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A Review of the US Joining Technologies for Plasma Facing Components in the ITER Fusion Reactor

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A REVIEW OF THE US JOINING TECHNOLOGIES FOR PLASMA FACING COMPONENTS IN THE ITER FUSION REACTOR

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ABSTRACT

This paper is a review of the current joining technologies for plasma facing components in the US for the International Thermonuclear Experimental Reactor (ITER) project. Many facilities are involved in this project. All of those facilities are not represented in the authors list but all contributions will be noted throughout the report and in the acknowledgements. Many unique and innovative joining techniques are being considered in the quest to join two candidate armor plate materials (beryllium and tungsten) to a copper base alloy heat sink (CuNiBe, OD copper, CuCrZr). These techniques include brazing and diffusion bonding, compliant layers at the bond interface, and the use of diffusion barrier coatings and diffusion enhancing coatings at the bond interfaces. The development and status of these joining techniques will be detailed in this report. Ultimately, these assemblies will be considered for possible use in selected regions of a full-scale, tokamak-designed fusion reactor.

Because beryllium reacts with all but a few elements to form intermetallics and is a strong oxide former, this study considered several different surface coatings as a means of both inhibiting these reactions and promoting a good diffusion bond between the two substrates. All bonded assemblies used aluminum and/or an aluminum-beryllium composite (AlBeMet 150) as the interfacial material in contact with beryllium. In all cases, a hot iso-static pressing (HIP) furnace was used to reduce oxidation and apply sufficient pressure to the bond area to produce metal-metal contact and subsequent bonding. Several different processing schedules were evaluated during the course of this study. Tensile testing followed by post-mortem examination of the fractures and cross-sections showed that several of the trial assemblies produced excellent strength (100-200 MPa) and ductility. Two of these assemblies including one brazed assembly and one diffusion bonded assembly were tested in the electron beam test system (EBTS facility) at SNL-NM. These actively-cooled mock-ups were subjected to heat loads of up to 2000 cycles at 5 MW/m² and 1000 cycles at 10 MW/m² without bond failure or damage to any tiles. Several tiles were subjected to short term heat loads as high as 250 MJ/m² (0.5s) to simulate vertical disruption events which caused surface melting of the beryllium tiles.

Several different techniques and geometries are being evaluated in joining a tungsten armor rod bundle (tungsten brush) to a CuCrZr heat sink. Methods being used to coat the tungsten rod bundles with copper include plasma spraying, electroplating, and ion sputtering. Also, in collaboration with the Efremov Institute in Saint Petersburg, Russia, research is underway to coat tungsten rod bundles using a copper casting technique. Following the coating process, these bundles are bonded to the copper substrate using a low temperature diffusion bonding technique which employs a HIP process at 400-500°C. Thin coatings (100-200 nm) of nickel are used at the bond line to enhance diffusion and produce excellent bond strengths between the heat sink alloys (CuCrZr, CuNiBe). Bonds have been produced in these combinations with fracture strengths greater than 310 MPa at room temperature and greater than 175 MPa at 300°C. At this time, the tungsten coating studies are in progress. Mechanical test results and microscopic examination of selected samples will be reported. The best of these coating and bonding techniques will be tested in the EBTS facility at heat loads commensurate with the divertor requirements and reported in this paper.

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A REVIEW OF THE US JOINING TECHNOLOGIES FOR PLASMA FACING COMPONENTS IN THE ITER FUSION REACTOR

1. Introduction

Development of joining technologies for the plasma facing components on today's fusion reactors has generated many unique challenges. One of the main factors contributing to these challenges is the combination of materials and processes used. This problem is particularly severe for carbon brazed to copper alloys. These materials differ greatly in their thermal expansion and are typically brazed with reactive metal (e.g., Ti-Cu-Ag) brazes at 800-850°C [1]. Cooling from the braze cycle produces high residual stresses that can promote subsequent cracking during service. Surviving the braze cycle itself is the first "operational" difficulty for such components. The problem of residual fabrication stresses was one factor that led the US ITER team to explore joining methods that could be

used at lower temperatures than those for reactive metal brazes. Other techniques for mitigating the residual stresses from fabrication and accommodating the thermal stresses during service include (a) castellation (segmenting) of the armor and (b) compliant interlayers that undergo some plastic strain during fabrication and thereby reduce residual stresses in the armor and substrate.

Beryllium and tungsten are the two PFC armor candidates being studied in the US joining effort; beryllium-armor for the primary first wall, baffle, limiter, and dome sections of the reactor and tungsten-armor for the divertor. The design options for the PFC's utilize a duplex structure whereby the armor plate is bonded to a water-cooled heat sink. The heat sink candidates include three copper alloys including: Hycon-3 (CuNiBe), Elbrador (CuCrZr) and Glidcop (DS copper), with CuCrZr being the leading candidate at this writing. These bonds must have good physical properties after irradiation, good mechanical properties, and be able to withstand cyclic heat loads without degradation. Both brazing and diffusion bonding are being considered as prime candidates for the joining technology. Two different coating techniques are being considered; plasma spraying and ion sputtering. Innovative coatings are being utilized as diffusion barriers and diffusion enhancers at the bonding surfaces. The following document will serve as a review of these joining technologies as they currently stand in the United States.

2. Brazing of beryllium to copper

The single most important characteristic of beryllium from a joining standpoint is its high reactivity. Beryllium forms stable compounds with almost every element in the Periodic Table. These compounds are typically both strong and brittle; their presence in a bond joint can reduce mechanical performance to unacceptably low levels of ductility and toughness. Also, beryllium readily oxidizes in extremely low vacuum pressures of 10^{-6} torr and less. Therefore, in order to form sound metallurgical bonds, a beryllium bonding surface must first be cleaned to remove surface oxides and be kept clean throughout all handling and processing steps until the bonding process is complete.

Several elements are compatible with beryllium, that is, they do not form intermetallic compounds and would thus provide a good base for filler metals or a compliant layer in contact with beryllium. Among these are silver, germanium, silicon, and aluminum. Silver has been successfully used by a number of investigators to join beryllium to copper [2,3,4,5] by both brazing and diffusion

bonding. However, concerns about activation and transmutation products in the ITER neutron flux environment have led to the decision that silver is unacceptable for plasma facing components [6]. Aluminum has been the focus for the silverless-braze filler metal candidate as well as a compliant layer between the armor and heat sink. Aluminum has reasonable thermal conductivity (about 2.4 W/m·°K at room temperature vs. 2.9 W/m·°K for the copper heat sink materials), good ductility and should act as a compliant layer to absorb most of the thermal stresses generated by the thermal expansion difference between beryllium and copper. The maximum residual stresses would be constrained to that of approximately the room temperature yield strength of the compliant layer material. 1100-Al alloy has a room temperature yield strength of 35 MPa. Another compliant material considered in these studies is AlBeMet-150 (50w/oBe-50w/oAl) which has a room temperature yield strength of 250 MPa.

Copper and beryllium form a number of intermetallic compounds which are stable to temperatures in excess of 900°C [7]. Direct bonding of beryllium to copper at temperatures as low as 350°C results in measurable thicknesses ($> 0.1 \mu\text{m}$) of the compounds BeCu and Be₂Cu after exposure times of 1 hour [7,8]. Two approaches have been used to circumvent this problem which include diffusion barriers and short temperature exposures. The incorporation of diffusion barriers [9] can isolate the copper from the beryllium, allowing higher bonding temperatures to be used. Other researchers [10] have used brazing cycles which employ rapid heating and cooling rates to limit the time of exposure at elevated temperatures.

While aluminum and beryllium are indeed compatible, reaction between aluminum and copper to form brittle intermetallic compounds precludes the direct brazing of beryllium to copper alloys using aluminum filler alloys. Instead, the approach taken was to utilize a diffusion barrier which would prevent any reaction between aluminum and copper. The diffusion barrier material chosen was a thin layer of titanium bonded as an interlayer between the aluminum and copper by using either an explosive bonding technique [11] (developed by Northwest Technical Ind, Sequim, WA) or ion sputtering the titanium and aluminum coatings [12] (Surmet Corp., Burlington, MA). Although titanium reacts with aluminum to form intermetallics, the bonding temperatures for this joining process are low and the reaction is kept to a minimum. By employing aluminum coatings on the beryllium armor, the task of bonding beryllium to the copper heat sink alloy was reduced to bonding aluminum to itself. In several trials, AlBeMet-150 (Brush-Wellman) was substituted for the aluminum compliant layer as a higher strength option. AlBeMet-150, consisting of 50w/o beryllium in an aluminum matrix, has improved strength (250 MPa) over that of the 1100-Al (35 MPa).

Two methods were employed to coat aluminum onto the beryllium surface; plasma spraying (PS) and ion sputtering (PVD). In all cases, the heat sink material is aluminum clad (by explosive bonding) CuCrZr with a titanium diffusion barrier as described above. The actual bonding sequence was accomplished either in a vacuum furnace or a hot isostatic press (HIP). Fabrication details of the final five brazing assemblies, bonding parameters, and fracture strengths are shown in Table 1. A schematic representation of a bond specimen is shown in Figure 1.

After processing, the candidate assemblies were removed for metallographic examination and mechanical testing. The characteristics of the bond interface were examined by both optical and electron microscopy. The fracture strength was determined using a transverse tensile test specimen geometry with a reduced cross-section.

The processing used to produce Specimen C-B was selected as the best of the brazing schedule candidates and was thus designated as the process to be used to produce a specimen for high heat flux testing. Accordingly, a specimen was fabricated for testing in the Electron Beam Test System (EBTS) at Sandia National Laboratory - New Mexico. This sample survived the 5 and 10 MW/m² exposures without incident. All beryllium tiles were intact and indicated no change in the surface

temperature. Following these tests, several tiles on the sample were subjected to heat loads in excess of 250 MJ/m² (0.5s) before some localized melting of the beryllium occurred.

Table 1
Brazed assemblies

Specimen ID	Al Coating on Be	Filler Metal	Bonding Parameters	Fracture Strength (MPa)
A-B	0.025 mm PVD	0.25 mm Al-12%Si	660°C/3 m/0.07 MPa	Broke while machining
B-B	0.30 mm PS	0.25 mm Al-12%Si	660°C/3 m/0.07 MPa	41.3 30.0
C-B	0.30 mm PS	0.25 mm Al-12%Si	625°C/15 m/103 MPa	115.4 118.2
D-B	0.30 mm PS	0.010 mm EP Cu	625°C/60 m/103 MPa	114.3 117.3
E-B	0.025 mm PVD	0.001 mm PVD Si	625°C/60 m/103 MPa	83.3 121.6

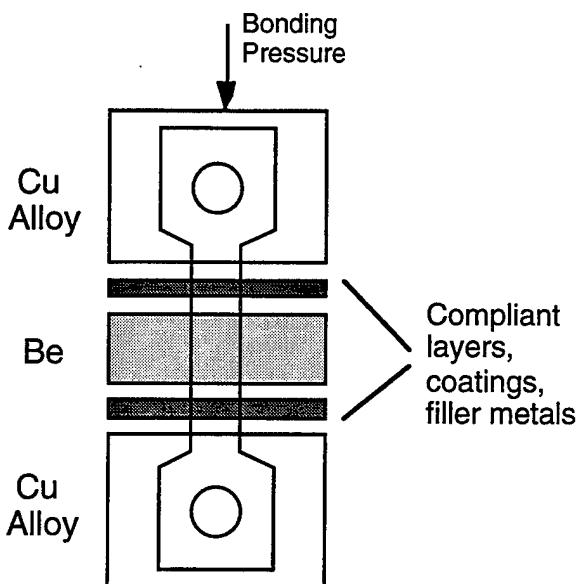


Figure 1
Bond assembly configuration showing orientation of specimen for transverse tensile testing.

3. Diffusion bonding of beryllium to copper

Diffusion bonding is defined as the coalescence of two metals through atomic transport and mechanical deformation at the bonding interface. For diffusion bonding to occur, the bare metal surfaces must come in contact at a distance close to that of the crystal lattice constant. When this condition has been met, temperature and pressure accomplish the bond. Without the need for filler metals, diffusion bonded joints can achieve weight reductions and better heat transfer properties. Additionally, for many nuclear applications, high atomic number metals used as filler materials are undesirable. Tenacious oxide films are hindrances to the bonding process for reactive metals such as beryllium and aluminum. The metal oxide films must be broken or eliminated in order to get

intimate contact and subsequent bonding between the metal surfaces [13,14].. Much of the effort in developing a diffusion bonding process is involved in minimizing the effects of this oxide layer [15].

The application of pressure during the bonding process can contribute in two ways. The first is the breakup of the oxide film by a shear component created by surface roughness. A flat surface leads to minimal shear displacements and limited distortion of the surface oxide. In contrast, a rough surface increases shear displacements, promoting oxide film rupture. Secondly, the deformation occurring at the surfaces enhances diffusion through the diffusion through the generation of point defects above that for the equilibrium concentration. The application of temperature serves to provide the thermal energy necessary for diffusion. As the bonding temperature is increased, those diffusion processes that create the bond (bulk diffusion, grain boundary diffusion, volume diffusion) become more active, resulting in void elimination at the interface and atomic exchange across the interface. The final five diffusion bonded assemblies, HIP parameters, and fracture strengths are shown in Table 2. The compliant layer was either 1100-Al or AlBeMet-150 which was explosively bonded to the copper alloy heat sink. In some cases, the beryllium was coated with aluminum using an ion sputtering process.

The first two specimens (A-DB, B-DB) used processing developed at Rockwell International Corporation to bond Al-62Be (Lockalloy) to itself and to beryllium foils [16]. Much of this early work marked the direction taken in these later experiments. In this study, investigators compared the use of etchants and thin coatings of metals to eliminate oxides and act as diffusion aids. The latter three specimens used a thin coating of copper on the aluminum surfaces to eliminate the aluminum oxide in favor of a thinner copper oxide, and more importantly, it provided a concentration gradient at the joint interface that promoted the diffusion process. The copper diffuses rapidly through the aluminum and increases the diffusion current. Provided the copper concentration is kept low no harmful intermetallics will form.

Table 2
Diffusion bonded assemblies

Specimen ID	Substrate Materials	HIP Parameters	Fracture (MPa)	Strength
A-DB*	AlBeMet bonded to Be	150 600°C / 1 h / 105 MPa	86.1 98.5	
B-DB*	AlBeMet bonded to PVD Al on Be	150 650°C/ 1 h / 105 MPa	59.9 19.3	
C-DB	AlBeMet bonded to PVD Al coated Be with 1 μ m of Si on the Al	150 625°C/ 1 h / 105 MPa	Broke during machining	
D-DB	1100-Al bonded to PVD Al with 1 μ m of Cu on the Al surfaces	625°C/ 1 h / 105 MPa	113.3 116.3	
E-DB	1100-Al bonded to PVD Al with 1 μ m of Si on the PVD Al surface	625°C/ 1 h / 105 MPa	82.6 120.5	

* These surfaces were chemically etched to remove the oxides prior to assembly.

After HIP processing, the assemblies were de-canned and the assemblies evaluated using both metallographic examination and mechanical testing. The characteristics of the bond interface were examined by both optical and electron microscopy [17]. Tensile tests were conducted primarily at room temperature; selected specimens were also evaluated at 300°C.

The processing used to produce Specimen D-DB was selected as the best of the diffusion bonded assemblies and was thus selected as the process to be used to produce an EBTS specimen for the thermal fatigue tests. This sample survived the 5 and 10 MW/m² exposures without incident. All beryllium tiles were intact and indicated no change in the surface temperature. Following these tests, several tiles on the sample were subjected to heat loads in excess of 250 MJ/m² (0.5s) before some localized melting occurred.

4. Vacuum Plasma Spraying of beryllium on copper

One of the requirements for selecting a joining process will be the ease of joining large flat and curved surfaces of beryllium directly to copper. Plasma spraying has been identified for both beryllium and tungsten as a potential primary or backup technology for fabricating the armor on the primary and limiter first wall modules (which is approximately 1000 m²) and the wing and gas box liner in the divertor [18]. Plasma spraying is preferred due to the potential for providing thick armor coatings of beryllium and tungsten directly on large flat and curved copper surfaces. Research investigations on plasma spraying of beryllium have focused mainly on developing this technology for in situ repair applications for damaged beryllium first wall armor surfaces [10,19]. In this investigation two Be/Cu first wall mockups were fabricated by vacuum plasma spraying (at Los Alamos National Laboratory) and subsequently high heat flux tested at ITER relevant conditions to demonstrate the feasibility of using this technology for fabrication of the beryllium first wall structure.

The beryllium was plasma sprayed on two different copper alloy heat sinks for this investigation; 1) CuNiBe (Hycon-3) and 2) CuCrZr (Elbodur) which had 1100 aluminum explosively bonded to the surface with a 1mm titanium diffusion barrier interlayer between the copper and aluminum. Explosive bonding of the copper to the aluminum was provided by Northwest Technologies of Sequim, Washington. The beryllium was plasma sprayed on a flat section of the copper heat sink 87 mm long x 25 mm wide. Prior to plasma spraying, the copper and aluminum surfaces were knurled using electro-discharge machining (EDM) in order to enhance the mechanical bond between the beryllium coating and the substrate. During deposition a helium cooling gas was introduced through the copper heat sink to control the temperature. Cooling of the copper heat sink was done to minimize the formation of brittle intermetallics between the copper and beryllium and to prevent melting of the explosive bonded aluminum layer. Controlling the temperature also minimizes the build-up of thermal stresses during the deposition process.

The beryllium plasma sprayed CuNiBe alloy was subjected to 3000 cycles at the 1 MW/m² heat flux level without incident. At the 3 MW/m² level after 10 cycles the surface temperature increased indicating delamination. The failure was attributed to unmelted beryllium particles trapped within the plasma sprayed coating. The beryllium plasma sprayed CuCrZr was tested at heat fluxes up to 40 cycles at 5 MW/m². Based on these results, it was concluded that the tiles could have survived the 3000 cycles at the 1 MW/m² heat load level [20,21].

5. Bonding of tungsten to copper

Tungsten is the leading candidate for the divertor section of the ITER reactor, because of the lower erosion rate predicted by the tungsten over the other armor material candidates. Tungsten joining to copper presents several unique challenges. One of the primary challenges is the large difference

in coefficient of thermal expansion between the two materials ($4 \times 10^{-6}/K$ for W vs $18 \times 10^{-6}/K$ for Cu), which results in large thermal stresses during cooling from the bonding temperatures. This was evidenced in early attempts to bond tungsten plate (7mm) to a copper heat sink. High residual stresses deformed and eventually cracked the interface between the tungsten and copper. A second challenge is that relatively low bonding temperatures ($<500^{\circ}C$) are dictated by the copper alloy heat sink material. The CuCrZr is age-hardened to optimum strength in the $480^{\circ}C$ range. Overaging occurs rapidly above $500^{\circ}C$ resulting in degraded mechanical properties or the need to re-solutionize anneal at $1000^{\circ}C$, followed by a rapid quench, which brings along a host of distortion and residual stress problems.

The US approach to these fabrication challenges utilizes a tungsten brush structure which is joined to the Cu heat sink. In this technique, tungsten rods (1.7, 3.25 mm diameter) are held in position using an Inconel honeycomb core. Various coatings are employed to improve the tungsten-copper bond strength. The pre-coated rods are then bonded to the copper heat sink using diffusion bonding techniques. The most favorable technique currently is hot isostatic pressing (HIP). The HIP temperature is commensurate with the aging characteristics of the CuCrZr heat sink alloy ($450 - 480^{\circ}C$). The heat sink material is clad with 3 mm thick OFHC copper which provides a soft compliant layer in which to bond the tungsten rods. When subjected to the temperature-pressure HIP cycle, the tungsten rods are driven into the OFHC copper layer. The resulting deformation at the rod interface enhances the diffusion process and results in a greater bonding surface.

Several different coatings are being evaluated on the tungsten rod tips [22]. Estimates of the bond strength enhancement are being evaluated by vacuum hot pressing (VHP) individual, coated rods into a copper-clad heat sink and subsequently extracting. The force required to extract the rods from the substrate and the subsequent surface examination using scanning electron microscopy are evidence of their effectiveness. The coating techniques include: plasma spraying and ion sputtering. Rod tip geometries and surface roughness are also being evaluated. In addition, coated rods have been subjected to temperature excursions to evaluate the value of pre-heat treating prior to bonding to the copper heat sink. Table 3 summarizes the test data at this writing. These results compare favorably with the ultimate tensile strength (100MPa) for OFHC copper at $300^{\circ}C$.

Attempts are also being made to directly cast or plasma spray copper onto the tungsten rods and diffusion bond or braze the copper casting to the copper alloy heat sink as a two-step process [22]. The main problem has been with the first stage of this two-step process. Namely, to stop the "wetting" of the molten copper along the entire length of the rod bundle. The result is a PFC surface which consists of tungsten and copper which is not acceptable for the PFC's. Attempts to chemically leach the copper has been less than desired. Plasma Processes Inc. has been working on the plasma spraying, Surmet Corp. and the Boeing Co. have been working on the ion sputter coatings. Sandia National Laboratories in collaboration with the Efremov Institute (Saint Petersburg, Russia) has been working on the casting effort. The Efremov Institute has been successful in casting directly on a tungsten plate section using their casting techniques. This section was tested successfully in the EBTS facility at Sandia National Laboratories to 10 MW/m^2 for 1000 cycles.

Two mock-ups from the United States have been fabricated and are currently in line to be tested in the EBTS facility.. One mock-up used the direct plasma spraying technique and the other used a VHP technique to bond the tungsten rods to the copper cladding.

Table 3: Results of the tensile tests on the tungsten rods are shown below. All coatings are approximately 150 nm. Individual rods are pressed into OFHC copper using a vacuum hot press. The fracture load is the load required to extract the rods from the copper heat sink. Unless otherwise noted, the coatings are in the as-coated condition.

Table 3.
Tensile tests on tungsten rods.

W-rod coatings	Fracture Load (Kg)	Test Temp. (°C)
PS Ni/PVD Ni*	142	280
PVD Nb/PVD Ni	108	300
PVD Nb	91	300
PVD Cu	56	300
PVD Ni	TBD	
PVDCu/PVD Ni	61	300
PS Cu/PVD Ni*	16	280
Cast Cu	TBD	

* Rod coatings were heat treated at 1000°C for 1 hour prior to bonding to the copper heat sink.

6. Summary

This document has attempted to review all of the joining activities currently being conducted in the United States and associated with the ITER PFC's. Work not included but in progress includes the bonding of beryllium to copper by the Brush-Wellman Company.

1. Three bonding techniques (brazing, diffusion bonding, vacuum plasma spraying) developed by the T-221 team have been successful in joining beryllium to a copper heat sink:
 - a. HIP brazing using aluminum or AlBeMet-150 as a compliant layer and Al-12%Si as filler metal.
 - b. HIP diffusion bonding using aluminum or AlBeMet-150 as a compliant layer.
 - c. Plasma spraying beryllium on an aluminum-clad copper alloy heat sink.

The first two bonding techniques (a, b) have been used to produce high heat flux mock-ups which survived 1000 cycles at the 10 MW/m². The latter processing (c) has survived the 1 MW/m² without incident and is being considered for first wall and wall repair applications.

2. Several bonding techniques are being evaluated for joining tungsten to a copper heat sink:
 - a. HIP diffusion bonding of tungsten rod bundles into a OFHC copper clad heat sink.
 - b. Plasma spraying copper directly on tungsten rod bundles.
 - c. Casting copper directly on tungsten rod bundles.

All these processes are using innovative W-rod surface preparations to improve bonding. W-Rod tensile tests have shown the value of judiciously selecting the interfacial coating for the bonding assembly.

7. Conclusions

1. The use of aluminum as an intermediate layer facilitates the joining of beryllium to copper. The results of this study indicate that after coating the beryllium armor plate substrates with a thin layer of aluminum and after cladding the copper alloy heat sink with aluminum (with an appropriate diffusion barrier), the two components can be successfully joined by several joining techniques. High heat flux testing of these bonded assemblies indicates that heat flux loads of up to 10 MW/m² can be tolerated without failure. This approach could be used to join a beryllium armor tile to a stainless steel structure.

2. The use of a copper film on aluminum surfaces to eliminate aluminum oxide and promote diffusion by presenting a concentration gradient was successful. Bond fracture strengths at room temperature were at 100% efficiency based on the tensile strength of pure aluminum (105 MPa). The fracture morphology was dimple rupture. Extensive necking occurred in the bond region which indicates good defect tolerance.
3. The approach of explosively bonding aluminum to copper, coupled with the use of a thin titanium (125-250 μm) diffusion barrier, results in a metallurgically stable system at the expected interface temperatures.
4. The HIP process can be used to produce superior Al-12%Si braze joints. By controlling the bonding parameters, consolidation of plasma sprayed deposits can be made to occur early in the cycle, prior to the formation of a liquid phase, resulting in improved microstructures and mechanical properties. However, control of both coating thickness and filler metal quantity is still required to prevent complete melting of the aluminum layer.
5. Copper can serve as a substitute for the Al-12%Si filler metal system. The quantity of copper needs to be controlled to prevent the problems of excessive (or inadequate) filler metal volume at the bonding temperature.
6. Rod pull tests on tungsten rods indicates that pre-coating the rods with judiciously selected films and thermal treatments will improve the bond strength between the w-rod and the copper alloy heat sink.

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