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Journal of Nuclear Materials 283–287 (2000) 1253–1257

journal of nuclear materials

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Refractory metal joining for first wall applications

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Abstract

The potential use of high temperature coolant (e.g. 900°C He) in first wall structures would preclude the applicability of copper alloy heat sink materials and refractory metals would be potential replacements. Brazing trials were conducted in order to examine techniques to join tungsten armor to high tungsten (90–95 wt%) or molybdenum TZM heat sink materials. Palladium-, nickel- and zirconium-based filler metals were investigated using brazing temperatures ranging from 1000°C to 1275°C. Palladium–nickel and palladium–cobalt braze alloys were successful in producing generally sound metallurgical joints in tungsten alloy/tungsten couples, although there was an observed tendency for the pure tungsten armor material to exhibit grain boundary cracking after bonding. The zirconium- and nickel-based filler metals produced defect-containing joints, specifically cracking and porosity, respectively. The palladium–nickel braze alloy produced sound joints in the Mo TZM/tungsten couple. Substitution of a lanthanum oxide-containing, fine-grained tungsten material (for the pure tungsten) eliminated the observed tungsten grain boundary cracking.

1. Introduction

A considerable effort has been undertaken in recent years to develop joining technology necessary to fabricate first wall structures which employ tungsten, carbon or beryllium armor materials bonded to a copper alloy heat sink [1,2]. However, the desire for improved operating efficiencies in fusion reactors may necessitate the use of heat sink materials possessing higher temperature capabilities. Specifically, several design scenarios utilize helium-cooled structures with nominal coolant temperatures of approximately 900–1000°C.

Refractory alloys exhibit excellent high temperature strength while possessing reasonably good thermal conductivity. The substitution of these materials for copper alloys in heat sink applications provides an additional benefit of minimizing bondline stresses between the actively cooled structure and tungsten armor due to the more favorable match of thermal expansion coefficients.

The high thermal stresses developed during operation of first wall components combined with the limited tensile ductility of refractory metals in general, and weld metal in particular, make fusion welding of these components a poor choice. Diffusion bonding and brazing processes can produce microstructures which are less prone to defects and more tolerant of imposed stresses generated either during fabrication or operation.

2. Experimental procedure

In the current investigation, several potential brazing processes for joining tungsten armor to either tungsten or molybdenum alloy substrates were examined. Two tungsten alloys and a molybdenum alloy were selected as heat sink materials; their compositions are listed in Table 1. The two tungsten alloys are ‘machinable’ grades that consist of a matrix of predominantly copper and nickel in a solid solution surrounding discrete tungsten particles. These alloys have superior fracture toughness (compared to pure tungsten) while retaining some modest strength level (~95 MPa tensile strength at 1000°C). TZM molybdenum was chosen for its superior thermal stability; it provides at least a 250°C advantage in recrystallization temperature compared to pure

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PII: S0022-3115(00)00178-1

Table 1
Compostion of test materials (wt%)

90W–6Ni–4Cu
95W–3.5Ni–1.5Cu
99.4Mo–0.5Ti–0.08Zr
Pd–40Ni ($T_s = T_l = 1238°C$)
Pd–35Co ($T_s = T_l = 1219°C$)
Zr–17Ni ($T_s = 982°C, T_l = 986°C$)
Ni–15Cr–3.5B–0.5Ti ($T_s = T_l = 1055°C$)

molybdenum (1450°C vs. 1200°C) [3]. Heat sink materials were obtained in bar form, approximately 4 cm in diameter.

Braze alloy compositions were selected based on the desire to obtain a bondline capable of sustained operating temperatures in excess of 1100–1200°C. Two palladium-based filler metals were selected for the Mo TZM to tungsten material combination. For the tungsten alloy/tungsten joining experiments, two additional filler metals were chosen. The Ni–Cr–B eutectic composition is commonly used to join nickel-based alloys (similar to the W heat sink matrix composition). A small Ti addition was added to this filler composition to improve wetting of the tungsten particles. Additionally, a Zr–Ni filler metal was examined. All filler metals consisted of 50–75 μm thick foil; their compositions and solidus–liquidus temperatures are listed in Table 1. The latter two filler metals, while having lower melting temperatures, were chosen based upon their capability to react with base metals, leading to high remelt temperatures of the joint.

Two tungsten armor compositions were examined. A baseline of commercially pure composition in the form of 1.5 mm thick sheet was compared with 3 mm thick sheet containing 1% lanthanum oxide. Both materials were procured from Plansee.

Small coupons of the heat sink materials measuring 2 cm × 2 cm × 3 mm were machined for bonding trials. Surface preparation consisted of mechanical polishing with 600 grit SiC abrasive followed by solvent cleaning in acetone. The tungsten 'armor' coupons, 1.5 cm × 1.5 cm thick, were polished and cleaned similarly.

Braze foil, cut to the dimensions of the heat sink material, was preplaced between the two coupons. A small mass, approximately 200 g, was placed on top of the assembly to provide a slight bonding load. The assembly was then vacuum brazed at a maximum pressure of 1×10^{-5} Torr. The thermal profile of all specimens consisted of a 20°C/min heating rate and a 300 s hold, at the peak temperature, followed by a free cool (in vacuum) to ambient temperature.

The material combinations examined are listed in Table 2.

3. Experimental results

The palladium-based filler metals produced sound joints with all material combinations. A typical braze microstructure is shown in Fig. 1. The braze filler metal retains its single phase, solid solution structure. Note that there are several irregularities in the tungsten–braze interface. It appears that several small grains of tungsten are missing. It is uncertain whether these features are a result of the mechanical polishing (during specimen preparation) or formed during the brazing step. Higher magnification (Fig. 2) reveals grain boundary cracking of the tungsten. This grain boundary cracking was found in all specimens that employed pure tungsten.

Some diffusion between filler metal and the tungsten alloy heat sink material were observed in all specimens. Depending on the local density of tungsten particles and matrix, diffusion of filler into the matrix material occurred to a depth of 5–15 μm away from the interface. Considerably less diffusion was observed into the tungsten particles, typically about 2 μm. It is expected that longer hold times at the brazing temperature would produce amplified diffusion distances leading to useful elevation of the remelt temperature of the braze joints.

Table 2
Materials and bonding temperatures used in brazing experiments

Heat sink material	Armor material	Braze alloy	Braze temperature (°C)
90% W	W	Pd–Co	1275
90% W	W	Zr–Ni	1000
90% W	W	Ni–Cr–B–Ti	1200
95% W	W	Pd–Co	1275
95% W	W	Pd–Ni	1275
95% W	W	Zr–Ni	1000
95% W	W	Ni–Cr–B–Ti	1200
95% W	W-La	Pd–Co	1275
Mo TZM	W	Pd–Co	1275
Mo TZM	W	Pd–Ni	1275
Mo TZM	W-La	Pd–Co	1275

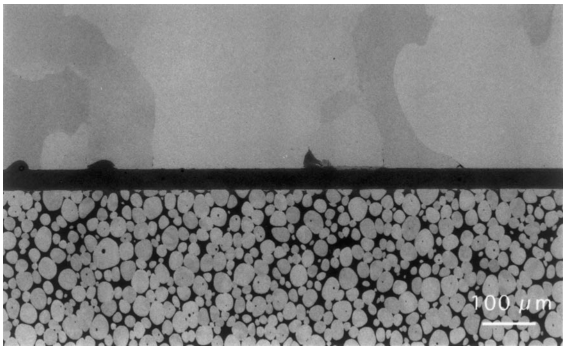

Fig. 1. Cross-section of 90% tungsten (bottom) brazed to pure tungsten (top) at 1275°C using Pd–35Co.

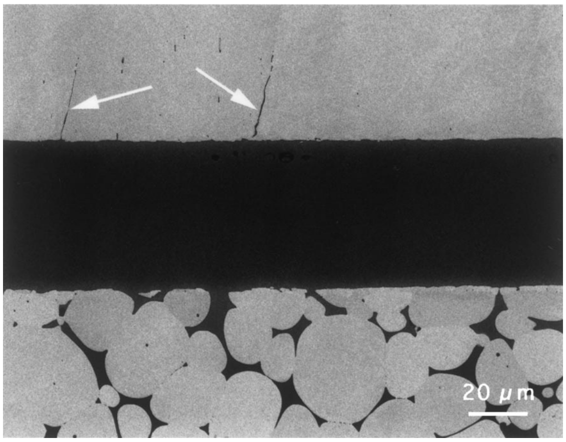

Fig. 2. Cross-section of 95% tungsten (bottom) brazed to pure tungsten at 1275°C using Pd–35Co. Grain boundary cracking of tungsten indicated by arrows.

The Mo TZM/tungsten specimens fabricated using the Pd–Ni filler produced joints free from second phase particles with little diffusion. However, the Pd–Co filler reacted with the Mo substrate to produce a continuous layer of intermetallic structure (Fig. 3).

Although brazed at a lower temperature, the Zr–Ni filler metal exhibited greater reactivity with tungsten and tungsten alloy substrates. Unfortunately, fillets formed in these joints were extremely brittle with considerable cracking. Cracks originating in the fillet area (Fig. 4) usually propagated through or across the braze joint into the pure tungsten; cracking of the tungsten heat sink material less common.

Finally, the nickel-based filler composition reacted strongly with both the pure tungsten and the tungsten particles in the heat sink materials. A reaction layer containing chromium, nickel and tungsten was found at all tungsten/braze interfaces (Fig. 5). Additionally, joints made this nickel-based filler metal contained small amounts (less than 5%) of porosity.

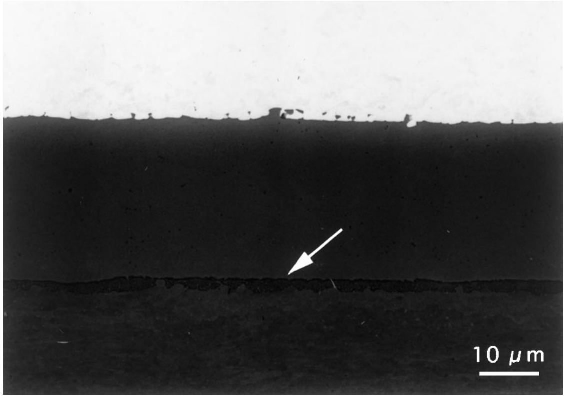

Fig. 3. Cross-section of Mo TZM (bottom) brazed to pure tungsten at 1275°C using Pd–35Co. Intermetallic Pd–Mo–Co structure indicated by arrow.

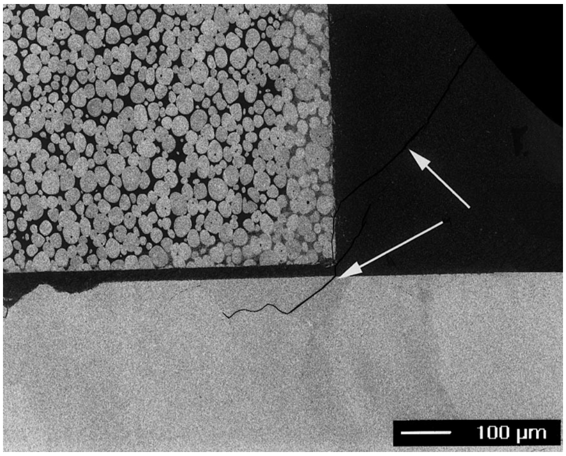

Fig. 4. Cross-section of 90% tungsten (top) brazed to pure tungsten 1000°C using Zr–17Ni. Arrows indicate crack formed in fillet which propagated into tungsten armor.

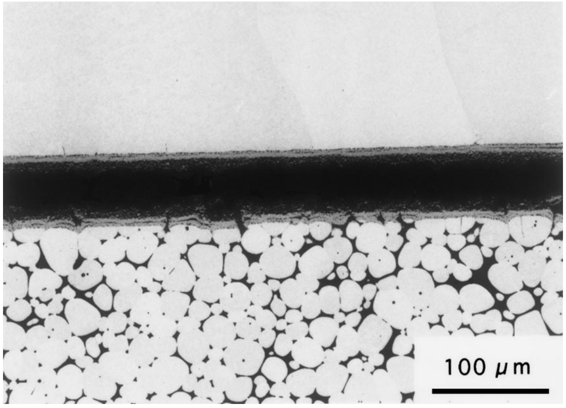

Fig. 5. Cross-section of 95% tungsten (bottom) brazed to pure tungsten at 1200°C using Ni–15Cr–3.5B–0.5Ti.

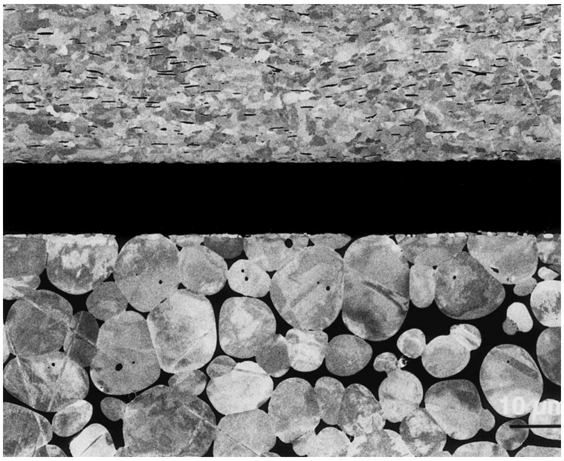

Fig. 6. Cross-section of 95% tungsten (bottom) brazed to lanthanum oxide-containing tungsten at 1275°C using Pd–35Co.

Substitution of the fine grained, oxide dispersion containing tungsten for pure tungsten did not appear to significantly change the metallurgical structure of braze joints. However, the lanthanum oxide-containing tungsten did prove to be more crack resistant, as evidenced by the lack of grain boundary failure in comparable joints (Fig. 6).

4. Discussion

The high tungsten alloys offer significant advantages in machining and fracture toughness compared to pure tungsten. However, there is a corresponding penalty in the material's thermal conductivity. The 90% tungsten alloy has a thermal conductivity value approximately 50% of pure tungsten (0.22 vs. 0.48 cal/cm °C s); the 95% tungsten alloy is somewhat better at 0.32 cal/cm °C s [4]. Since both of these tungsten alloys appeared to braze similarly, the higher tungsten content material would be preferred for its improved cooling capability.

In order to minimize bondline stresses between the tungsten armor and the tungsten alloy heat sink material, it is desirable to match thermal expansion coefficients as closely as possible. The 95% tungsten alloy has an expansion coefficient of 4.9×10^{-6}°C compared to a value of 5.4×10^{-6}°C for the 90% tungsten material [4]. Since pure tungsten expands art a rate of 4.4×10^{-6}°C, the higher tungsten content alloy will also be preferred to reduce the likelihood of cracking the tungsten armor.

Similarly, both palladium-based braze alloys produced good joints in the tungsten alloy/tungsten combinations. Since the Pd–40Ni composition has a 20% higher thermal conductivity [5] than the Pd–35Co alloy, the nickel-bearing filler metal would offer a slight advantage from a thermal standpoint. (Note that both filler metals have less than 25% of the thermal conductivity of pure tungsten.) The Pd–Ni composition would also be preferred in Mo TZM/tungsten joints due to the absence of second phase structures that were found in joints made with Pd–Co filler.

The palladium-based filler metals exhibited excellent wetting on tungsten and tungsten alloys. This, combined with the observed homogeneity and apparent ductility of the braze microstructures, suggests that these filler metals would be excellent candidates for improved brazing procedures which might utilize a reduced braze filler thickness and an increased bonding pressure. Such an approach would have several benefits. A reduction in braze joint thickness would permit increased dilution of the braze alloy by both tungsten and elements from the heat sink matrix material, thereby increasing the joint's temperature capability. Note that the solubility of tungsten in palladium exceeds 20 at.% [6].

Design of some plasma facing components has incorporated armor consisting of a brush structure bonded to a heat sink [7]. Whether the brush structure consists of close-packed rods or closely aligned rectangular rods, capillary forces will act to draw molten brazing filler away ('wicking') from the bondline causing potential defects, particularly porosity. By combining a minimum quantity of preplaced filler metal with higher bonding pressures, acceptable quality joints could be made while avoiding the wicking concern.

5. Conclusions

Brazing techniques can be viable joining processes for bonding tungsten armor to tungsten- or molybdenum-based heat sink materials. Solid solution filler metals based on the palladium–nickel or palladium–cobalt binary systems can produce joints capable of service at temperatures up to 1100–1200°C. Zr–17Ni and Ni–15Cr–3.5B–0.5Ti braze alloys offer the potential of enhanced diffusion accompanied by increased joint remelt temperature, but with limitations stemming from lack of ductility and formation of second phase microstructures.

The use of lanthanum oxide-containing tungsten for armor applications appears, from a fabrication standpoint, to be superior to pure tungsten due to the decreased grain size and improved crack resistance of the oxide-bearing material.

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