

# High-Strength Low-Temperature Bonding of Beryllium and Other Metals

Bonds exhibiting a beryllium fracture strength of 40 ksi can be made at 225° F on beryllium plated with 500  $\mu$ in. of silver

BY J. L. KNOWLES AND T. H. HAZLETT

**ABSTRACT.** An investigation has been carried out to determine the feasibility of developing techniques for bonding beryllium as well as other metals at low temperatures and to establish the relationship of the bond parameters to the bond strength. This study was guided by the following conditions:

1. Bonding temperature should be less than 600° F.
2. The plastic deformation of the base metal should be minimized.
3. The bond should be nearly as strong as the base metal.

The final condition, that the bond be nearly as strong as the base metal, was intended to apply to type S200C beryllium. Most of this study utilized beryllium but additional tests were made on molybdenum, stainless steel, tungsten, and commercially pure copper.

It was established early in the investigation that bonds could not be accomplished within the required temperature range and with small base metal distortion without the use of an intermediate material. Consequently, all the results reported are for specimens with a plated dissimilar metal, generally silver, on the surfaces being joined. It was also demonstrated that a special cleaning treatment was required after this second material was applied.

Bonding atmospheres varying from a vacuum of  $2 \times 10^{-6}$  torr to helium, oxygen, and air were studied. These widely differing bonding environments have no significant influence on bond strength.

Variations in the bonding procedure, resulting in a difference in strengths obtained, lead to the conclusion that the elastic strain energy in the substrate acts to destroy some portion of the bond by fracturing a portion of the bond area.

For a fixed applied bonding compressive stress, an increase in bond strength

resulted when the modulus of elasticity of the substrate was increased. The thickness of the intermediate material was also significant in determining bond strength for the same processing conditions. When the plating thickness was varied from 35 to 200  $\mu$ in., the bond strength was approximately doubled.

The presence of the plated material and the subsequent preparation of the surface resulted in a very small notch in the surface of the specimen at the bond interface. When this imperfection was removed after bonding, the bond strength was again almost doubled.

Data from beryllium plated with 200  $\mu$ in. of silver show that a partial bond was formed at a temperature of slightly less than 225° F. At a bond temperature of 280° F, bond strengths in beryllium can be obtained approaching the bulk material strength when the notch is removed. Also, bond strength increases linearly with increased applied bonding compressive stress. This result fits the adhesion model of bonding.

It may be concluded from this study that bonds (exhibiting beryllium fracture strength of 40 ksi) on beryllium plated with 500  $\mu$ in. of silver (especially prepared after plating) can be made at 225° F. The data also invalidate a conclusion reached by at least one other investigation that the minimum theoretical temperature for such bonding is 390° F, i.e., the temperature at which Ag<sub>2</sub>O decomposes.

Experimental data obtained from this investigation indicate that an adhesion or cohesion model is the bonding mechanism. This conclusion is based on the linear relationship of the bond strength-to-bonding compressive stress and the independence of bonding time.

Bond strengths are dependent upon the thickness of the plating; bond strengths increase with increasing thickness up to 600  $\mu$ in.

## Introduction

Fusion welding and brazing are the most common metallurgical techniques employed to join metals because they are usually the most economical.

Not all applications lend themselves to these techniques, however. Fusion welding is not normally used to join dissimilar metals having widely differing properties. In addition, because of the high temperatures required, unfavorable metallurgical changes often occur in the materials being welded, and high residual stresses may lead to distortion or even failure of the structure. Many similar and dissimilar metal couples cannot be brazed for lack of a material which will adhere to or "wet" the mating metals. Again, relatively high temperatures are usually required which may lead to degradation of the material properties and/or physical distortion of the structure arising from the thermally induced stresses. The necessary high temperatures also limit the selection of materials which may be used in the vicinity of the weld. Beryllium, for example, is used quite extensively in inertial guidance instruments. Many nonmetallic components used inside these instruments cannot survive the temperatures used to braze or weld the assembly. Thus, mechanical joints are used that increase the weight of the system—an undesirable feature for many applications.

A review of the literature on solid state welding of beryllium by Hauser et al.<sup>1</sup> revealed that it is possible to achieve solid state bonds; however, the welding temperatures required were far in excess of 600° F. By utilizing an ultrahigh vacuum and ion-sputter surface-cleaning technique before bonding, Batzer and Bunshah<sup>2</sup> measured bond strengths of approximately 16% of the bulk strength of beryllium after bonding at 572° F. Although this procedure for achieving high-strength low-temperature bonding appears promising, the techniques employed were not considered feasible from a production point of view.

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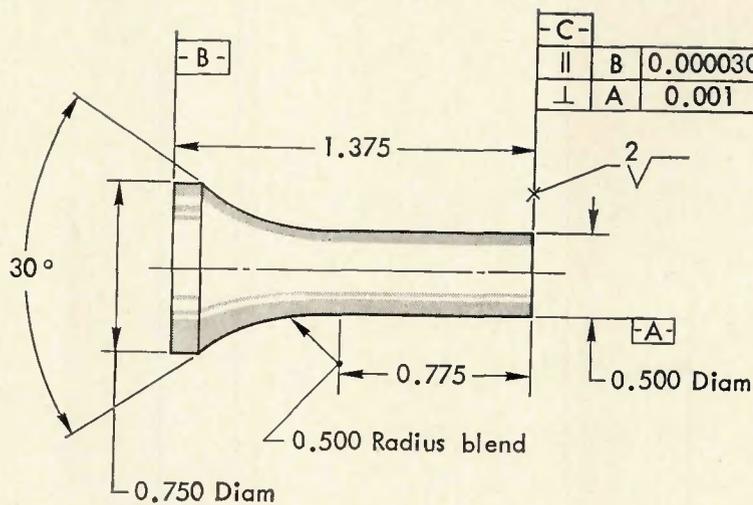


Fig. 1—Specimen details

Beryllium has also been successfully joined by introducing another metal at the interface and elevating the temperature until a liquid phase is achieved. This process has been referred to as diffusion or eutectic bonding but generally is considered to be a braze joining process. The minimum bond temperature reported was 1330° F with an aluminum foil.<sup>3</sup> Maloof and Cohen<sup>4</sup> brazed beryllium with silver foils, silver electroplates, and a ductile beryllium-20 at. % silver alloy foil. During the brazing cycle, intergranular penetration of silver into beryllium was suspected but was not considered harmful due to the ductile nature of the resulting alloy. It was also established that liquid silver would wet the beryllium surface.

Crane et. al.<sup>5</sup> have diffusion-bonded aluminum to stainless steel with electroplated silver on each surface at temperatures of 375 to 700° F. Their bonds were made in an air environment, and the compressive load was adjusted to achieve a deformation of approximately 0.005 in. in the 0.125-in. thick aluminum. Metallurgical examinations of the silver-silver joint for all time-temperature combinations tested revealed a void-free

fine-grained, recrystallized silver structure. The ability to develop a solid state bond at the silver-silver interface in an air environment was attributed to the lack of detrimental oxides or other non metallic films, since the free energy of formation of Ag<sub>2</sub>O is essentially zero at approximately 390° F.<sup>6</sup>

In summary, it was concluded that beryllium could not be joined to itself under the limitations imposed by this investigation without an intermediate material. There was no evidence to indicate that a beryllium-metal diffusion bond could be developed at less than 600° F. Thus, it was decided to plate the bond surface of the beryllium with another metal and rely upon the plating process to achieve a strong beryllium-plate adhesion. The plated surfaces would then be joined while subjected to a compressive load at a low temperature. Silver was considered a logical selection, because it wets beryllium and because silver-silver bonds had been achieved at 500° F.

It is generally accepted by most brazing technologists that a thin void-free braze joint is stronger than a thick one. Although this relationship was mentioned often in the early trade literature, Crow<sup>7</sup> is credited with the first systematic study. This relationship was further reinforced by Leach and Edelson,<sup>8</sup> who showed that, in brazing stainless steel with a quaternary silver alloy, a maximum butt-joint strength was obtained with 0.0015 in. thick braze joint. Either an increase or a decrease in the thickness of the joint resulted in a significant decrease in the bond strength. Later investigators concluded that bond strength continuously increases with decreasing joint thickness as long as the braze joint is void-free.

It was also observed that the strength of the braze joint approached that of the base metal which was

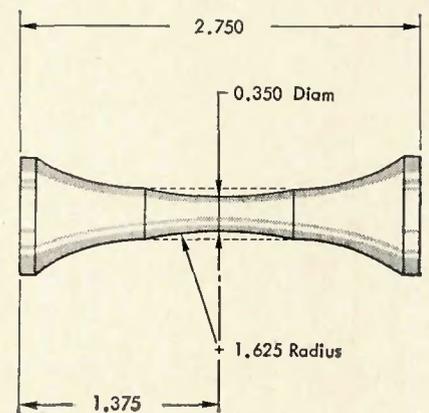


Fig. 2—Necked bonded specimen

often several times that of the bulk braze-material strength. Bredz<sup>9</sup>, for example, reports that a braze joint 0.00029 in. thick made with pure silver (ultimate tensile strength 21 ksi) on 1020 steel (ultimate tensile strength 69 ksi) fractured at 67.2 ksi. It is now well-established that a ductile material, if constrained from deforming due to the resulting stress system imposed, is capable of supporting extremely high stresses as compared to its unconstrained strength.

Meissner and Baldauf<sup>10</sup> concluded that if the triaxial stresses in the joint were in fact responsible for the increase in thin-film strengths, the joint strength should be a function of the geometry of the specimen. Bond strengths measured from polystyrene on steel, paraffin wax on steel, and eutectic lead-tin solder on brass revealed that the bond strength was related to the thickness-to-diameter ratio of the joint. Although this relationship is now generally accepted, the shape of the curve expressing this relationship has not been established. The curves found in the literature vary from linear, to exponential, to hyperbolic, to nondescript.

From the foregoing discussion it may be concluded that solid state bonds made from silver-plated metals

Table 1—Typical Chemical Analysis of Beryllium (S200C)<sup>a</sup>

Elements	— Weight-percent —	
	Heat 1	Heat 2
Be	98.2	98.33
BeO	2.0	1.90
C	0.15	0.14
Fe	0.17	0.18
Al	0.08	0.09
Si	0.05	0.04
Mg	< 0.02	< 0.02
N	< 0.04	< 0.04
Mn	< 0.01	< 0.02
Total (Cr, Cu, Ni, Ti)	< 0.20	< 0.20
Others, each	< 0.04	< 0.04

<sup>a</sup> Material specification: MEL-507D.

Table 2—Some Typical Mechanical Properties of Silver Plate and Substrate Materials

Material	Tensile modulus of elasticity, 10 <sup>6</sup> psi/in./in.	Tensile yield stress, ksi	Ultimate tensile stress, ksi
	Ag	10	8 <sup>a</sup>
Cu	17	44	48
SS(304)	28	30 <sup>b</sup>	75 <sup>b</sup>
Be	40	32-37 <sup>c</sup>	40.8
Mo	47	75	90
W	59	220	220

<sup>a</sup> Annealed.

<sup>b</sup> Minimum.

<sup>c</sup> Material supplier's data (range of four tests).

could exhibit strengths in excess of the ultimate strength of silver. However, the formation of a solid state bond differs considerably from that of a liquid braze joint in that a compressive load is necessary to establish contact of the interface surfaces. The stresses at the interface are not purely compressive but will also be influenced by the constraints imposed by the base metal. Since there is general agreement that, as a result of the triaxial state of stress, the bond strength increases as the joint thickness decreases, it would appear to be a logical extension that, for a given compressive load, the bond formation (i.e., contacting area at the interface) decrease as the joint thickness decreases. The strength of bonds thus formed will depend not only upon the triaxial state of stress, which is dependent upon the joint thickness-to-diameter ratio, but also upon the bond area. In view of the fact that the curve expressing this relationship for the simpler case of brazed joints has not been established, no prediction of the curve shape could be made for the solid state bonds investigated in this study.

### Experimental Equipment and Procedure

Silver-plated, cylindrical specimen pairs were joined by applying an axially compressive load at an elevated temperature. Preliminary studies revealed that the adherence of electroplated silver to beryllium by conventional processes was considerably less than the bulk strength of beryllium, but a vacuum-evaporation plating technique developed by Westlund<sup>11</sup> provided the necessary adhesion of silver to beryllium.

Thickness of the silver plate was measured nondestructively by X-ray fluorescence, and a polishing and cleaning technique was developed for preparing the silver surfaces before

bonding. A hydraulically actuated loading fixture was used to bond the specimens as well as to test the bond for its ultimate tensile strength. Vacuum bonding was achieved by installing the bonding fixture in a second vacuum system.

The specimens used (see Fig. 1) were solid cylinders with an enlarged tapered end to permit tensile testing. The bond end of the specimen (surface C) was polished flat to within 20 $\mu$ in. and parallel to the loading end (surface B) to within 30 $\mu$ in. Surface finish on the bond surface was 2 rms or better, except on the copper specimens for which it was 3 to 4 rms.

Necked bonded specimens were standard specimens, bonded, and then reduced in diameter to 0.350 in. by machining a 1.625 in. radius notch centered along the bond interface—Fig. 2.

All specimens were machined from 0.750 in. diameter stock. The chemical analysis of the beryllium substrate is shown in Table 1. The other materials were obtained from available stock and may be classified as follows:

1. Copper: Hard temper, electrolytic tough pitch. Material specification: QQ-C-502C, ETP.
2. Molybdenum: Arc cast, centerless ground, hot-cold rolled, leveled, fully stress-relieved. Material specification: AMS 7805.
3. Stainless steel: Cold finish and centerless ground, class 304, annealed, round. Material specification: QQ-S-763.
4. Tungsten: 99.9% pure, centerless ground. Material specification: MIL-T-14507A.

Some mechanical properties of the substrates used are shown in Table 2. The ultimate tensile strength of beryllium was obtained from an average of tests conducted on necked solid beryllium specimens tested in the bond fixture.

The plating and bonding systems were evacuated with diffusion pumps with a silicone base oil and achieved base pressures of  $2 \times 10^{-7}$  torr. These pumps were separated from the bell jar by a baffle and a liquid nitrogen cold trap. A typical plating sequence consisted of heating the specimens to a predetermined temperature (the temperature depending on the base metal) and then condensing silver vapor on the bond surfaces. The degree to which silver adhered to the substrate depended upon the cleanliness of the vacuum chamber and the specimen temperature at the time of plating. It diffusion pump oil migrated past the cold trap and into the chamber, silver could be peeled from the beryllium substrate. In the case of silver on beryllium, the beryllium must be heated to approximately 1100° F or higher to achieve an adhesion which exceeded the strength of beryllium. Table 3 lists the plate and substrate materials, substrate plating temperature, maximum bond strength achieved, and notes the fracture location. Except for the silver-beryllium couple, no effort was made to optimize the plating procedure. Thus, a cleaner vacuum system or higher substrate temperatures (lower for the gold-beryllium couple) would probably improve the plate-substrate adhesion.

Specimens were bonded as-plated or polished. It was found that a significant decrease in bond temperature could be realized if the as-plated silver bond surfaces were either etched or mechanically polished. The polishing procedure proved more repeatable and was accomplished by hand polishing with aluminum oxide and a standard lapping oil on a thick nap polishing cloth. Approximately 20 $\mu$ in. of silver was removed, and the surfaces were cleaned with a cotton swab saturated with acetone. This was followed

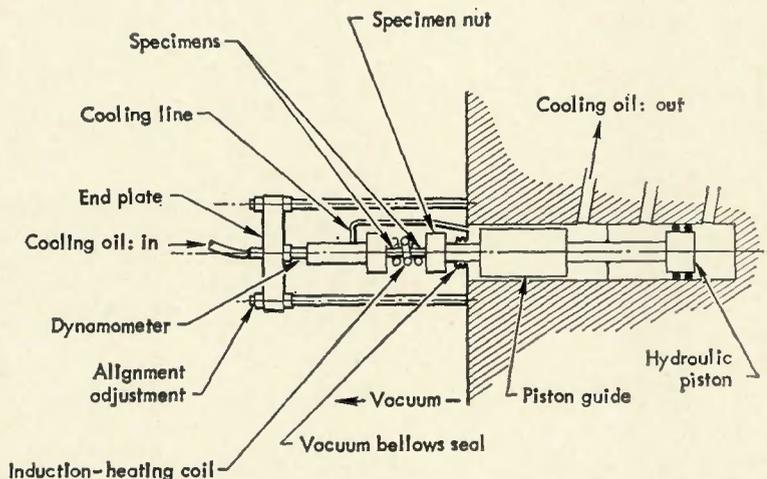


Fig. 3—Schematic drawing of bonding fixture

Table 3—Plating Parameters

Plate	Substrate	Substrate temperature, °F	Maximum adhesion, ksi
Ag	Be	1110-1380	44 <sup>a</sup>
	Cu	570	42 <sup>a</sup>
	Mo	1430	58 <sup>b</sup>
	SS(304)	1380	55 <sup>b</sup>
	W	1600	42 <sup>c</sup>
Au	Be	1430	10-20 <sup>b</sup>
	SS(304)	930	<sup>d</sup>
Cu	Be	1380	33 <sup>e</sup>
	Be	1200	10-15 <sup>e</sup>

<sup>a</sup> Substrate fractured, i.e., no plate peel.  
<sup>b</sup> Nearly 100% plate peel.  
<sup>c</sup> Fracture surface showed substrate fracture and plate peel.  
<sup>d</sup> Plate removed with transparent tape; intermetallic probably formed.  
<sup>e</sup> Fracture occurred at the Au-Au interface.

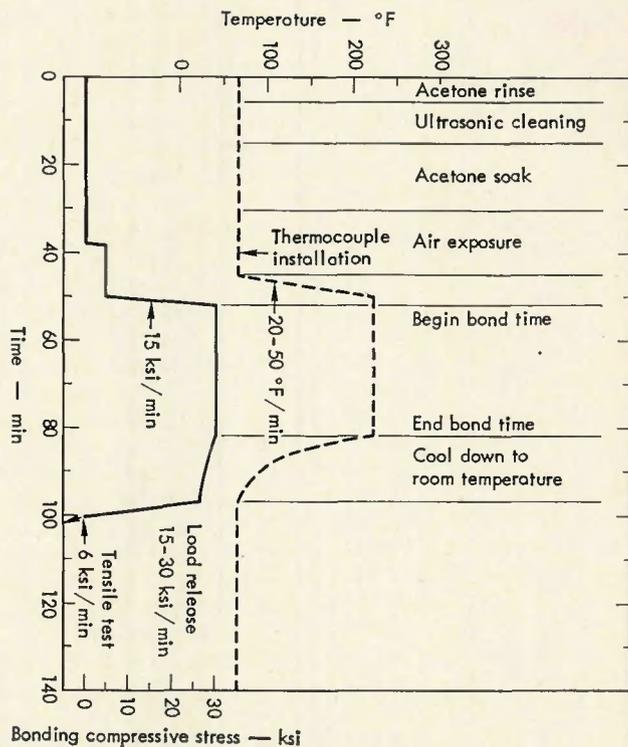


Fig. 4—Cleaning, installation, and bonding cycle

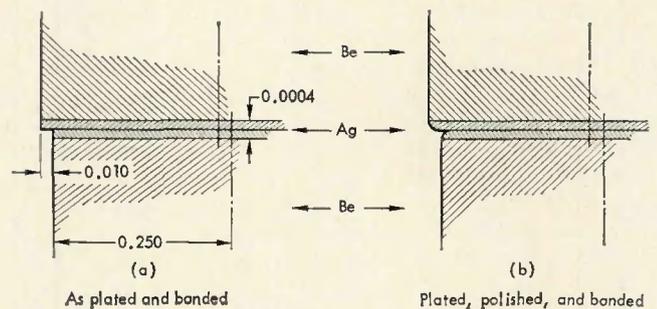


Fig. 5—Bonded specimen pairs with built-in notch

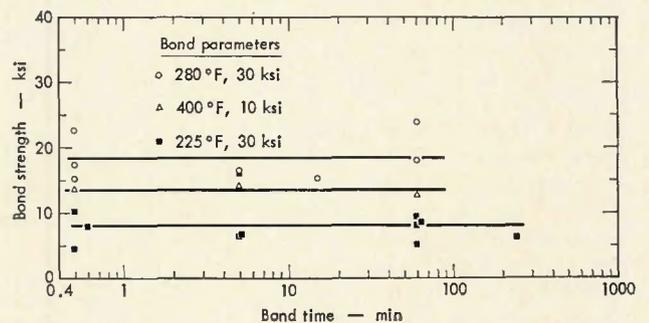


Fig. 6—Effect of bonding time for beryllium

by a low-power ultrasonic cleaning in acetone for approximately 10 min.

After cleaning, the specimens were installed in a bonding fixture— Fig. 3. The tapered ends of the specimens were held by a nut and collet arrangement. One specimen was moved axially by a double acting hydraulic piston acting through a close-fitting piston guide; the other specimen was attached to a dynamometer that was rigidly affixed to an end plate and in turn to four load-bearing rods attached to the system. The end plate could be adjusted to rotate about two perpendicular radial axes thus permitting the bond interfaces to be parallel. Cooling oil pumped through the dynamometer and around the piston guide prevented overheating of the dynamometer strain gages and excessive thermal expansion of the piston guide. The hydraulic loading system was limited to 5,000 lb tension which corresponds to a tensile stress of 25 and 50 ksi on the 0.50 and 0.35 in. diameter necked specimens, respectively. Bond strength data exceeding these values were obtained on a commercial tensile tester.

A typical cleaning, installation, and bonding cycle is shown in Fig. 4. The specimens were preloaded in air to 4.2 ksi, heated to their bond temperature, and loaded to the bonding compressive stress. This condition was maintained for the desired bond time and was terminated by cooling to room temperature.

As-bonded specimens (i.e., un-

necked, whether polished or as-plated) revealed 95 to 100% beryllium fracture surfaces and bond strengths of 20 to 36 ksi when the bond parameters were maximized. These strengths were considered low, since bulk beryllium fractures at 40 ksi. It was previously mentioned that the bonding fixture was aligned such that the bond interface surfaces were parallel. However, at the bond interface there was a radial misalignment of approximately 0.010 in. Thus, a bonded specimen pair had a built-in "notch," which would act as a stress-riser. This notch was even more severe, as shown in Fig. 5, for specimens polished at the silver interface, because the silver tended to round off at the edges as a result of the hand-polishing technique used.

Notches were removed by machining the shank as shown in Fig. 2. These necked specimens again exhibited 95 to 100% beryllium fracture for high-strength bonds; however, the bond strengths were 35 to 44 ksi (i.e., bulk beryllium strength). Most of the results reported herein were obtained on the 0.500 in. diameter specimens tested as-bonded for bond strengths less than 20 ksi.

## Results

The following definition of a standard bond will be assumed in this section unless otherwise noted:

1. Silver-plated substrate.
2. Nominal 200  $\mu$ in. of silver on each specimen half.

3. Silver polished before bonding.
4. Air preload (4.2 ksi) before bonding.
5. Bonded in air.
6. Bonding compressive load released at room temperature.

## Bond Time

Beryllium specimens bonded for times ranging from 0.5 to 240 min. reveal that the bond strength is insensitive to the bond time. These data, shown in Fig. 6, were further substantiated by results from stainless steel (310  $\mu$ in. of silver) and molybdenum specimens.

## Effect of Bonding Compressive Stress

Results obtained from beryllium (Fig. 7) indicate a temperature-dependent linear relationship between bond strength and the bonding compressive stress.

Numbers adjacent to the data points indicate the total number of bond strengths comprising the average value plotted. The absence of a number implies one test.

Results obtained at 225° F deviate from a straight line at approximately 27 ksi. This deviation may be explained by the onset of the plastic deformation of the beryllium as will be discussed later.

Two lines are shown for bonds made at 280° F. The higher strength data were obtained at the end of the investigation on specimens not ultrasonically cleaned in acetone. Additional tests also indicated that prolonged exposure of the polished silver

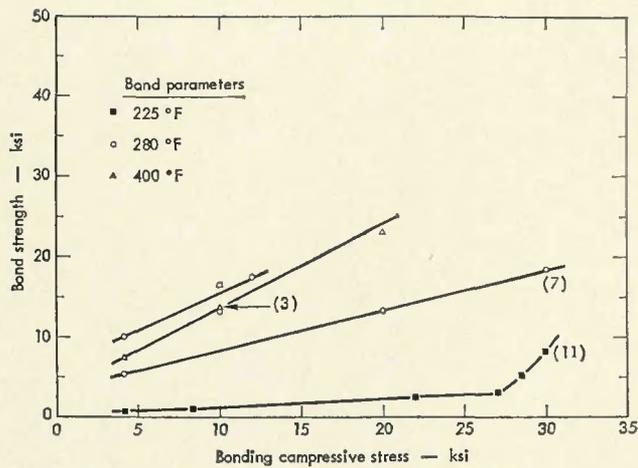


Fig. 7—Effect of bonding compressive stress for silver-plated beryllium

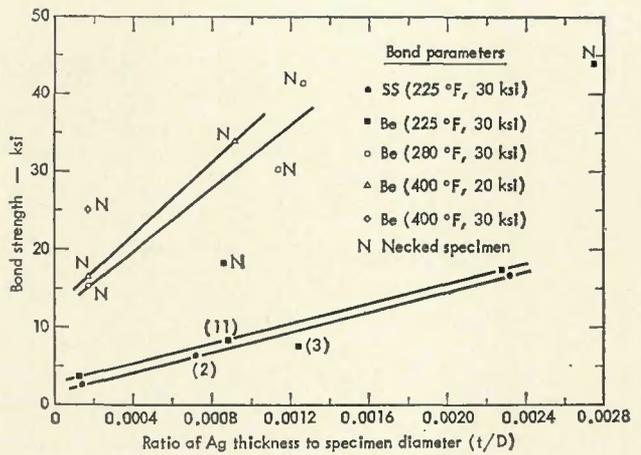


Fig. 8—Effect of ratio of total silver thickness-to-specimen diameter ( $t/D$ )

to acetone is detrimental to bond strength.

### Effect of Plating Thickness

Unnecked beryllium and stainless steel specimens plated with 30 to 580  $\mu$ in. of silver indicate a linearly increasing bond strength with plating thickness. These data (Fig. 8) have been normalized in the conventional manner by plotting bond strength as a function of the ratio of total silver thickness to specimen diameter ( $t/D$ ). Data for necked beryllium specimens bonded under the same conditions clearly show the increase in bond strength that results by removing the built-in notch. Additional data from necked beryllium specimens bonded at different temperatures and

compressive stresses are also shown.

### Effect of Bonding Temperature

Various substrate materials bonded at 30 ksi for temperatures ranging from 225 to 280° F reveal a linear relationship between the room-temperature bond strength and the bonding temperature—Fig. 9. The change in bond strength per unit change in temperature is independent of the substrate material within the temperature range reported. Additional data for beryllium bonded in a vacuum ( $2 \times 10^{-6}$  torr or less) confirm this linear relationship and also reveal that the bond strength is insensitive to air vs vacuum bonding environments.

Beryllium specimens bonded in vac-

uum in the as-plated condition at 24 ksi bonding compressive stress indicate a rapid increase in bond strength with change in bonding temperature near  $0.5 T_m$  of silver—650° F (Fig. 10).

### Effect of Release of Bonding Compressive Stress and Effect of Modulus of Elasticity of Substrate

Also shown in Fig. 10 is the influence of the stored compressive energy. Bond strengths comprising the lower curve were obtained from specimens for which the bonding compressive stress was reduced to 4.2 ksi after the bond cycle was completed, i.e., when the specimens began to cool to room temperature. Higher bond strengths were obtained on those spec-

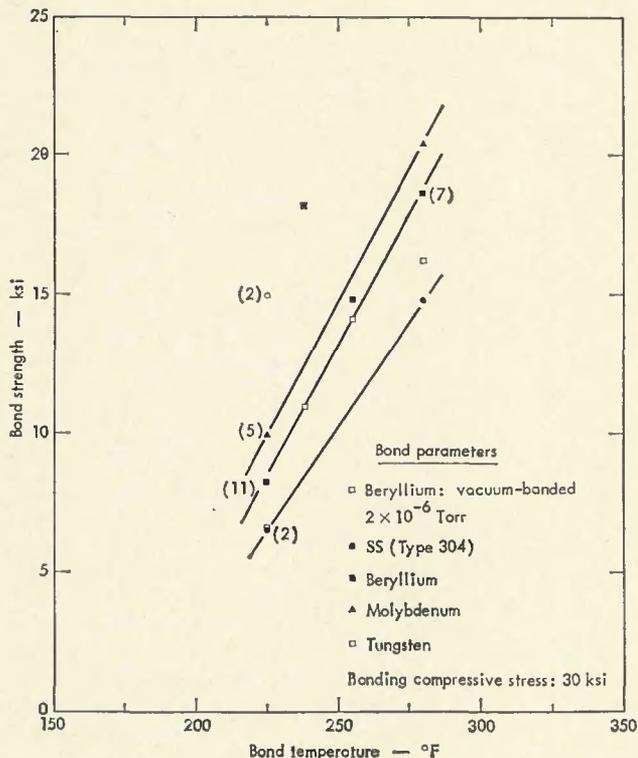


Fig. 9—Effect of bonding temperature for various silver-plated metals

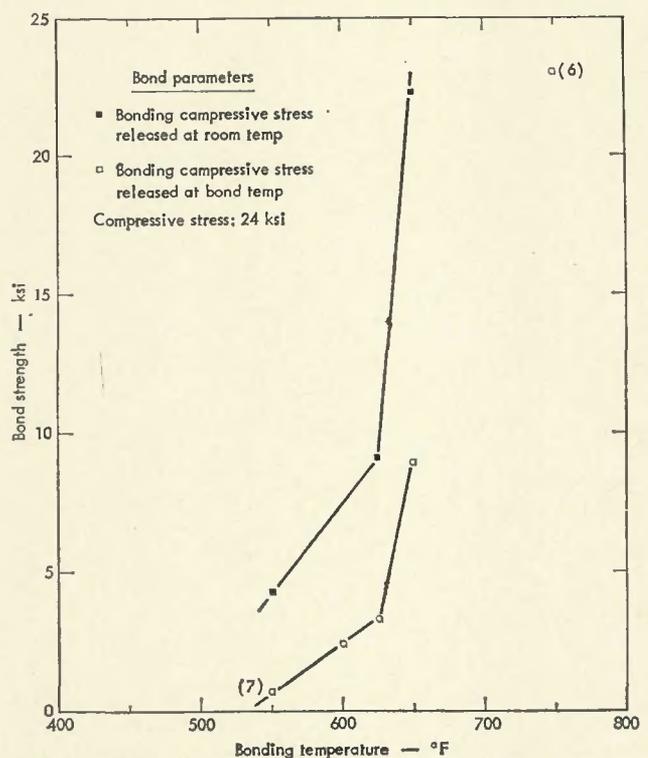


Fig. 10—Effect of bonding temperature for as-plated specimens of beryllium

imens for which the bonding compressive stress was maintained until the specimens reached room temperature.

The bond strength of various silver-plated substrates linearly increase with the modulus of elasticity of the substrate at a rate independent of the bonding temperature—Fig. 11. Data from Moffatt and Wulff<sup>12</sup> on lead-brazed joints in steel and molybdenum show a similar increase with substrate modulus.

#### Effect of Bonding Environment and Silver-Surface Conditions

This study was begun under the assumption that bonds made in vacuum would be superior to those made in air. This assumption proved to be invalid, as shown in Fig. 9. Bonding in other extreme environments, such as helium and oxygen, has no effect on the bond strength.

Two specimens were polished with the standard aluminum oxide but the lapping oil normally used was replaced by distilled water. The bond

strength expected for the selected bond parameters, assuming a standard polishing, was 7.5 ksi, and the actual bond strength was 0.0 and 2.5 ksi. The latter specimen was repolished with the standard lapping oil and rebonded. This bond strength was 9.3 ksi.

Five standard specimens were heated to their bond temperatures, held for periods ranging from 2 to 60 min. without a compressive load being applied, and then bonded. Two of these specimens were bonded in vacuum. All results indicate no significant effect from this bake-out cycle.

#### Plastic Strain Induced by Bonding Process

One of the major objectives of this study was to develop a low-temperature bonding process that would result in little if any permanent deformation of the parts being bonded. The diameters of all specimens were measured adjacent to the bond interface before and after bonding, and the plastic strain was computed as twice the change in diameter divided

by the original diameter. For all bonds reported herein on copper, stainless steel, molybdenum, and tungsten, no permanent upset was obtained.

The plastic strain or upset in beryllium is shown in Fig. 12 for various bond temperatures as a function of the bonding compressive load. Referring again to Fig. 7 and the bond strengths obtained for the 225° F bonds, a correlation between plastic strain and bond strength can be observed. The rapid increase in bond strength with increase in bonding compressive stress above 27 ksi is attributed to a corresponding increase in bond area resulting from plastic deformation.

#### Data from Necked Specimens

Results presented thus far have been aimed toward an understanding of the bonding mechanism and the relationship of bonding parameters to bond strength. The present study was concluded with the bonding of specimens under relatively optimum conditions, the necking of the specimens to remove the built-in notch (or stress riser) as shown in Figs. 2 and 5, and the determinations of the ultimate strength of the bond. The results are shown in Table 4.

#### Metallography and Related Examinations

An electron microprobe analysis of the silver-beryllium interface did not reveal any diffusion of silver into beryllium. The structure of the as-plated silver as determined by X-ray diffraction was randomly oriented polycrystalline. Metallographic examination revealed that the silver had a columnar structure; the grains were oriented perpendicular to the bonding surface and had a diameter of about 2 μ as viewed on the bond interface—Fig. 13.

The polishing procedure used to prepare the silver surfaces before bonding made the surfaces highly reflective, decreased the surface roughness, and introduced some amount of cold work into the surface as evidenced by smearing that occurred at the intersection of polishing lines. The polishing lines resulted from the 0.3μ aluminum oxide used. The observations concerning roughness and smearing were made with a scanning electron microscope.

Bonded specimens, metallographically processed to view a plane perpendicular to the bond interface, were etched to reveal the silver grain structure. A beryllium specimen, plated with 500 μin. per specimen half, bonded at 225° F, and fractured 100% in the beryllium at 44 ksi, is shown in Fig. 14. The columnar grain structure of the silver is evident and

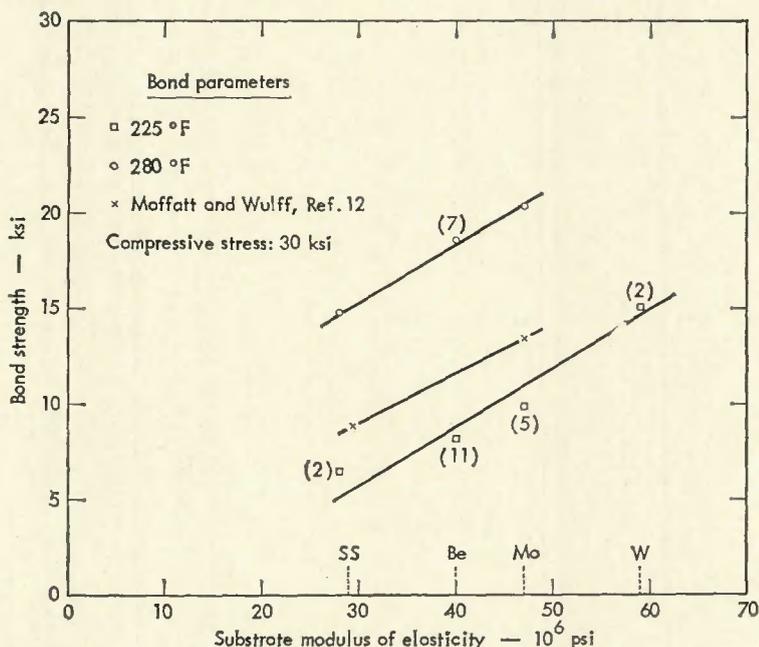


Fig. 11—Effect of substrate modulus of elasticity

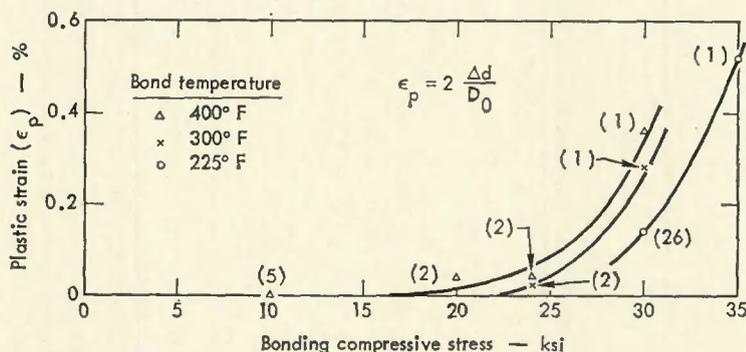


Fig. 12—Plastic strain induced in beryllium by bonding cycle as related to bonding compressive stress

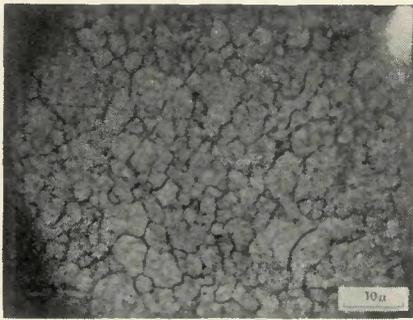


Fig. 13—Polished and etched plated silver (reduced 58% in reproduction)

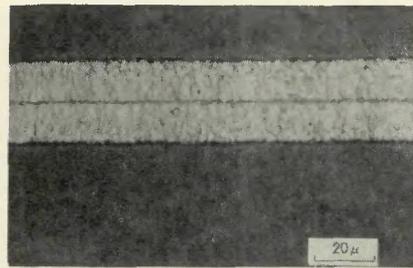


Fig. 14—Photomicrograph of low-temperature bonded section after standard bond cycle (thick silver plate)—(reduced 50% in reproduction)

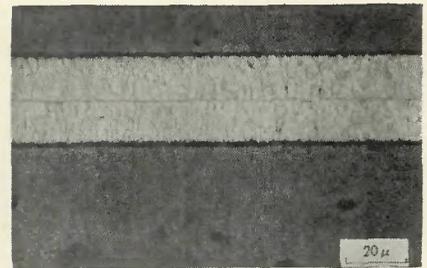


Fig. 15—Photomicrograph of low-temperature bonded section, vacuum-annealed at 750° F, 1 hr (reduced 50% in reproduction)

the silver-silver interface is severely etched. This specimen was reheated to 750° F in a vacuum for 1 hr without a compressive load. This temperature (750° F) is well above the recrystallization temperature of cold-worked silver. Metallographically reprocessed and etched twice as long as previously, the specimen did not etch as severely at the interface, and careful examination revealed some grain growth across the interface—Fig. 15.

A specimen bonded at 255° F and polished at a shallow angle to the bond interface surface also revealed preferential etching at the polished interface. The wide-etched bond zone is shown in Fig. 16. Changing the focus of the microscope revealed the preferential etching, and a sketch below the figure reveals the approximate surface profile. The heavy line shows the surface contour of the viewed surface and reveals why the plate thicknesses appear to be different.

Careful microscopic examination of all silver-silver fracture surfaces did not reveal any evidence of transfer of silver from one surface to the other.

## Discussion of Results

### Bonding Mechanism

The term *cohesion*, or *adhesion* for dissimilar metal couples, is generally used to explain the formation of a solid state bond that cannot be attributed to another possible bonding mechanism such as diffusion or recrystallization. A cohesive bond is generally assumed to be derived from atomic forces. This bond is achieved by bringing the two surfaces to be bonded into such intimate contact that the force fields of the atoms on both sides of the interface interact. If either or both of the surfaces are contaminated, the distance separating the atoms may be too large to permit the interaction and a bond will not be achieved unless the contaminating barrier can be invaded. The force required to separate the interfaces will be related to the total area of intimate contacts developed. If it can be shown that the bonding mechanism is not diffusion or recrystallization—the other recognized possible bonding mechanisms—it can be concluded that the bonding studied herein is cohesion.

The results clearly indicate that the bond strength is not a function of bonding time. Both diffusion and recrystallization are time-dependent. The bond temperatures, 225 to 280° F, for which most of the data are obtained are considerably less than the temperature generally associated with diffusion-activated phenomena (0.5  $T_m$  or 650° F for silver) and less than the recrystallization temperature of silver (392° F).

Photomicrographs of sections through the bond interface show a more rapid attack of the silver-etching reagent at the bond interface than elsewhere in the silver material. One specimen which was reheated to above the recrystallization temperature and re-etched revealed a more

uniform etch and some grain growth across the interface. Since the polishing procedure used on the interface prior to bonding introduces a cold worked layer and since strain-hardened materials are generally more susceptible to attack by acids, these results provide further evidence that recrystallization does not occur at the bond interface during the low-temperature bonding cycle.

The as-plated bonds required temperatures near 0.5  $T_m$  for silver before a bond is formed, and in these cases the bonding mechanism is suspected to be diffusion. A surface contamination barrier is believed to exist and, with the high thermal energy supplied, the barrier is overcome by diffusion of silver atoms through this barrier or diffusion of the contaminant into the silver. When this contaminating layer is removed by mechanical polishing, bonds can be achieved

Table 4—Bond Strengths for Some Necked Specimens

Material	Ag thickness, <sup>a</sup> ×10 <sup>-4</sup> in.	Temperature °F	Compression, ksi	Bond strength, ksi	Fracture location <sup>b</sup>	Plastic strain, <sup>c</sup> %
Mo	1.9	550	30	58.1	P	
W	2.2	700	30	31.4	P <sup>d</sup>	
	3.2	400	30	22.9	P	
	3.0	550	30	11.9	P	
Cu	3.1	700	30	42.2	P	
	2.6	280	10	40.3	S	
	2.4	400	10	41.5 <sup>e</sup>	S	
SS	2.6	400	10	42.3 <sup>f</sup>	S	
	4.7	400	30	49.5	P	
	4.2	700	30	54.8	P	
Be	1.8	225	35	18.2	Ag	0.5
	5.2	225	30	44.0	S	0.2
	0.4	280	30	15.5	Ag	0.4
	2.2	280	30	30.5	S + 5% Ag	0.3
	2.5	280	30	41.5	S	0.2
	1.8	350	25	40.4	S	0.1
	0.3	400	20	16.4	Ag	
	0.5	400	30	25.1	P + 30% Ag	0.4
	2.0	400	20	34.1	S + Ag <sup>g</sup>	
5.4	500	10	37.5	S + 5% Ag		

<sup>a</sup> Plate thickness per specimen half before polishing.

<sup>b</sup> P—plate-peeled; S—substrate; Ag—Ag-Ag.

<sup>c</sup> Plastic strain was zero unless otherwise noted.

<sup>d</sup> This specimen was back-heated prior to plating to 740° C; the other specimen was heated to 775° C.

<sup>e</sup> Bond time: 60 min.

<sup>f</sup> Bond time: 0.5 min.

<sup>g</sup> Ag fracture surface was evident only along unbonded regions of beryllium surface scratches.

at much lower temperatures. It might be argued, however, that the polishing introduces a high state of strain at the bond interface which, upon bonding, is somewhat released by diffusion or recrystallization at the interface on such a microscopic scale that it cannot be detected by the available techniques.

This argument can be refuted by the results of two different bond techniques used at the low temperatures. First, a thin layer of silver was removed from the bond interface surface by acid etching. Thus the contaminating layer was removed, and the bond interface was not strain-hardened. A 300° F bond agreed closely with the results obtained on polished specimens. Second, several polished specimens, preheated to their bonding temperature in air and in vacuum for various periods of time while separated, showed no degradation in bond strength. If recrystallization is the bonding mechanism, the preheat cycle should have caused recrystallization, and a lower bond should have ultimately resulted. Thus, it is concluded that the surface preparation after plating serves only to remove a contaminating film from the silver surface.

#### Plating Thickness

The experimental data presented reveals that the bond strength increases with increasing plate thickness—Fig. 8. Two questions regarding the data should be addressed:

1. Why does the bond strength in-

crease with increasing  $t/D$  ratios?

2. Can the data be extrapolated to large  $t/D$  ratios?

Many investigators have shown that braze and adhesive bonds increase in strength with decreasing  $t/D$  ratios as long as the bond formation is void-free. The triaxial state of stress responsible for the increase in bond strengths applies equally to this study; however, a void-free bond cannot be assumed. In fact, the bond formation which depends upon intimate contact at the silver interfaces is impeded by lower  $t/D$  ratios, i.e., thinner silver plates. For thin plates the stress at the interface will be triaxial compression for an applied bonding compressive load, because the yield stress of the plate is less than the substrate. For thicker plates the radial compressive stresses are less, thus permitting more plastic deformation and therefore increasing the contact area. It is concluded, therefore, that the increase in bond strength with increase in  $t/D$  results from increased area of contact.

There must be a limiting value of  $t/D$  beyond which either full bond formation has been achieved, or the decrease in bond strength resulting from the increasing  $t/D$  ratio becomes more significant than the increasing bond formation. This relationship is shown in Fig. 17. The region A-B represents the  $t/D$  ratios tested. The bond strength increases to point C beyond which the bond strength decreases to the limiting value shown at D. This limiting strength represents

the ultimate uniaxial tensile strength of the plated material. An upper limit might also exist depending upon the bonding parameters and the substrate material. In this event the curve would be truncated by a horizontal line. For the silver-beryllium combination, plating techniques were developed that produced a plate adhesion that exceeded the bulk strength of beryllium; thus this general curve would be truncated at the ultimate strength of beryllium, 40 ksi. For the silver-stainless steel combination, the plating technique resulted in a plate adhesion strength of approximately 55 ksi, thus truncating the curve at this stress level.

#### Bond Strength as Related to Stress and Strain

The bond strength for unnecked specimens is linearly related to the bonding compressive stress over the range tested. For the lowest bond temperature used (225° F), the bond strength was found to increase at a greater rate when the compressive stress was sufficient to cause a plastic strain in the beryllium substrate. The increase in area of contact resulting from increased compressive loads would account for the increase in bond strength. The results of extrapolating the curves in Fig. 7 to zero bonding compressive stress indicate some significant bond strength,  $\sigma_0$ . One would not expect a bond to form without the application of a bonding compressive load necessary to bring the surfaces into intimate con-

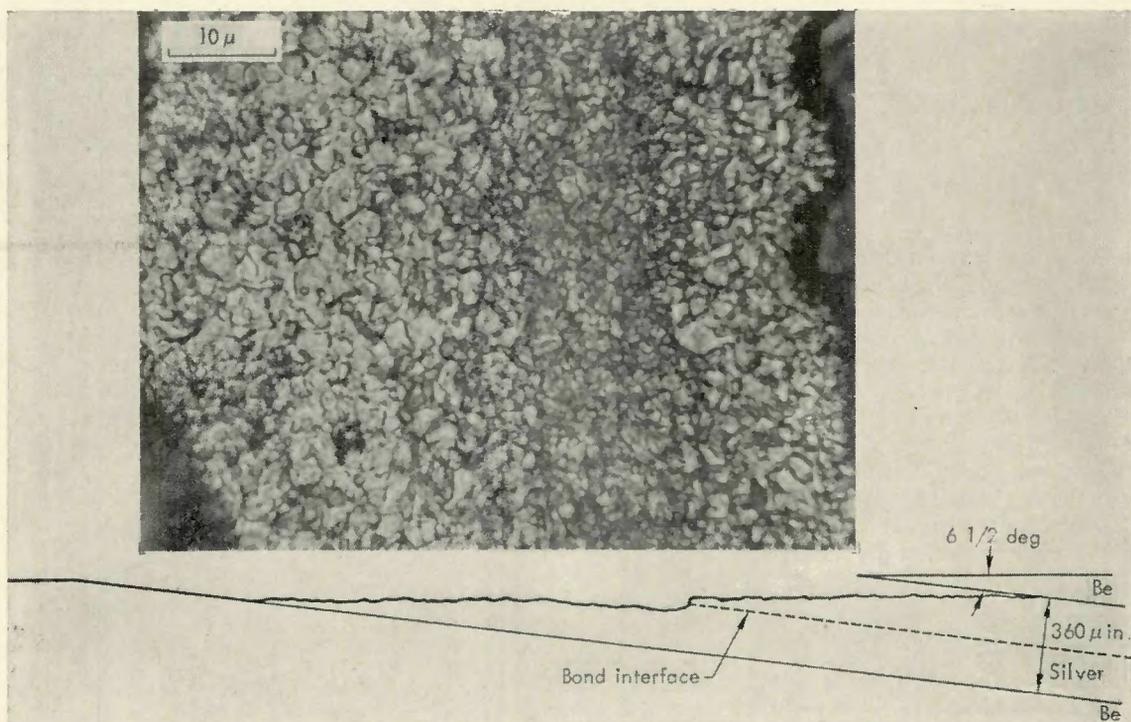


Fig. 16—Photomicrograph of low-temperature bonded section viewed 6½ deg from plane of bond interface (reduced 28% in reproduction)

tact. This condition probably results from the fact that bond formation was accomplished at some temperature,  $T_B$ , while the bond destruction or bond-strength determination was conducted at room temperature,  $T_0$ . If we assume that the bond area remains constant at  $T_B$  and  $T_0$ , then the bond strength at  $T_0$  should be greater than at  $T_B$  because there is a difference in the strength of silver at these temperatures. Thus  $\sigma_0$  represents the increase in strength resulting from the difference in bond and test temperatures.

The bond and test procedures differed in a second way, however. The bonding compressive stress was applied at the bond temperature  $T_B$ , and released at room temperature,  $T_0$ . The results, shown in Fig. 10, reveal that by releasing the compressive load at  $T_B$  rather than at  $T_0$ , a significant decrease in bond strength is obtained. This fact may be explained by assuming that the stored strain energy,  $\int F d\epsilon$ , fractures some of the bond when released. Assuming that the strain energy released is not a function of the specimen temperature when the load is released, and since the strength of silver is less at  $T_B$  than at  $T_0$ , we see with the aid of Fig. 18 that more of the bond is destroyed by the load at  $T_B$  than at  $T_0$ . Thus a decrease in bond strength measured at  $T_0$  is expected by releasing the load at  $T_B$  rather than at  $T_0$ .

Further insight can be gained by considering the results obtained for low-temperature bonds on materials with varying moduli of elasticity—Fig. 11. The stored elastic-strain energy in the substrate material decreases with increasing modulus for a fixed compressive stress. Thus, if we focus our attention only on the substrate stored elastic energy, we would expect a greater bond strength for materials with higher moduli of elasticity. The analysis is not so simple, however. The triaxial state of stress in the silver is the result of differences in yield strengths and moduli of elasticity of the plate and substrate. Assume that the area of contact and the degree of bonding for some temperature, plate thickness, and bonding compressive stress are independent of these mechanical parameters. Then the tensile radial stresses in the silver during the tensile test would be greater for the more rigid (i.e., greater modulus of elasticity) substrate material. It follows that the measured bond strength would be greater the higher the modulus of elasticity. Thus, the observed increase in bond strength with increasing substrate modulus of elasticity can be explained by the decrease in stored elastic strain energy

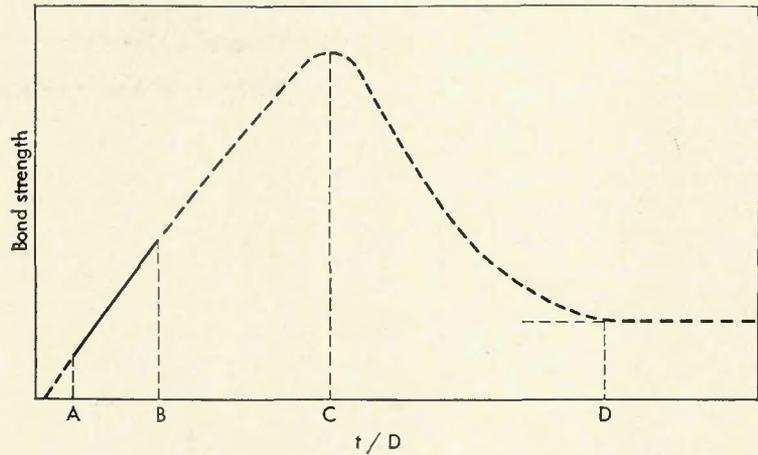


Fig. 17—Predicted bond strength of specimens vs. thickness-to-diameter ratio

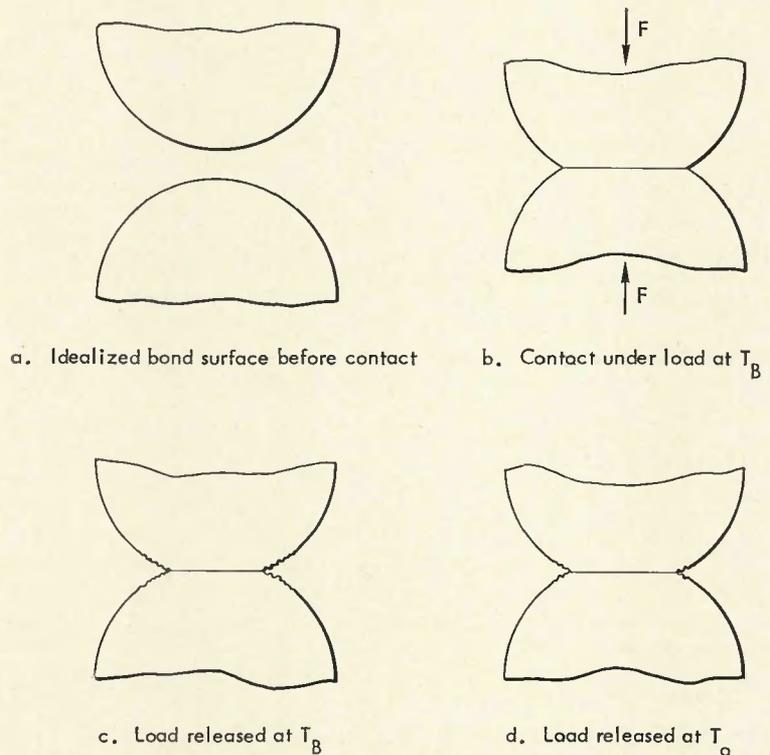


Fig. 18—Effect of released bonding compressive load for idealized contact

and/or the increase in the radial stresses within the silver.

Also shown in Fig. 11 are the results from Moffatt and Wulff<sup>12</sup> of lead braze joints in steel and arc-cast molybdenum. The  $t/D$  ratios were between 0.0005 and 0.001, and the molybdenum bond contained a "few small nonwetted regions." The slope of the line agrees well with the data obtained during this study ( $t/D = 0.0008$ ). The braze joints were not loaded in compression and, therefore, did not contain any stored elastic-strain energy except for that introduced by thermal contraction. It is concluded that thin joint strengths linearly increase with increasing modulus of elasticity of the substrate. The

change in bond strength per change in modulus is a constant not dependent upon the filler metal (lead or silver) or upon the bond parameters (braze or solid state bonds at different bond temperatures) for a given  $t/D$  ratio. The increase in bond strength with substrate modulus is primarily attributed to the increase in radial stresses in the plate or braze material and secondarily to the reduction of the stored elastic energy.

### Summary

A technique considered feasible for production has been developed for joining beryllium in air at temperatures of 225 to 280° F. The results

are bond strengths exceeding the strength of beryllium (type S200C). The interfaces to be joined are polished flat, vacuum evaporation-plated with silver, polished with aluminum oxide and lapping oil to remove contaminants from the silver bond surface, cleaned with acetone, and bonded in air at the required temperature and compressive load. This bonding procedure, resulting in room-temperature bonds which exhibit beryllium fracture strengths, develops an upset in the parts of 0.2% strain or less.

The plating system consisted of a silicone oil diffusion pump. To assure good adhesion of silver to beryllium it was found necessary to plate the beryllium while heated to at least 1100° F and to maintain the vacuum system free of silicone-oil vapors. Neither requirement would prevent the use of this method on a production line, but the vacuum system must be either monitored for silicone oil or designed to eliminate this source of contamination.

The bond strength for beryllium specimens was sensitive to the radial alignment and interface flatness. The radial alignment is not a problem peculiar to this bonding process and is not considered to be any more restrictive than any other presently used bonding technique. Thus, the loss in bond strength resulting from the notch effect due to this misalignment is a common problem and does not reduce the potential of this process. Specimens were polished to a flatness of 20  $\mu$ in. across the 0.500 in. diameter end. After the silver was hand-polished on a thick-nap cloth, the specimens were flat to within 30  $\mu$ in. except at the extreme outside diameter, which was rounded off severely as shown in Fig. 5. This built-in notch caused stress concentrations, and the measured bond strength was less than that achieved by machining away the notch, i.e., necked specimens. The rounded-off silver edge can be minimized by using a polishing cloth, such as silk or nylon, which has no nap. This procedural change is recommended for production application.

The bonding environments and surface cleanliness are generally consid-

ered to be very important considerations for successful solid state bonding. The results of this study show that the bonding environment is not important (air, oxygen, helium, or vacuum). The surface preparation is indeed critical, however. Polishing the surfaces with aluminum oxide and water resulted in much lower bond strengths than was the case after polishing with aluminum oxide and lapping oil. Polished surfaces were cleaned with acetone; extended exposure to acetone reduced the bond strengths. Methods different from those used in this study for polishing and cleaning may reduce even more the required bond temperature.

Although most of the results were with beryllium bonded specimens, copper, stainless steel, molybdenum, and tungsten specimens were successfully bonded to themselves. Substrate fracture strengths were obtained on beryllium and copper while plate-substrate failures limited the bond strengths of the remaining metals. The only problem in applying this technique to other materials appears to be in achieving good plate-substrate adhesion. Although no attempt was made to join dissimilar metal pair substrates, no difficulties can be foreseen.

## Conclusions

1. The bonding mechanism is one of cohesion for the polished silver-plated specimens and is probably a diffusion mechanism for the as-plated specimens.

2. Results indicate that the bond strength:

- (a) Is independent of bonding time.
- (b) Linearly increases with the compressive bonding stress (for elastic substrate stresses).
- (c) Linearly increases with plating thickness (35 to 600  $\mu$ in.).
- (d) Linearly increases with bonding temperature (225 to 280° F).
- (e) Linearly increases with substrate modulus of elasticity.
- (f) Is greater at room temperature if the bonding compressive stress is maintained until the

specimen has cooled to room temperature.

- (g) Is not sensitive to the bond environment (oxygen, air, helium, vacuum of  $10^{-6}$  torr).
- (h) Is not influenced by baking-out at the bond temperature before compressively loading.
- (i) Decreases if the polishing fluid is water rather than lapping oil.
- (j) Decreases with extended exposure to acetone.
- (k) Is not influenced by 1½ hr air exposures after polishing and cleaning.

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