CONF-9710153--

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Solid State Bonding of Beryllium-Copper for an ITER First Wall Application

(To be published in the *Proceedings of the Third IEA International Workshop on Beryllium Technology for Fusion*, Mito City, Japan, October 1997)

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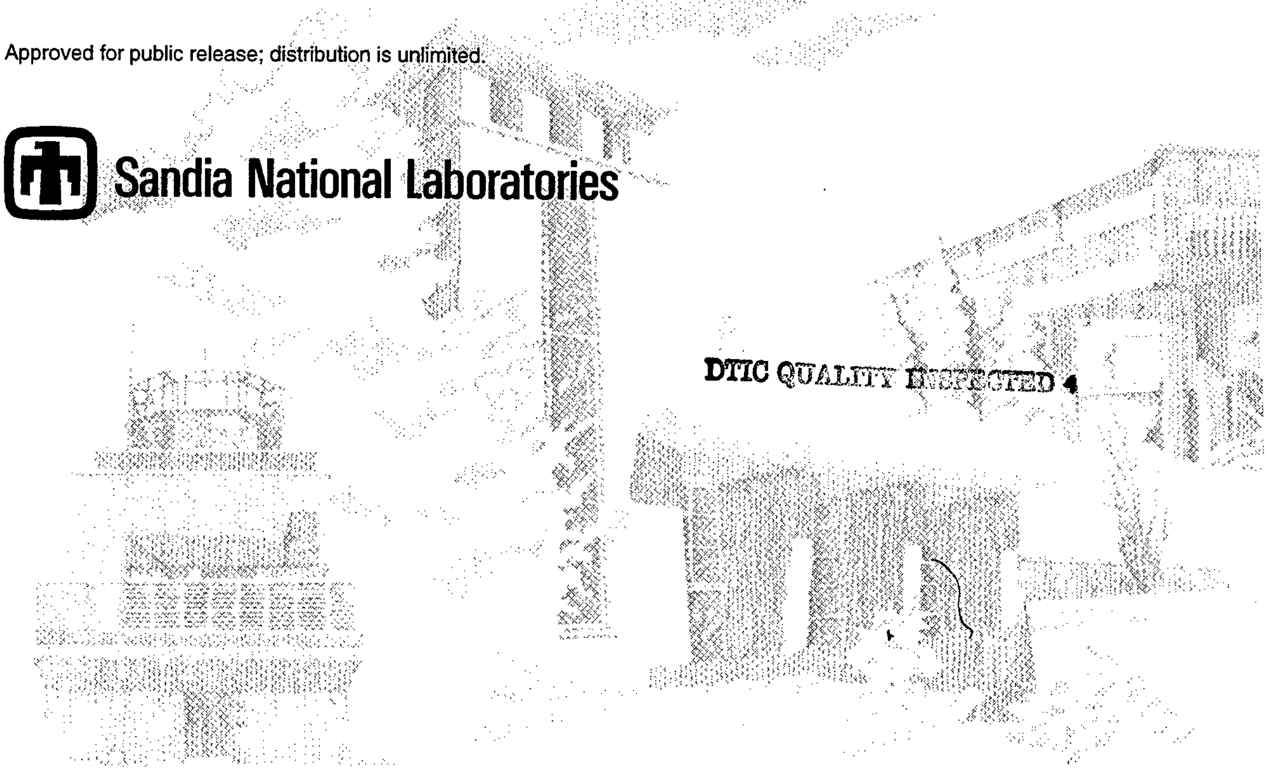

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SOLID STATE BONDING OF BERYLLIUM-COPPER FOR AN ITER FIRST WALL APPLICATION

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ABSTRACT

Several different joint assemblies were evaluated in support of a manufacturing technology for diffusion bonding a beryllium armor tile to a copper alloy heat sink for fusion reactor applications. Because beryllium reacts with all but a few elements to form intermetallic compounds, this study considered several different surface treatments as a means of both inhibiting these reactions and promoting a good diffusion bond between the two substrates. All diffusion bonded assemblies used aluminum or an aluminum-beryllium composite (AlBeMet-150) as the interfacial material in contact with beryllium. In most cases, explosive bonding was utilized as a technique for joining the copper alloy heat sink to an aluminum or AlBeMet-150 substrate, which was subsequently diffusion bonded to an aluminum coated beryllium tile. In this approach, a 250 μm thick titanium foil was used as a diffusion barrier between the copper and aluminum to prevent the formation of Cu-Al intermetallic phases. In all cases, a hot isostatic pressing (HIP) furnace was used in conjunction with canned assemblies in order to minimize oxidation and apply sufficient pressure on the assembly for excellent metal-to-metal contact and subsequent bonding. Several different processing schedules were evaluated during the course of this study; bonded assemblies were produced that failed outside the bond area indicating a 100% joint efficiency.

ACKNOWLEDGEMENTS

The authors would like to thank Mssrs S. Sastri (Surmet Corp), W. Bonivert (SNL/CA) and D. Butler (Northwest Technical Industries, Inc) for providing the coated beryllium and copper samples and Mssrs. K. Slattery and R. D. Watson for their valuable contributions in consulting, manufacturing, and testing throughout this study. A special thanks to C. Rood, A. Gardia, and N.Y.C. Yang for their assistance in the evaluation of the surfaces using optical and electron microscopy. Finally, we would like to acknowledge the work done by S. Goods and M. Tootle in the set-up, testing, and evaluation of the tensile specimens.

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1. INTRODUCTION

The method of affecting a bond between two dissimilar or similar metals without the presence of a liquid phase is called diffusion bonding. For diffusion bonding to occur, the bare metal surfaces must come in contact at a distance close to that of the crystal lattice constant. In order for that to happen, the surfaces must be free of contamination and oxides. When these conditions have been met, temperature and pressure accomplish the desired bond. Reactive metals such as beryllium and aluminum form tenacious oxide films which are especially difficult to break up. However, these metal oxide films must be broken in order to obtain intimate contact and subsequent bonding between the metal surfaces. Much of the effort in developing a diffusion bonding process concerns minimizing or eliminating the effects of these oxide layers by adjusting the bonding parameters.

The application of pressure during the bonding process creates a shearing action at the bond interfaces which breaks up the oxide film and exposes bare metal surfaces. Also, through this shearing action, the diffusion process is accelerated through the generation of line and point defects. These bare metal-metal contact points mark the genesis of the diffusion bonding process. Relatively rough surfaces and high bonding pressures will enhance the diffusion process. 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 and atomic exchange across the interface.

The application of a thin coating of metal which is compatible with the substrates provides a concentration gradient which further enhances diffusion. The other benefit gained by this coating is the replacement of the aluminum and beryllium oxides with a less tenacious oxide. Finally, a judicious material selection has to be made if the materials are to be compatible with both the beryllium armor tile and the copper heat sink material. That is, they do not form intermetallic compounds with either substrate. Those elements that do not form intermetallic compounds with beryllium include aluminum, germanium, silicon, and silver. Silver was precluded because of the transmutation products produced in the high neutron ITER environment [1]. The remaining possibilities were evaluated either in the brazing study conducted in joining these two materials [2] or in this study on diffusion bonding. Aluminum seemed the likely candidate for a compliant layer selection. It possesses good ductility at the expected interface service temperature (180°C) and excellent thermal conductivity but limited strength. However, the beryllium tiles would be sitting on a compliant layer which could not sustain residual stresses (derived from the thermal expansion differences between beryllium and copper) much greater than the flow stress of the aluminum (<100 MPa). Further, the compliant layer material would undergo annealing at the service temperatures which would further limit the retention of residual stresses. The material selection premise was to promote ductility over strength for this bond configuration.

In this study, the copper heat sink material (Hycon-3) was clad with a 1 mm layer of either 1100-Al alloy or AlBeMet-150 (50w/oBe-50w/oAl) using a thin titanium diffusion barrier (0.25 mm) as the interfacial material isolating the copper from the aluminum. This layered composite was produced using an explosive bonding technique. Titanium as well as tantalum have been used in industry to separate these two materials. The chemical stability of these composite layers has been tested to the melting point of the aluminum [3]. Beryllium armor tiles were coated with a thin layer of aluminum. Thus, the diffusion bonding problem was reduced to bonding aluminum to itself if the material selections had been chosen correctly. The surfaces of the aluminum were coated with a thin coating of copper or silicon to eliminate the aluminum oxide and enhance the diffusion flow by producing a concentration gradient. If the titanium diffusion barrier could eliminate the reactions between beryllium-copper and copper-aluminum, the process should work. Much of the work in selecting materials for this study was made easier by an earlier study where brazing was used as the joining process [4].

2. EXPERIMENTAL PROCEDURE

The beryllium used in this study was Brush-Wellman grade S-65C, a product made by the vacuum hot pressing of impact ground powder. The precipitation hardenable copper alloys utilized were CuCrZr (Elbrodur G, Kablemetal) and CuNiBe (Hycon-3, Brush-Wellman). Both of these alloys were age hardened in the same temperature range [460-480°C] and soften quite rapidly at temperature above 500°C. The beryllium and copper alloy components were coated, with aluminum or aluminum alloys, by either explosive bonding, plasma spray (PS) or ion sputtering (PVD) techniques. Chemical composition of the various test materials are shown in Table 1.

Table 1
Nominal composition of test materials used in this study

Alloy Designation	Composition (wt. %)
CuCrZr (Elbrodur G)	Cu - 0.65%Cr - 0.10%Zr
CuBeNi (Hycon-3)	Cu - 1.4-2.2%Ni - 0.2-0.6%Be
S-65C	Be - <1.0%Be - Fe/Al impurities
AlBeMet-150	50%Be-50%Al
1100-Al	99% Al (minimum)

Several different bonding sequences were evaluated in this study. All specimens consisted of essentially the same cylindrical geometry, shown schematically in Figure 1. A beryllium disk (S-65C) was sandwiched between two cylinders of coated copper. All of the components were 48 mm in diameter. Each specimen was assembled in a cylindrical stainless steel can and vacuum baked for 4 h at 100°C to remove the majority of surface moisture. A cap was then electron beam welded to the end of the can completing the encapsulation process and ensuring an inert environment during subsequent processing. The canned assemblies were then hot isostatically pressed (HIP).

Table 2 lists the specimen identification, materials, and HIP processing parameters for each specimen. The 1100-Al and AlBeMet-150 layers were explosively bonded to the copper alloys using an intermediate layer of 250 µm thick titanium designed to act as a diffusion barrier. Note that silicon and copper films were used to provide a concentration gradient and eliminate the aluminum oxide film at the bond surface. After first sputter etching the bonding surface to remove native oxide films, these thin coatings were applied by ion sputtering. Where these thin coatings were not employed, bond surfaces were: (1) chemically etched in an aqueous solution containing 45%HNO_3 and 10% HF, (2) rinsed in de-ionized water and (3) forced air dried immediately prior to assembly.

After HIP processing, the candidate assemblies were de-canned from the stainless steel and specimens were removed for metallographic examination and mechanical testing using wire electron discharge machined (WEDM). The specimen orientation is shown in Figure 1. 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. These specimens were inexpensive to both machine and test, although they did not duplicate the in-service loading stresses found in real components. However, this test was used to compare various bonding sequences and not to determine absolute bonding strengths. Tests were conducted primarily at room temperature; selected specimens were also evaluated at 300°C.

3. TEST RESULTS

The room temperature tensile test results are shown in Table 3. Despite etching to remove surface oxides immediately prior to assembly, DB-1 and DB-2 exhibited problems associated with the presence of oxides at the bond interface. These oxides were visible, using optical metallography, as a thin black line at the interface. Tensile bar fracture surfaces appear as mixed mode consisting of dimple ductile rupture and regions of poor bonding where the surface had an oxide film.

DB-3 and DB-5 exhibited problems associated with the presence of a silicon-rich film at the beryllium interface. There is very little solubility of silicon in either beryllium or aluminum at room temperature. For reasons that are not completely understood, silicon in this system tends to segregate to the beryllium interface, particularly if the aluminum coating thickness (on beryllium) is kept low. In DB-3, this film was nearly continuous and failure occurred at a very low stress while machining. In DB-5, the silicon-rich film was much less prominent and consisted of a discontinuous phase. DB-4 exhibited no detectable bond line which indicates that sufficient diffusion had occurred to eliminate the presence of oxides and porosity at the bond interface. The copper promotes diffusion by providing a concentration gradient and eliminating the presence of an aluminum oxide. However, it is not of sufficient concentration to promote the formation of beryllium intermetallics at the beryllium surface. The fracture stress is essentially that of the tensile strength of pure aluminum.

Table 2
Identification, coatings and HIP parameters for diffusion bonded specimens

Specimen ID	Be Coating	Cu Coating*	HIP Parameters
DB-1	none	AlBeMet-150	600°C / 1 h / 105 MPa
DB-2	25 µm PVD Al	AlBeMet-150	625°C / 1 h / 105 MPa
DB-3	25 µm PVD Al + 1 µm PVD Si	AlBeMet-150	625°C / 1 h / 105 MPa
DB-4	300 µm PS Al + 100 nm PVD Cu	1100-Al + 100 nm PVD Cu	625°C / 1 h / 105 MPa
DB-5	25 µm PVD Al + 1 µm PVD Si	1100-Al	625°C / 1 h / 105 MPa
DB-6	100 µm PVD Al + 300 nm PVD Cu	AlBeMet-150 + 300 nm PVD Cu	625°C / 1 h / 140 MPa

*The explosive bonding of copper/titanium/aluminum plates was performed at Northwest Technical Industries, Inc. in Sequim WA, USA.

Necking was extensive in the bond regions which indicates excellent defect tolerance. The fracture surface exhibited dimple ductile rupture.

Based upon the results obtained for DB-4, another specimen (DB-6) was fabricated using PVD techniques to coat both the beryllium and AlBeMet-150 bonding surfaces. This specimen exhibited joint microstructures similar to DB-4 but fractured at a higher stress, presumably due to the improved strength of AlBeMet-150 compared to 1100-Al. Figure 2 shows the tensile bars from DB-6. The ductility of the bond region is evidenced by the extensive necking in these tensile bars. Microstructural features from cross-sections of this specimen are shown in Figures 3 and 4. Figure 3 shows an over-all view of the Be/AlBeMet-150 interface. Figure 4 shows an overall view of the AlBeMet-150/Hycon-3 interface. In Figure 4, the explosive bonding process has produced intrusion of the AlBeMet-150 into the titanium diffusion barrier. This was seen in several locations along the interface. Electron diffraction analysis provided the identity of these extrusions.

The processing used to produce DB-6 was selected as the best of the diffusion bonding series and was thus selected for high heat flux testing in Sandia National Laboratories Electron Beam Test Site. A specimen was fabricated according to the schematic shown in Figure 5. The sample survived the 5 and 10 MW/m^2 exposures without incident. All beryllium tiles were intact and indicated no change in the interface temperature. Following these tests, several tiles were subjected to approximately 250 MJ/m^2 (0.5s) heat loads to simulate vertical disruption events. The beryllium tiles melted to form dome-like shapes where the flat tiles had once existed.

4. DISCUSSION

Etching with a dilute solution of HF/HNO_3 to remove oxides prior to bonding as was the case for the first two bonding configurations was not practical on a full-scale unit and at best was very difficult with lab-scale units. The etchant reacted very quickly with the beryllium. Cooling the etchant in an ice-bath to reduce the reactivity to a controllable rate resulted in a 10s immersion before a visible film began to form on the beryllium surface. Aluminum was etched with a NaOH solution. The transfer time to a vacuum chamber was critical if re-oxidation was to be kept at a minimum. Earlier studies by investigators had attempted to bond beryllium to Lockalloy (62%Be-38%Al) using a similar process [5]. They found that the beryllium removal rates using the HF/HNO_3 were as high as 1mm/m. In that study, an etching technique was developed in conjunction with a subsequent ion sputtered copper or silver coating on all surfaces to eliminate the oxidation problems and improve the likelihood of a good bond. In the current study, etching alone was insufficient surface preparation to obtain consistently good bonding (DB-1, DB-2), leading to the use of sputtering to both remove surface oxides and provide diffusion enhancers. Germanium was tried in the brazing experiments and excluded from this series of tests. The germanium was found to segregate to the beryllium interface and significantly reduce the bond strength. Silicon, if used in small volumes appeared to work, although, silicon-rich precipitates were observed at the beryllium interface. Copper worked the best and resulted in good bond strengths with no interfacial precipitates when the copper film thickness was kept low and aluminum volumes were sufficient to dilute the copper.

5. CONCLUSIONS

1. Diffusion bonding of aluminum surfaces to themselves is difficult because of the oxides formed by this very reactive material. Attempts to remove the oxide by using a $HF-HNO_3$ dilute solution (DB-1, DB-2) just prior to canning were only moderately successful

2. Diffusion bonding of aluminum surfaces using a silicon film to eliminate aluminum oxide were unsuccessful (DB-3, DB-5). The silicon migrates to the beryllium interface and segregates as a

silicon-rich phase. The dilution of this segregate can be improved by using thicker aluminum coatings (DB-5).

3. 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.

Table 3
Metallographic and Tensile Test Results

Specimen ID	Metallographic and SEM results and remarks	Fracture Stress at Room Temperature
DB-1	oxide film still visible at the bond interface	86 MPa 99 MPa
DB-2	oxide film still visible at the bond interface	60 MPa 19 MPa
DB-3	semi-continuous, silicon-rich phase at the beryllium interface	broke while machining
DB-4	no apparent oxides, bond line was not visible	113 MPa, 116 MPa
DB-5	semi-continuous, silicon-rich phase at the beryllium interface	83 MPa 120 MPa
DB-6	no apparent oxides, bond line was not visible	195 MPa 98 MPa (300°C)

4. The diffusion bonding process developed using the aluminum-beryllium compliant layer can produce beryllium-copper bonds that will survive at the 5-10 MW/m^2 level without failure.

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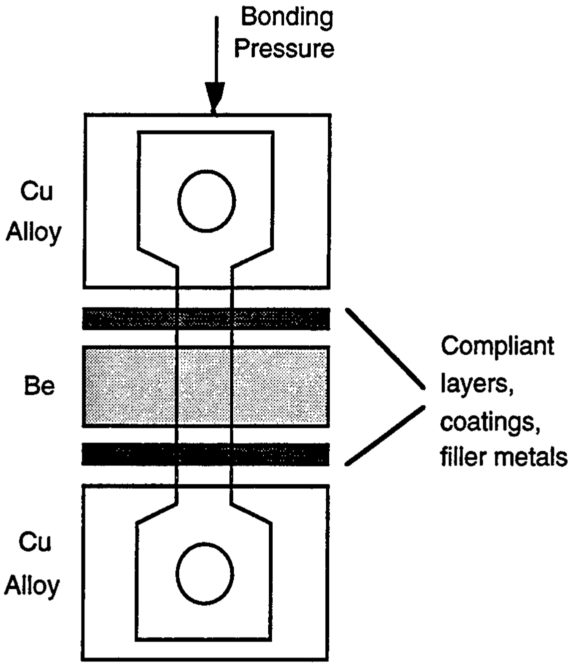


Figure 1. Bond assembly configuration showing orientation of specimen for transverse tensile testing.

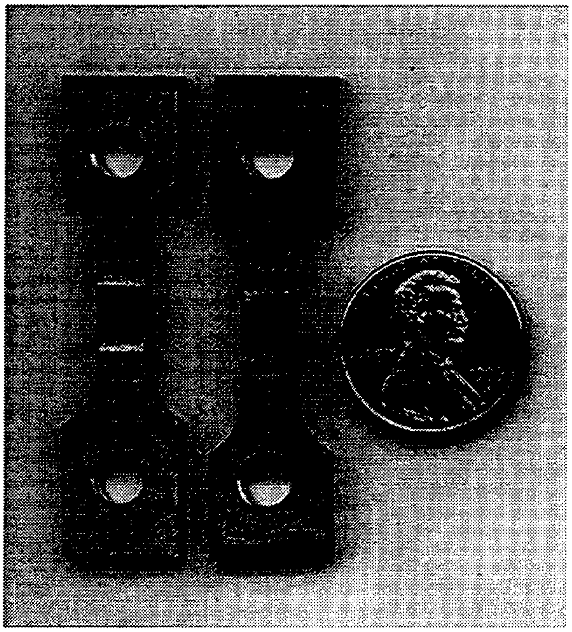


Figure 2. Photograph showing tensile bars taken from the diffusion bonded sample (BD-5). Note the extensive necking in the sample on the left.

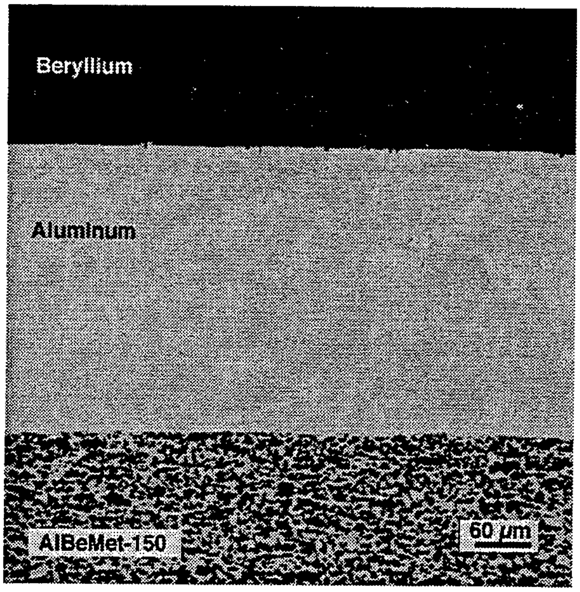


Figure 3. Micrograph showing the beryllium-aluminum and aluminum-AlBeMet-150 diffusion bond in specimen DB-6.

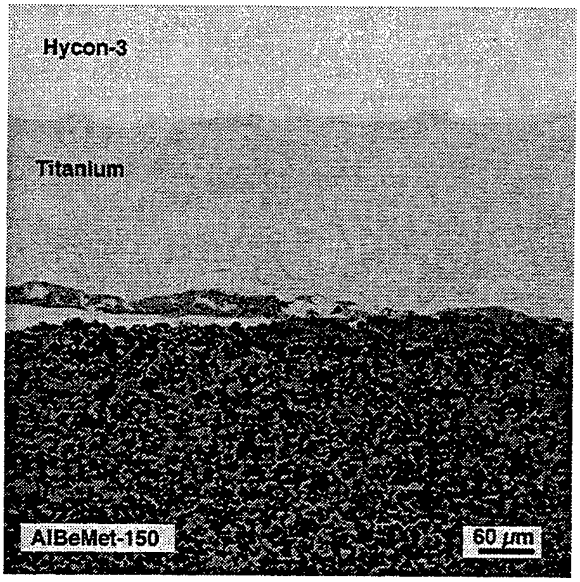
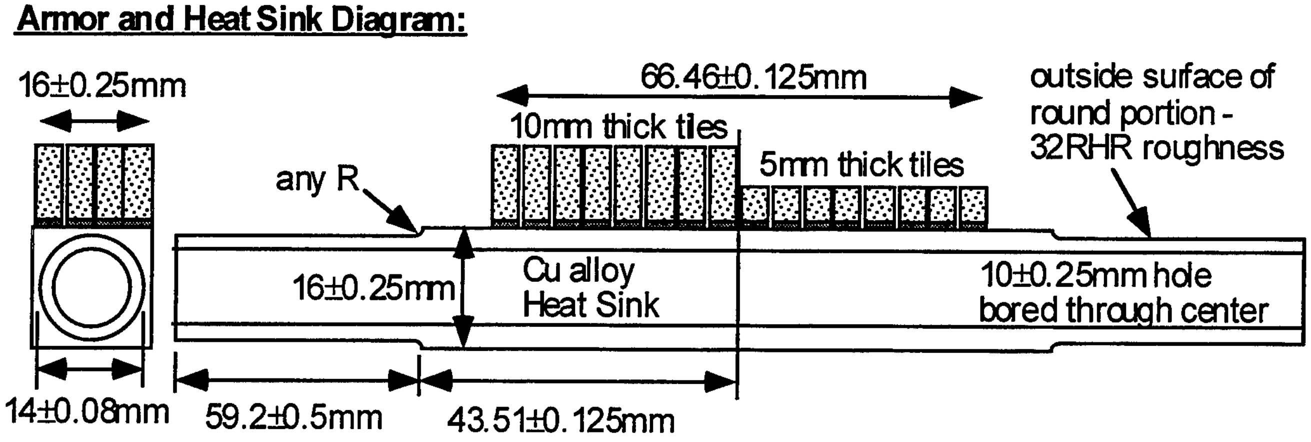


Figure 4. Micrograph showing the AlBeMet-150 to copper alloy explosive bond. Note regions of the AlBeMet-150 have intruded into the titanium.

Figure 5.
EBTS Test Specimen geometry



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Report Number (14) SAND--97-8637

CONF-9710153--

Publ. Date (11) 199802

Sponsor Code (18) DOE/ER , XF

UC Category (19) UC-404, DOE/ER

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