(b)). The developed character of the W/FeNi interface (interface 1) demonstrates that there is a strong erosion of the tungsten surface.

After thermocycling as a result of induced thermal stresses, the formation of slightly branched cracks shows up in tungsten along the whole sample length, at the distance of about 100 μ m from the brazed seam (Fig. 3(c)).

The appearance of these micro-cracks has a number of causes. The rolled tungsten has a layer structure and low (practically zero) ductility (Table 1) in the direction perpendicular to that of rolling. In addition, residual thermal stresses form a complicated stressed-deformed state (SDS) during cooling of the brazed joint because of the differences in the thermal expansion coefficients (α_W and α_{FS}), Young's modulus (E_W and E_{FS}), and Poisson's ratios of tungsten and steel. The SDS of a brazed joint can be demonstrated by finite-element methods [8,9], or estimated by calculation of the material resistance to deformation. It is also possible to calculate the distribution of stresses. Both methods result in the formation of a triaxial stress state in brazed materials. Obviously, because of the triaxial stress state and small thickness of the interlayer $(h_{\text{Bz}} \ll h_{\text{W}} < h_{\text{FS}}$ here $h_{\text{FS}} = h_{\text{EP}} + h_{\text{FeNi}}$ its relaxation ability is not very pronounced [10] and its elastic characteristics were close to that of the steel (Table 1). The normal stress (σ), perpendicular to the plate of a brazed joint (Fig. 4(a)), has a maximum in a material with less plasticity that is located at some distance (h_{CR}) from the zone of brazing (Fig. 4(b)). The distance depends on the thickness ratio of brazed materials ($\chi = h_{FS}/h_W$) [9].

A simple estimation of the deformation resistance in a brazed seam, when $h_W \ll h_{FS}$ (Fig. 4(a)) and the interlayer properties are close to that of the steel, gives the following estimations of residual thermal stress level. Any cooling of brazed joints during thermocycling tests ($\Delta T \cong 700$ °C) results in the formation of compression stresses in tungsten owing to a difference of thermal expansion coefficients ($\alpha_{FS} - \alpha_W$) and Young's modulus for tungsten E_W and the steel E_{FS} :

$$\sigma_{y} = \sigma_{z} = -(\alpha_{\rm FS} - \alpha_{\rm W})\Delta T E_{\rm W} (1 + E_{\rm W} h_{\rm W} / E_{\rm FS} h_{\rm FS})^{-1}.$$
(1)

The absence of deformation constraint in cross direction results in the formation in this direction (x) of tensile strains:

$$\varepsilon_{\rm x} = 2\nu_{\rm W}(\alpha_{\rm FS} - \alpha_{\rm W})\Delta T (1 - E_{\rm W}h_{\rm W}/E_{\rm FS}h_{\rm FS})^{-1}.$$
 (2)

An estimation according to Eq. (2) gives $\varepsilon_x \approx 0.005-0.006$. It is an unacceptable high elastic deformation for tungsten. According to the second theory of strength for this ε_x of uniaxial tension, an equivalent tensile stress in tungsten will be equal to:

$$\sigma \cong -2\nu_{\rm W}\sigma_y = -2\nu_{\rm W}\sigma_z$$

= $-2\nu_{\rm W}(\alpha_{\rm FS} - \alpha_{\rm W})\Delta T E_{\rm W}(1 + E_{\rm W}h_{\rm W}/E_{\rm FS}h_{\rm FS})^{-1}.$ (3)

An estimation of σ from Eq. (3) shows that under the assumption of full absence of relaxation the value of



Fig. 4. Distribution of normal thermal stresses in a brazed joint: (a) method of joining and (b) distribution of the σ -stress from the joint height.

normal thermal stresses in W/FeNi+EP450 can attain 800 MPa, which is comparable to the ultimate strength of polycrystalline W-2%CeO2 (Table 1), where, as seen from Fig. 4(b), the maximum of equivalent tensile stress is in a less ductile material (tungsten) at the h_{CR} -distance from the joint. In the process of brazing a noticeable relaxation of thermal stresses takes place of course owing to plastic deformation of the steel and the spacer, but nevertheless the level of residual normal thermal stresses, acting in tungsten, seems to exceed the ultimate strength of powder metallurgical tungsten. It is important that the resulting compressing stresses of σ_v and σ_z in tungsten, which are maximum at $0.05 < \chi < 0.5$ [9], prevent the deviation of the crack moving direction from that which is parallel to the joint surface. On the other hand, the crack extension direction was predetermined by the initial rolling texture of tungsten plates. Because of this, the peeling of tungsten is parallel to the joint surface and takes place at some distance from it $(h_{\rm CR} \approx 100 \ \mu m$ for chosen thicknesses).

The general view of a monocrystalline W/FeNi/EP-450 brazed joint is shown in Fig. 5. To decrease chemical erosion, monocrystalline samples were thoroughly polished and a chromium layer ($\leq 10 \mu$ m in thickness) was applied onto the contact surface. It has significantly improved the structure and the contact surface topography. The W/FeNi brazed seam has a three-layer structure. An X-ray spectrometry analysis has shown that the 15–25 µm thick light zone of a brazed seam is a diffusion area of the interaction of W, Cr, Fe with Ni. The W/FeNi brazed seam (a zone between interfaces 1 and 2) does not contain tungsten particles.

No brazed joints with monocrystalline tungsten were broken during the thermocycling tests. It is important to note that the structure of a brazed seam of monocrystalline W/FeNi/EP-450 is also stable during thermocycling tests.



Fig. 5. Cross-section of a monocrystalline W/St-9 zone after the thermocycling tests (20-700 °C).

4. Conclusion

The brazing of polycrystalline $W-2\%CeO_2$ and monocrystalline pure tungsten to a Crl3Mo2NbVB ferritic/martensitic steel was carried out by 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 decrease the residual thermal stresses in a brazed joint.

No brazed joints with monocrystalline tungsten have been broken during the thermocycling tests in an inert atmosphere (100 cycles under the regime of heating to 700 °C for 20 min – air and water cooling for 3–5 min). It is necessary to point out that the monocrystalline W/ FeNi/EP-450 brazed joint microstructure has a high stability during the thermocycling.

A significant physicochemical interaction of the filler metal with powder metallurgical tungsten $(W-2CeO_2)$

has been established. The thermocycling tests have resulted in formation of a slightly branching crack in tungsten along the whole sample length at about $100 \,\mu m$ from the seam, i.e. in the zone of the highest tensile stresses.

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