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Thermomechanical Characterization of Joints for Blanket and Divertor Application Processed by Electrochemical Plating

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Fusion technology requires in the fields of first wall and divertor development reliable and adjusted joining processes of plasma facing tungsten to heat sinks or blanket structures. The components to be bonded will be fabricated from tungsten, steel or other alloys like copper. The parts have to be joined under functional and structural aspects considering the metallurgical interactions of alloys to be assembled and the filler materials. Application of conventional brazing showed lacks ranging from bad wetting of tungsten up to embrittlement of fillers and brazing zones. Thus, the deposition of reactive interlayers and filler components, e.g. Ni, Pd or Cu was initiated to overcome these metallurgical restrictions and to fabricate joints with aligned mechanical behavior.

This paper presents results concerning the joining of tungsten, Eurofer and stainless steel for blanket and divertor application by applying electroplating technology. Metallurgical and mechanical characterization by shear testing were performed to analyze the joints quality and application limits in dependence on testing temperature between room temperature and 873 K and after thermal aging of up to 2000 h. The tested interlayers Ni and Pd enhanced wetting and enabled the processing of reliable joints with a shear strength of more than 200 MPa at RT.

Keywords: Joining, shear testing, tungsten, Eurofer, electroplating, interlayer

1. Introduction

Tungsten and its alloys are designated in fusion technology to be applied as plasma facing materials with functional background, e.g. due to the high sputtering resistance and excellent heat conductivity or as structural alloy in the field of He-cooled divertor development, where heat loads of up to 15 MW/m^2 should be removed. Whereas DEMO relevant divertor design based on water cooling will have a heat sink fabricated from copper alloys. Looking onto the blanket designs, the structural material of the first wall will be of type Eurofer steel, which may be covered by tungsten to enhance its performance [1,2].

Analyzing such design concepts, different alloys will be applied and have to be joined. Combinations will range from tungsten with itself or vs. Eurofer steel over steel type joints (if intermediate connections are also considered, e.g. for mechanically or thermally graded applications) up to joints with copper alloys. Brazing of He-cooled divertor components had shown that tungsten joints easily suffer from reliable wetting due to passivating surface scales or missing miscibility if, e.g. copper is used as filler metal [3] or from not desirable reactions of filler and base material forming brittle layers [1]. To overcome such restrictions, electrochemical plating tools were developed to deposit reactive interlayers, e.g. on tungsten together with filler component copper to process promising and reliable joints for fusion relevant alloy combinations [4]. In the first development phase the comparison of brazing and diffusion bonding as joining methods was in the foreground. This contribution will focus mainly on the characterization of the processed joints in the state as fabricated and also after annealing to point out an eventually appearing aging behavior. The mechanical performance of the produced joints was analyzed by shear testing at different temperatures to give hints for application ranges.

2. Experimental procedure

2.1 Fabrication of joints

2.1.1 Selection of interlayers

Brazing tests of tungsten (W) with itself or other materials showed that insufficient wetting easily occurred if copper fillers were applied as usual in standard industrial processing without active components. Thus, an applicable tool to overcome this lack will be the deposition of reactive interlayers on top of the parts to be joined. Elements like Ti, V, Cr, Fe, Pd or Ni promise the ability to activate surfaces during joining processes as it can be evaluated from phase diagram analyses [3], e.g. due to miscibility or phase formation with tungsten compared to pure copper – tungsten reactivity. Electrochemical plating was chosen as deposition technology for reactive interlayers due to its common use in industrial applications, its ability to coat complex shapes, good controllability of layer thickness by processing time and plating current, and also considering costs of future industrial application.

For analyzing the general applicability and demonstrating the benefits of electrochemical plating in joining technology, Pd and Ni were selected as master elements for interlayers. Both can be processed from aqueous electrolytes similar to copper which is used as filler component for these joining tests.

2.1.2 Electrochemical plating

The cylindrical samples used for the joining tests had a diameter of 8 mm and length of 10 and 26 mm and

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were fabricated from rod material by electro discharge machining (EDM) and turning. After joining the samples were characterized mechanically by performing shear tests at room temperature (RT) and elevated temperatures of up to 873 K. Fig. 1 shows schematically the deposition sequence of the interlayers and filler components on the cross sectional area and additionally an image of a pair of coated pieces. However, a general requirement for application of coating technologies and also for electrochemical plating as tool for deposition of reactive interlayers and/or filler components is the adherence of the deposited layers on the surfaces of the substrates. Especially tungsten will be covered ab initio due to its physicochemical nature at least by oxide / hydroxide scales and sometimes also by layers coming from machining (e.g. Zn impurities from EDM cutting). Such scales have to be removed and also a surface activation has to be performed to increase adhesion before functional layers for joining are deposited. The etching process given in [4] applying $K_3[Fe(CN)_6]$ * KOH as reactant was used for the tungsten conditioning. Eurofer (E) and stainless steel (S, DIN 1.4571) surfaces were cleaned by etching to remove oxides based on (Fe, Cr, Ni) in a solution of H_2SO_4 - HCl - H_2O at the ratio of 9 / 18 / 73 for 5 minutes at 50°C and of HNO_3 - HF - H_2O at the ratio of 20 / 3 / 77 for 30 seconds at 50°C, respectively.

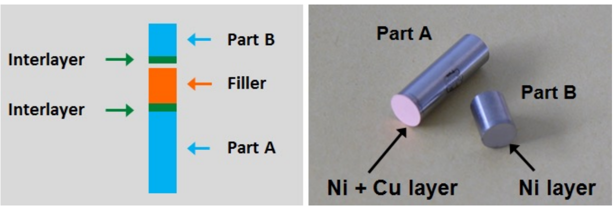

Fig. 1. Electroplating procedure and plated parts for joining.

The selected interlayers Ni and Pd were deposited by electroplating from aqueous electrolytes. The electrolyte used for Ni plating consisted of a mixture of nickel sulfamate with a Ni^{2+} content of 76 g/l, boric acid and a fluorinated surfactant. A consumable Ni electrode was applied for keeping the Ni concentration constant during the electroplating. Homogeneous and well adherent layers were obtained applying current densities near 10 mA/cm^2 and a working temperature of about 50°C (Fig. 2). The pH of the electrolyte was held constant in a range of 3.3 to 3.5. The layer thickness was about 10 μm for the test series to activate the reaction with the filler.

Pd deposition was done using a commercially available electrolyte from AMI DODUCO company, which is designed to work near a pH value of 7.5. The current densities were in the range of 0.4 to 0.8 mA/cm^2 and an insoluble Ti plated Pt anode was used. Deposition experiments of Pd directly on tungsten have shown that the parameter window is rather narrow to obtain well adherent layers due to low hydrogen overvoltages favoring H_2 formation and therefore increased palladium hydride formation. This risk can be reduced by covering the tungsten surface by seeds or a thin Cu layer of sub micrometer dimension.

The main filler component Cu was subsequently electroplated onto the reactive interlayers. For electrodeposition an electrolyte based on $CuSO_4$ was used, as it is described in literature [4]. The Cu plating was conducted at a current density of about 30 mA/cm^2. A consumable Cu anode was used as reservoir to keep Cu concentration constant during electroplating. Homogeneous layers could be processed in the thickness range of 5 to 200 μm in reproducible manner by simple control of deposition time. A layer of 70 μm could be deposited within roughly 1 hour as used for the brazing tests. The characterization of deposited layers showed well adherence and layer structures without voids as visible from Fig. 2 for Ni electroplated on tungsten or reported in more detail in [5].

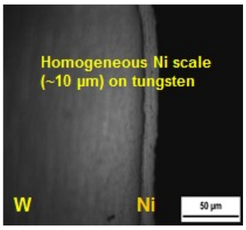

Fig. 2. Micrograph of Ni interlayer deposited on tungsten with homogeneous and defect free structure.

2.1.3 Joining of samples

The pieces coated with interlayer and filler were mounted for joining in a brazing support to guarantee best axial alignment of the assembled parts. The brazing temperature was selected well above the melting point of copper (T_M = 1084°C / 1357 K) at, e.g. 1150°C (1423 K). The brazing was performed in a tubular furnace under inert argon gas atmosphere and the support system was moved by a manipulator system into the hot brazing zone. The whole duration of brazing, consisting of heating up and dwell time, lasted 10 min for processing at 1423 K. The temperature of the support system was monitored.

Fig. 3 shows joined samples fabricated for the thermomechanical characterizations. Visible is that some small traces of the filler were pressed out from the brazing gap. This material formed droplets indicating that melting temperature of the filler was clearly exceeded.

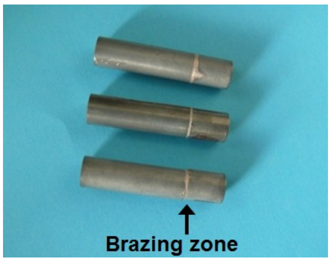

Fig. 3. Brazed samples for thermal treatments and shear testing.

The performed metallographic examinations revealed that a homogeneous gap filling took place together with wetting of the surface by the filler independently of alloy type, i.e. tungsten, stainless steel or Eurofer. More detailed information concerning the reactions in the brazing gap was collected by performing SEM (Scanning Electron Microscopy) and EDX analyses (Energy Dispersive X-Ray spectroscopy). Fig. 4 shows a line scan across the brazing zone of a tungsten – Eurofer joint. The Ni interlayer reacts with both materials as it can be seen by the rounded steps of the concentration curves for W or Fe as main Eurofer component in the contact zone of the parts joined. Between the interlayer and copper diffusion was also monitored which leads to the formation of solid solution of Ni-Cu. The degree of mixing is dependent on brazing temperature and time. Metallographic examinations and SEM analyses confirmed that brazing by electroplated Ni and Cu layers leads for both materials tungsten and Eurofer to good and reproducible wetting as main criteria for bonding.

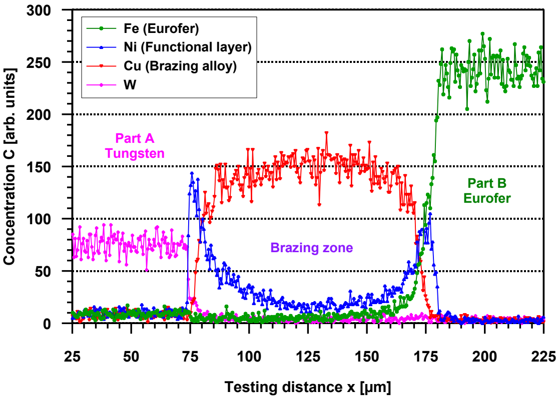

Fig. 4. Line scan along the cross section of a brazed W – E joint with interlayers of Ni and filler component Cu.

2.1.4 Thermal treatment of joints

The microstructural analyses pointed out, that interlayers with an adjustable thickness and a well adherent quality electroplated on tungsten, stainless steel and Eurofer react during brazing with the base materials and the main filler component. The reaction of the interlayer with the base materials and filler are dependent on temperature and reaction time. This means that the mixing of the elements in the brazing zone will proceed with time and temperature due to diffusion. The concentration profile directly after brazing is shown in Fig. 4. Thus, mechanical characteristics of the joint may change during operation at elevated temperatures. For quantification of such aging behavior brazed samples were annealed in evacuated quartz tubes at assumed operation temperature of 973 K for durations ranging from 200 to 2000 hours. The aged samples were characterized by shear testing and metallographic tools similarly as the “as-brazed” samples.

2.2 Mechanical testing

The mechanical stability of the processed samples will be essential for the application in future fusion plants beyond of the homogeneous wetting by the filler components during joining. Cross sections of brazing zones showed that the requirement of homogeneous gap filling is achieved [4], however, the mechanical behavior has to be qualified by separate tests. Shear testing was chosen as first qualification tool due to the simple preparation and testing configuration.

2.2.1 Shear testing

The tests were performed in an universal mechanical testing machine “Instron 4505“ equipped with a furnace for high temperature application as shown in Fig. 5a. Shear testing was performed in a temperature range from room temperature (RT) to 873 K under air for W – W joints and up to 723 K for the other combinations. The specimens had a cylindrical shape and consisted of two parts, which are brazed together as illustrated in Fig. 1. The long part was inserted into the specimen holder and fixed by two screws. A stamp is moved downwards and positioned at the short part of the specimen. To prevent a too large bending moment during the test, the lever arm has to be kept small. For this purpose a gap between the specimen holder and the stamp is set to about 1 mm. It should be assured that the brazed seam is localized within the gap and therefore completely free for shearing off along the joint interface during the test. The detail of the experimental setup is depicted on the schematic diagram in Fig. 5b. For high temperature testing, the specimens are heated by a radiant heater. During heating the specimens are subjected to a constant preload of 0.03 kN. The temperature is controlled by NiCr-Ni thermocouples. The shear tests with a displacement rate of 0.01 mm/s are conducted after the specimen is homogeneously heated up to the desired temperature. The test is stopped if cracking or to large bending of the samples is monitored.

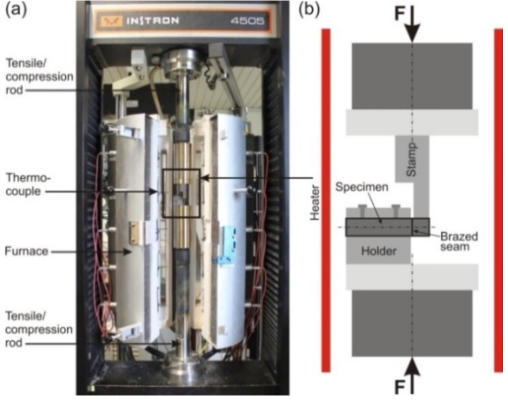

Fig. 5. (a) The used universal mechanical testing machine equipped with a high temperature furnace and force loading pistons, (b) schematic diagram of the experimental setup

3. Thermomechanical results and discussion

The three different types of joints, W – W, E – E and S - S, were analyzed concerning shear strength. The samples were tested in the state as brazed and aged at 973 K. Testing conditions was RT and elevated temperatures.

3.1 Behavior of joints at RT

The overall behavior of the shear tested joint combinations was not uniform concerning alloy combinations of joint, pretreatment and testing temperature. An overview for shear tests at RT is given in Tab. 1. The failure of the W-rod appeared at room temperature due to its high ductile to brittle transition temperature (DBTT) as known by charpy tests of W alloys [6]. Other effects may be excluded due to the fact that the brazing was performed below recrystallization temperature of W for short time

Table 1: Fracture characteristics and observed highest loads of different combinations of joints during shear testing at RT.

Material combination	Fracture characteritics	Highest Loads
W – W	Brittle fracture of W base material	11kN
E – E	Shearing-off in the brazing zone	12kN
S – S	Bending	9kN
W – E	Shearing-off in the brazing zone	9kN
W – S	Shearing-off in the brazing zone	9kN

and that no changes of the microstructure, e.g. by recrystallization, were observed. The parts exhibited irregular structure at the fracture area and partly cracks penetrated into the brazing zone. The filler showed ductile behavior in such case [5]. Thus, only a lower shear strength limit of about 230 MPa can be given for W – W joints at RT.

Stainless steel – stainless steel joints showed only bending of the samples without failure in the brazing zone. In contrast to these combinations joints of type E – E showed shearing-off at RT without bending or cracking of the samples. The failure occurred in the brazing zone with ductile fracture behavior of the filler and without detachment from the parts.

3.2 Influence of annealing temperature and elevated test temperatures

In contrast to the shear testing at RT, 'classical' shearing of the parts was observed independently of alloy combinations for elevated testing temperatures. No cracking of the tungsten rods took place at 723 and 873 K compared to the RT testing.

Eurofer joints were the only ones which showed shearing-off in the brazing zone at RT, thus this type may be a benchmark for comparing different testing conditions. Shear loads were found to be near 12 kN at RT for Ni activated surfaces of E – E joints with main filler component copper in the state as-brazed. Lower shear strength was measured after aging of the joints at 973 K. The values observed were near 7.5 kN as shown in Fig. 6. The macro- and microstructural analyses did not indicate that defects were growing during annealing. This can be extracted from the brazing zone inspection performed by X-Ray tomography before annealing and from the optical appearance of the shear tested brazing zone as shown in Fig. 7. Defects visible have not grown during annealing but they will reduce the value observed by the ratio of active joined area to unbraced ones. Thus, the reduction in shear strength extrapolated to defect free joints will be less as implied by Fig. 6. Surely, diffusion processes appeared in the brazing zone during aging leading to an increase of the interlayer element in the main filler component copper. This behavior was found at all combinations tested. Fig. 8 illustrates this for the example interlayer of nickel, filler copper and steel parts by EDX line scans determining the concentration profiles for the state as joined and after aging. Clearly visible is that the Ni concentration in the center of the filler increased by aging. Shearing of joints was observed near 4 kN loads for testing at elevated temperature of 723 K. The value given in Fig. 6 for Eurofer joints is also representative for bonded tungsten and stainless steel parts at same testing conditions. The reduction in strength to about one third of the RT value is plausible and corresponds with the yield strength reduction of copper [7] the main filler component in dependence of testing temperature. Tungsten – tungsten joints were additionally tested at 873 K due to the higher strength of the base material. The shear strength was near 20 MPa or a load of 1 kN was needed to observe shearing-off for this type of joint. The damage of the joint appeared in the filler and the surfaces of the parts were well covered by a filler layer indicating that the Ni interlayer activated the surfaces well for reaction with the filler.

3.3 Behavior of mixed joints

Under this aspect joints were fabricated with different material combinations: W – E and W – S with nickel as interlayer. Shear testing was performed at RT and 723 K with an upgraded clamping support. Both ends of the sample were mechanically fixed by screws to reduce further bending during shear loading. At RT sliding was observed at loads of 9 kN (see Tab. 1). The strength degreased for both configurations at testing temperature of 723 K to roughly 100 MPa or a load of 5 kN. These tests confirmed that the joining by electroplating is also applicable for mixed combinations

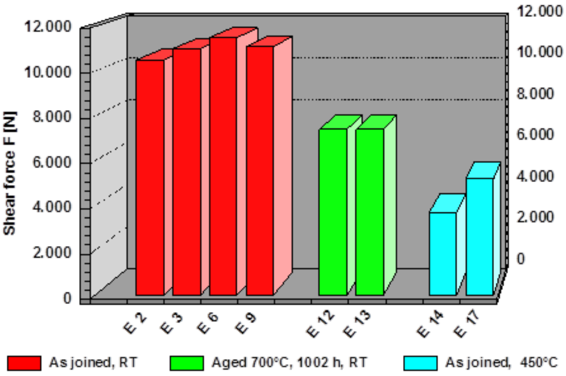

Fig. 6. Shear testing behavior of E – E joints in dependence on testing and treatment conditions.

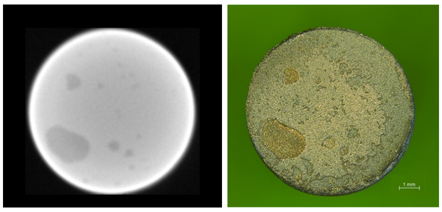

Fig. 7. Brazing zone of Eurofer joint E 12 in state as joined (left) and after aging and shearing-off (right). Left picture is

X-Ray tomography and right one is the optical macro view after testing.

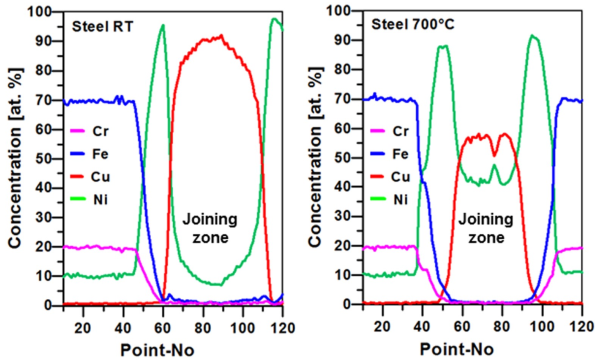

Fig. 8. EDX line scans of steel joints. The concentration profiles belong to parts analyzed in the state as brazed and brazed and annealed.

of base materials. Furthermore these tests showed that the shear strength depends more on application / testing temperature than on alloy combination.

3.4 Applicability of palladium as interlayer

Palladium belongs to the elements which can activate surfaces to achieve better wetting by fillers. In earlier work [9] it was shown that palladium can be electroplated on tungsten. For the shear testing campaign joints were manufactured with an interlayer of approx. 10 μm Pd on Eurofer and tungsten. Eurofer joints were tested at RT and 623 K and loads of about 16 and 12 kN were needed to see shearing, respectively. Tungsten joints failed at a load of roughly 9 kN at 623 K. These tests indicate that shear strength of joints can be increased by applying other interlayers.

4. Conclusions

Blanket and divertor development will need adapted joining technologies to assemble components consisting of different materials, e.g. the armor alloy tungsten to first wall made from Eurofer [1, 2]. The former work under development of He-cooled divertor components showed that wetting of surfaces by fillers or transfer of loads are essential. Such complicity can be reduced or overcome by well-designed and adapted material sequences. Electroplating is a tool which has the capability to deposit layers of different materials and thicknesses on base materials. Thus this tool was selected and further developed to deposit well-adherent and homogeneous layers on materials which are designed for application in fusion technology. It was earlier demonstrated that copper can be used as filler together with nickel interlayer for processing of tungsten – tungsten joints. The actual development showed that deposition of metallic well adherent layers (interlayers Ni or Pd and filler Cu) on tungsten, Eurofer and stainless steel is possible in a reliable manner. Metallurgical analyses indicated that wetting of the components is given together with gap filling during brazing.

The main focus of this work was directed towards the thermomechanical behavior of the processed joints as the second main requirement in the development for future technological application. The tests with Ni as an interlayer showed that reproducible values were obtained as demonstrated for Eurofer joints tested at RT. The performed aging of the joints indicated that no additional defects are formed in the brazing zone which reduces the strength of the joint.

Testing at elevated temperature showed that all combinations tested performed similar and showed strength values reasonable compared to the strength of main filler component at testing temperature. The W – W joints showed that they exhibit some resistance against shearing even at higher operation temperatures.

The tests with an interlayer of palladium showed that this process is not restricted to nickel interlayers, only. It was also demonstrated by the test series with palladium that the strength can be designed upwards for higher application temperatures.

Surely, additional development and qualification work is absolutely necessary in the fields of processing, mechanical tests and brazing solder / filler adjustment. However, the successfully performed tests showed clearly the potential of the new tool.

Acknowledgements

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