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Mechanical Characterization of Electrochemically based W – Cu Joints for Low-Temperature Heat Sink Application

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Joining of the armor material tungsten to other alloys and especially to copper components which will act as heat sinks in divertor application is rather challenging due to the restricted miscibility of tungsten and copper. This difficult behavior leads to bad or missing metallurgical W – Cu reactions with the consequence of reduced mechanical stability. Introducing adapted interlayers can overcome these limitations if they exhibit some extended miscibility with both partners to be joined.

Electrochemical plating was chosen as deposition technology for such reactive interlayers and demonstrators were processed with a 10 µm thick reactive Pd interlayer and joined by diffusion bonding. Their metallurgical behavior was characterized in dependence on processing temperature, reaction time and applied pressing load. The fabricated joints were mechanically qualified by shear testing. Cracking of the joints never appeared at the boundary of interlayer to W. The demonstrators revealed reasonable and applicable shear strength of around 100 MPa. The observed shear strength values and formed microstructures in the joining zone will be displayed and discussed in dependence on the applied processing parameters. The developed bonding process by applying electrochemically plated interlayers has proven to be a reliable tool with industrial application potential.
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Keywords: Divertor, Joining, Electrochemical plating, Shear testing, Heat sink.

1. Introduction

DEMO, which will be a facility somewhere between ITER and a commercial power plant, will need a design which is favoring more conservative technologies [1]. According to that requirement, divertor designs will also follow such a guideline and rely on water cooling instead of advanced He cooling as analyzed under He cooled divertor design [2]. This water cooled design will work with low temperature heat sinks which use W-alloys as armor material in combination with Cu-alloys for fabrication of the cooling channels. Both materials have to be joined in reliable manner which is a critical issue due to the miscibility gap between W and Cu. This limiting behavior under joint processing can be overcome by adding reactive interlayers between the parts to be assembled. Interlayers of Ti, V, Cr, Fe, Pd or Ni offer such an ability of surface activation as it can be deducted from phase diagram analyses [3], which report miscibility or phase formation with tungsten. Such reactive elements can be added to fillers or deposited on surfaces for activation and enabling metallurgical reactions between W and Cu based alloys.

Electrochemical plating was selected as deposition technology for reactive interlayers among other things due to its common utilization in industrial applications. Other advantages also are the ability to coat complex shapes, the good controllability of layer thickness by processing time and plating current. The development of brazing technology by applying electrochemical plating showed that W can be effectively brazed to W or steel by deposition a thin reactive Ni interlayer and a pure copper layer on W. Such braced joints showed good wetting and appropriate shear strength [4] in the range of 200 MPa. Similar values obtained from industrially brazed steel joints by applying conventional fillers. These results demonstrated the general applicability and the benefits of electrochemical plating in brazing technology. However, the processing temperature for W – Cu joints have to be lower to avoid melting or degradation of the low-temperature heat sink Cu-alloy compared to W – W or W - steel brazing by Cu fillers at roughly 1100°C.

Fig.1 shows the general configuration of armor material, interlayer and heat sink to realize such a joint. The objective for this development is the deposition of interlayers by electrochemical plating and joining of the components by diffusion bonding. The processing parameters, e.g., pressing load or time, have to be adjusted to the reactivity of the shown configuration and the mechanical stability of the joint depending on the characteristics of the interlayer. As candidate material acting as reactive interlayer was selected palladium (Pd) due to activation considerations and observed higher shear strength values at brazed W-W joints compared to Ni interlayers [4].

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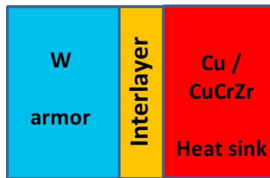

Fig. 1: Schematic view of alloy sequence for joining development by diffusion bonding.

2. Processing of joints

2.1 Development of electrochemical plating

The elements (Ni, Cr, Fe or Pd) which can act as potential interlayers can be deposited due to their chemical behavior in aqueous electrolyte systems. The deposition of a Nickel interlayer was developed earlier for brazing of, e.g., W or steel parts by plating. The surfaces of such materials can be easily coated after a cleaning and activation process by approx. 10 µm thick Ni layers as given in literature [5]. However, Pd plating is more complex since Pd exhibits some tendency to form hydrides during plating due to its affinity to H and the more exposed position in the electrochemical chain of elements. Surely, deposition of Pd can similarly to Ni be performed applying water-based electrolytes.

The development of Pd plating was started with fundamental coating tests. The used electrolyte contained the palladium in form of an ammonia complex and deposition was performed at T = 40°C. In contrast to Ni plating no consumable anode was used. General testing showed that a homogeneous deposition of Pd is possible on electrochemically 'neutral acting' surfaces at current densities near 8 mA/cm² at a pH value of 7 to 8. A variation of the layer thickness was possible in the range 10 – 100 µm by controlling deposition time and the layers were homogeneous and fully covering the surface.

The deposition of Pd interlayers was first intended to be conducted on the W parts in analogy to the successfully performed plating and brazing tests with Ni interlayers. This type of plating was favored due to coating of cleaned W surfaces after etching with an alkaline hexacyanoferrate solution. This processing would avoid adherent intermediate scales (e.g. WO_x) on top of W surfaces between W and interlayer. Fig. 2 shows the surface of the cleaned W part before and after Pd plating. The applied electrolyte was fresh with a pH value of 7.5. However, small deviations in processing conditions can lead to spalling behavior of the deposited layer as shown in Fig. 2c. Negative effects for improper adherence range from, e.g., W quality up to changes of electrolyte composition. The delaminated Pd layers were found to be brittle. The spalling can, but must not happen. This layer behavior is possible in the electrochemical Pd – W system due to, e.g., low hydrogen overvoltages favoring H_2 formation and therefore increased palladium hydride formation. Further tests showed that the occurring overvoltages can be manipulated by mechanical or electrochemical surface modifications of the W part by seeds of Ni or a thin Cu coating of the W surfaces. The valuation is that this doorway can be applied in lab testing [4], but should be avoided at least at the actual stage in industrial processing. Thus, the alternative route was applied of the Pd interlayer deposition on the Cu part for the diffusion bonding development.. Fig. 3 shows the microcut of a coated Cu piece together with the corresponding line scan for the two elements Pd and Cu. The Pd layer is homogeneous and the plating reshaped the profile of the Cu surface accurately. The SEM (Scanning Electron Microscopy) picture and the concentration diagram taken by EDX (Electron Dispersive X-rays) analyses show a similar layer thickness of about 10 µm, which is in agreement with the electrochemically transferred charge.

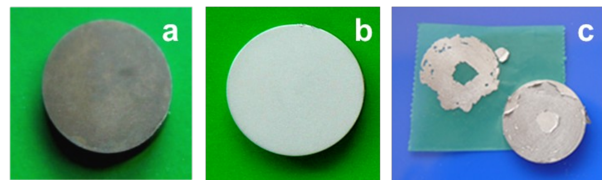

Fig. 2: W surface as activated (a), Pd coated W surface (b) and W part with spalling of Pd layer (c).

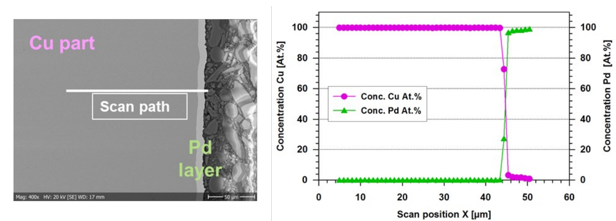

Fig. 3: Pd interlayer deposited on Cu part together with SEM / EDX line scan

2.2 Joining of the parts

For joint fabrication, cylindrical parts were cut from a standard rod material with a diameter of 8 mm with a length of 10 and 30 mm, respectively. The selection of the sample geometries was a contribution to the dimensions required for mechanical testing. Raw materials were tungsten delivered by Plansee Group, Austria and copper of type Cu-ETP DIN 2.0065. The joining of the parts was performed under vacuum (10-4 mbar) in a hot pressing furnace. The arrangement of the parts in the joining unit is depicted in Fig. 4a. The joining of the components was done in the solid state by diffusion bonding.

The bonding behavior was analyzed in respect to processing temperature, pressing load (F) and processing time. The bonding temperatures chosen for the tests were 600 and 700°C, respectively. The highest pressing load was 500 N or 10 MPa with respect to the flow behavior of copper at 700°C. The lowest tested load was 100 N at

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an area of 50 mm² to be joined. In general, any bonding between W and Pd coated Cu was observed at both processing temperatures and loads without deformation of Cu parts. Fig. 4b shows a joint ready for a shear test which was processed at 700°C.

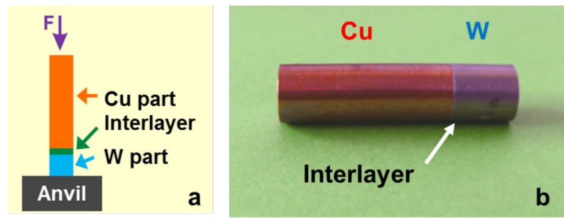

Fig. 4: Schematic view of the sample arrangement in the bonding equipment (a) and bonded specimen (b)

3. Results

3.1 Behavior of W-Cu joints

3.1.1 Microstructure of joined parts

In a first test campaign W – Cu joints were processed at 600 and 700°C. The samples removed from the joining furnace showed by optical inspection correct straight-lined shape and bonding. Fig. 5 shows SEM pictures of the bonding zones of two samples joined at 600 and 700°C, respectively. The bonding load was 500 N or 10 MPa and the dwell time at the designated temperature lasted 1 hour. Both pictures show a significant diffusion reaction of the interlayer Pd with the Cu part, which is identifiable by the grey colored areas. Clearly visible is also the change over from W (bright section) to the interlayer and Cu part (dark section). EDX line scans showed that the deposited interlayer reacted with both joining partners W and Cu.

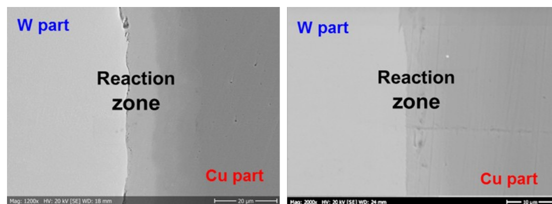

Fig. 5: Diffusion zone of samples bonded at 600 (left) and 700°C (right)

Fig. 6 depicts an element analyses across the reaction zone. The Pd interlayer had an initial thickness of 10 µm and expanded by diffusion into the Cu matrix by roughly 20 µm. The concentration curves for Pd and Cu show that a continuous mixture of both elements took part as expected by phase relationships [3]. The W piece also showed diffusion reaction with the Pd interlayer as can be seen by the curved shape of the W and Pd concentration lines.

The metallurgical analyses confirmed that the desired impact of the interlayer – reaction with both joining partners appeared. These microcuts showed a formation of a reaction zone without defects.. The analyses pointed out that the degree of the (at least) W-Pd reaction depends on the processing parameters, e.g., 600 or 700°C bonding temperature.

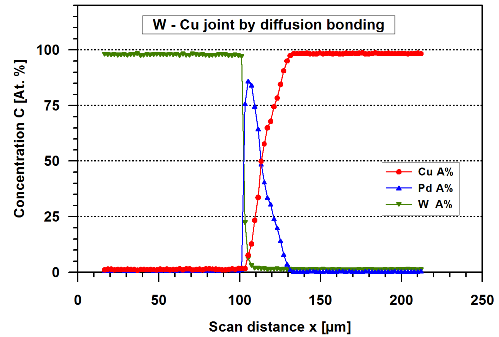

Fig. 6: Concentration profile for the 3 elements W, Pd and Cu of a diffusion bonded sample at 700°C for 1 hour with a load of 350 N

3.1.2 Mechanical behavior of joints

The mechanical behavior of W – Cu joints was analyzed by shear testing of the diffusion bonded demonstrators. The tests were performed at RT in an universal mechanical testing machine "Instron 4505" which is designed for operation in the range of RT to 700°C [4]. The shear tests were performed with a displacement rate of 0.01 mm/s and the load was recorded. The test was stopped if cracking or too large bending of the samples occurred.

3.1.3 Cracking behavior

The visual nature of the cracked surfaces after shear testing showed a clear dependency on bonding time and temperature. Fig. 7 and Fig. 8 show a representative sequence of pictures collected from samples processed at 700° C and pressing time of 1 and 4 hours, respectively. The pressing load was 7 MPa. The sliding appeared in the contact zone of W to Pd – Cupart for the samples pressed for 1 hour. Fig. 7 gives an impression of the bonding progression. The turning structure of the surfaces is still present and indicates that a point or line contact took place at the beginning of the diffusion bonding process. The cone ends of the tool marks of the W part had contact to the Pd layer. From there the diffusion started. The indication is the detected W amount on top of the Pd coated Cu part. The bright areas of the SEM picture (Fig. 7) visualized an increased W fraction. EDX analyses showed quantities up to 50 %. This may indicate that a cracking of the cone ends of W appeared during shear testing. The same configuration bonded for 4 hours showed a different structure. In this case, the cracking took place in the volume of the bonding zone. The bonding of W to the interlayer Pd was steady.

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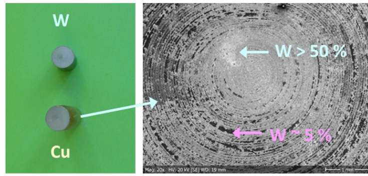

Fig. 7: Surfaces of shear tested fragments. The sample was bonded at 700°C and bonding time was 1 hour

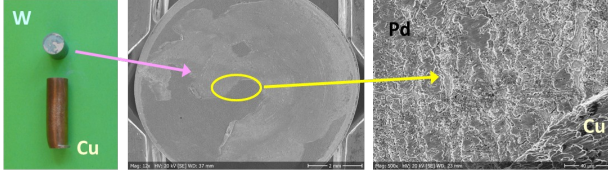

Fig. 8: Surfaces of shear tested fragments. The sample was bonded at 700°C and bonding time was 4 hours. Centre: SEM macro view of W part. Left: Structure of sheared area. The diameter of the sample is 8 mm.

The sliding mainly appeared in the Pd – Cu solid solution zone. The macro view (Fig. 8) even shows some area with red Cu color sticking on top of the W part. Further analyses showed that no brittle failure of the joint was present. The deformation of the Cu part suggests that the shear strength was above of the copper's strength. A similar appearance of 'cracking patterns was observed at bonding temperature 600°C. However, samples bonded for 1 hour showed extreme point contacts. Such samples failed at rather low shear loading force (5 MPa range). A behavior as shown in Fig. 8 was observed after 8 hours at 600°C.

3.1.4 Shear strength

The shear testing was performed at RT and the the tested area was 50 mm². The processing parameters of the samples for shear testing are given in Tab. 1. Applying too short bonding times and too small bonding loads produced samples, which failed rather early and showed very low shear strength as listed in Tab. 2 (sample D 10 and D 02). The structure of the cracked surfaces were similar to Fig. 7 and indicated that bonding appeared only at same points due to the roughness of the surfaces. Diffusion and flow of material under load were not sufficient for obtaining an overall contact of the surfaces – the general condition for bonding. This issue was overcome by increasing the bonding load – compare samples D 02 and D 12 or by enlarging the bonding time (sample D 10 vs. D 15). Tab. 2 illustrates that the degree of bonding depends on all three listed parameters. Similar shear strength was obtained at bonding temperature 600°C by roughly 10 times longer processing time than at 700°C. Fig. 9 shows the shear strength behavior in dependence on the bonding time for joints processed at 700°C and a pressing load of 350 N. The diagram illustrates that the shear strength increased with bonding time.

Table 1: Processing parameters of samples

Temperature [°C]	Time [h]	Force [N]
600	1	350
	1	500
700	0.5	100
700	0.5	350
	1	350
	4	350

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Table 2: General behavior of diffusion bonded samples

Sample	Time [h]	Shear strength [MPa]	Bonding force [N]	Temp. [°C]
D 10	1	1	500 N	600°C
D 15	4	40	350 N	600°C
D 02	0.5	1	100 N	700°C
D 12	0.5	50	350 N	700°C

The thickness of the W – Pd zone enlarged and the W distribution homogenized in the course of time. Surely, the Pd – Cu mixing also progressed. The shear strength reached values of roughly 120 MPa at bonding times of 4 hours. Such a value is comparable with strength data (around 200 MPa) observed at applying conventional steel brazing with Cu – based fillers [6]. This indicates that the developed joining technology is applicable under industrial demand.

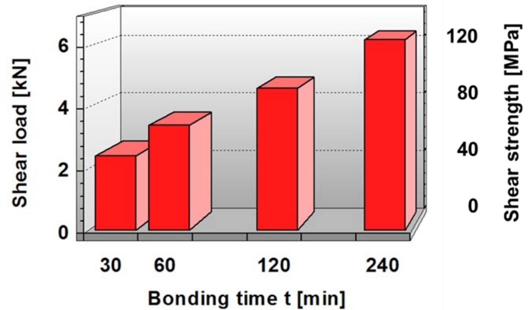

Fig. 9: Shear strength in dependence on pressing time for bonding temperature 700°C

3.2 Behavior of W-CuCrZr joints

In a second test campaign W – CuCrZr joints were processed at 600 and 700°C. The processing was the same as used for fabricating the W – Cu joints. A pressing load of 500 N was chosen for the sample joined at 600°C. The bonding at 700°C was executed at a load of 350 N to avoid deformation of the CuCrZr cylinder.

3.2.1 Microstructural analyses of the joints

Bonded samples could be fabricated at both bonding temperatures. EDX line scans performed at cross-cuts of bonded samples revealed that diffusion took place of W and Pd in a ratio known from the W – Cu joints. The SEM picture (Fig. 10) shows that a tight reaction between W and Pd occurred without forming defects. A mixing of Pd and CuCrZr by diffusion was also

observed. The Pd mapping image indicated a mixing zone of about 20 µm. The element analyses confirmed the presence of both reaction types. The concentration profiles are similar to the dependency shown in Fig. 6.

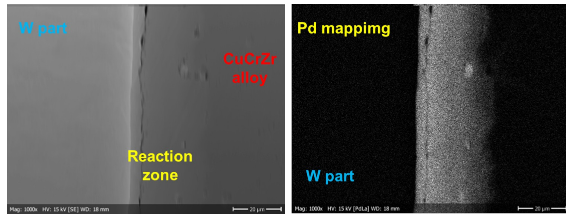

Fig. 10: Micro-cut (SEM picture, left) and Cu mapping (right) of a sample bonded at 700°C for 4 h.

Table 3: Shear strength of W – CuCrZr joints by Pd interlayer

Sample	Time	Shear strength	Temp.
D 1a	8 h	20 MPa	600°C
D 2a	2 h	1 MPa	700°C
D 3a	4 h	30 MPa	700°C
D 5a	8 h	90 MPa	700°C

3.2.2 Shear strength of the joints

The shear testing of the W-CuCrZr joints was done with the same setup used for W-Cu testing at RT. A summary of representative values measured is given in Tab. 3 in dependence on pressing time and temperature. The easiest extractable information will be that required pressing times were clearly longer for obtaining resistant joints compared to W bonding onto pure Cu. This is valid for both tested temperatures. The increase of shear strength with pressing time was also observed at testing of W-Cu joints.

3.2.3 Cracking of W-CuCrZr joints

All the processed joints showed the appearance of an intense reaction of W and Pd and that Pd always covered the W part after shear testing. EDX point analyses showed a cover layer containing amounts of Cu. From this it can be concluded that also a Pd reaction with the CuCrZr took place. The cracking patterns as illustrated in Fig. 11 reveal two different zones. The first one is copper-colored and the other one is silvery shining.

Fig. 11: View on crack position after shear testing

This view indicates the outcoming of cracks from the Pd-CuCrZr contact zone towards the alloyed Cu rich ‘matrix’. More detailed analyses of microcuts give hints that defects or microcracks may be present even after cooling down of the joined sample. It may be assumed that the higher strength of the CuCrZr and the large expansion mismatch of W and Cu caused this feature.

4 Conclusion

The performed development showed that electrochemical deposition of reactive interlayers (e.g., Ni or Pd) is possible on both components W and Cu, which are designed as materials for divertors with low temperature heat sink. Well adherent interlayers were processed with constant and controllable thickness.

Diffusion bonding of W and Cu parts with a 10 µm thick Pd interlayer was successfully processed. The impact of the processing parameters bonding temperature, bonding pressure and bonding time on metallurgical and mechanical behavior was analyzed. Stable joints could be fabricated with both applied bonding temperatures of 600 and 700°C in rod shape of 8 mm, which was directly applicable for shear testing.

The bonding tests showed that at least roughly 5 MPa pressing load should be used to have sufficient contact of the parts during diffusion bonding and to overcome point contacts caused by surface roughness. The metallurgical analyses showed that the diffusion process at the W – Pd boundary is slower than the Pd – Cu reaction. SEM analyses indicated that the W – Pd reaction zone should have a thickness of near 5 µm to guarantee well stable joints. Such a configuration was obtained for temperature / time combinations of 600°C / 4 h or 700°C 0.5 h or at longer duration.

Shear testing of demonstrators fabricated at 700°C showed that the shear strength increases significantly with processing time. Samples bonded with dwell times of 4 hours revealed strength values of 120 MPa and a homogeneous bonding zone without any defects was found.. A comparison with classical steel brazing implies that the processed joints have sufficient strength. The tests performed with W-CuCrZr demonstrator revealed similar results. However, they showed that longer reaction times were necessary. Beyond that, new challenges appeared with CuCrZr. The strong expansion mismatch created defects in the reaction area Pd to CuCrZr during the cooling down step. As counter action additional soft Cu interlayers between Pd and CuCrZr for stress reduction seem to be suggested.

The whole test campaign proved that the developed joining process is reliable without any strange parameter limits. However, a further qualification of this process is recommended with respect to the behavior of the joint under high heat flux conditions.

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References

[1] G. Federici, R. Kemp, D. Ward, C. Bachmann, T. Franke, S. Gonzalez et al., Overview of EU DEMO design and R&D activitiesG., Fusion Engineering and Design, 89, (2014), 882–889.

[2] P. Norajitra, R. Giniyatulin, T. Hirai, W. Krauss, V. Kuznetsov, I. Mazul et al., Current status of He-cooled divertor development for DEMO, Fus. Eng. Design, 84, (2009), 1429-1433.

[3] ASM Intern., Materials Park, OH, USA, 2006-2013 http://www1.asminternational.org/AsmEnterprise/APD/

[4] W. Krauss, J. Lorenz, J. Konys, W. Basuki, J. Aktaa, Thermomechanical characterization of joints for blanket and divertor application processed by electrochemical plating, Fus. Eng. Design, In Press, Corrected Proof, Available online 29 December 2015, doi:10.1016/j.fusengdes.2015.12.043

[5] W. Krauss, J. Lorenz, J. Konys, Joining of HHF materials applying electroplating technology, Fus. Eng. and Design, 89, (2014), 1213-1218.

[6] Sebastian Buhl, Christian Leinenbach, Ralph Spolenak, Konrad Wegener, Microstructure, residual stresses and shear strength of diamond–steel-joints brazed with a Cu–Sn-based active filler alloy, Int. Journal of Refractory Metals and Hard Materials, 30, (2012), 16–24.

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