

Comparing Metal-Ceramic Brazing Methods

The advantages and disadvantages of the various methods for joining metals to ceramics are outlined

BY C. A. WALKER AND V. C. HODGES

Designers and engineers have many options to choose from when considering how to join metals to non-metals for structural, electrical, and packaging applications. These options could include mechanical means of fastening such as screws, bolts, rivets and other fasteners, or an elevated-temperature means such as soldering or brazing. Metal-ceramic brazing, the topic of this article, is particularly useful for fabricating high-reliability devices such as those used in high-voltage applications or requiring hermetically sealed joints. This article is intended to familiarize the designer with brazing methods commonly used to join metals to ceramics, discuss the advantages and disadvantages of each method, and show the relative tensile strengths obtained from samples fabricated using these methods. Alumina is one of the most commonly used engineering ceramic materials, offering high hardness and wear resistance with excellent electrical insulation properties. Alumina ceramic is commonly available in purities ranging from 88 to 99.9%, with high-temperature glasses making up the balance of the composition. For most cases discussed, 94% alumina ceramic (6% glassy phase) ASTM-F19 tensile button samples were joined to Fe-29Ni-17Co alloy using a gold-or silver-based braze filler metal. The versatile design of the ASTM-F19 tensile specimen allows a helium mass-spectrometer leak detection test to be performed prior to tensile testing (Ref. 1).

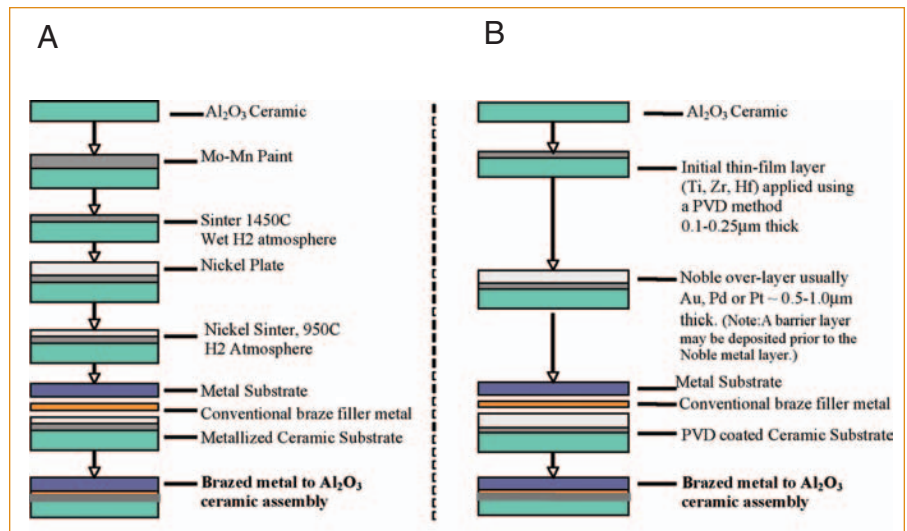


Fig. 1 — Commonly used ceramic metallization methods. A — Moly-manganese metalization process; B — thin-film metallization process.

Metal-to-ceramic brazing can be accomplished by first applying a metallic layer onto the ceramic surface or by brazing directly to the unmodified ceramic (oxide) surface. Several metallization methods have been proven to work effectively; however, this article is limited to the two metallization methods most commonly used (Refs. 2-4) for joining metals to ceramics: the molybdenum-manganese/nickel plating method and physical-vapor deposition or thin-film method.

Molybdenum-Manganese/Nickel Plating Method

The molybdenum-manganese/nickel plating method, also known as moly-manganese metallization, is performed as follows: A coating of molybdenum and manganese particles mixed with glass additives and volatile carriers is applied to the ceramic surface to be brazed — Fig. 1A.

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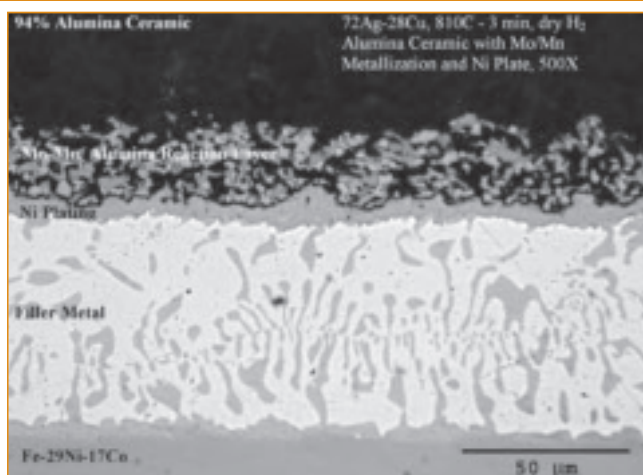


Fig. 2 — Ceramic-to-metal braze using molybdenum-manganese/nickel plate metallization.

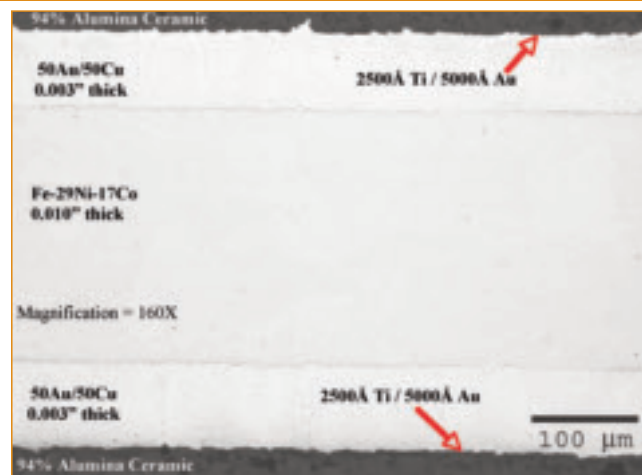


Fig. 3 — Au/Cu brazed metal-to-ceramic sample made using thin-film metallization.

Table 1 — Moly-Manganese/Nickel Plate ASTM-F19 Tensile Button Test Results

Filler Metal	Nonmetal Substrate	Metal Substrate	Brazing Temperature/Time	Furnace Atmosphere	Average Tensile Strength ^(a)
65 Cu/35 Au	94% Alumina	Fe-29Ni-17Co	1040°C/3 min	Dry Hydrogen	14.5 ksi/100 MPa
50 Au/50 Cu	94% Alumina	Fe-29Ni-17Co	1000°C/3 min	Dry Hydrogen	17 ksi/118 MPa
72 Ag-28 Cu	94% Alumina	Fe-29Ni-17Co	810°C/3 min	Dry Hydrogen	14.3 ksi/99 MPa
77 Au-13Ag-10 Ge	94% Alumina	Fe-29Ni-17Co	495°C/3 min	Dry Hydrogen	15.6 ksi/108 MPa
77 Au-13 Ag-10 Ge (2x thk)	94% Alumina	Fe-29Ni-17Co	455°C/5 min	Dry Hydrogen	16.1 ksi/111 MPa

(a) Tensile strength averages are ± 2 ksi/14 MPa.

The application of the coating may be hand-painted, sprayed, or robotically applied. After air drying, the coating is fired in a wet hydrogen environment (15°–30°C dew point) at 1450°–1600°C leaving a “glassy” metallic coating 300–500 micro-inches (7.6–12.7 microns) thick. The fired coating is subsequently plated with a 0.001–0.003 in. (25.4–76.2 microns) layer of nickel. The nickel plating is sinter-fired at 850°–950°C in a dry hydrogen (–50°C dew point or less) atmosphere leaving a finished metallic surface that can be readily brazed using standard braze filler metals.

Some of the advantages of the molybdenum-manganese/nickel plating method are as follows:

1) Having been developed in the 1930s (Ref. 2), moly-manganese metallization is a mature technology with a proven history of success;

2) Postmetallization, ceramic materials can be easily brazed using standard braze filler metals; and

3) Commercial suppliers are available

to provide the necessary metallization component materials or metallization services.

The molybdenum-manganese/nickel plating method also has several disadvantages. Included in these are the following:

1) **Expense.** Specialized high-temperature furnaces and plating equipment are necessary — Fig. 1.

2) **Lengthy time requirements.** Multiple high-temperature furnace operations are required as well as the care and maintenance of plating baths.

3) **Rework limitations.** Excessive nickel depletion into the braze filler metal can lead to poor braze joint performance.

4) **Geometric constraints.** Large sizes and thick cross sections are difficult to process.

5) **Batch size.** Process development for small quantities is often cost prohibitive.

Table 1 shows the average strengths typically obtained [14–17 ksi (99–117 MPa)] when using various gold- and silver-based brazing filler metals to braze 94% alumina ceramic to Fe-29Ni-17Co alloy, a

controlled expansion alloy often used when brazing to ceramics. All of the brazed samples shown in Table 1, as well as those shown in the subsequent tables (Tables 2–4) passed a helium mass-spectrometer leak detection test (leak rate $< 2.0 \cdot 10^{-9}$ atm-cc/s) prior to being tensile tested. The crosshead speed used for the tensile tests was $3.3 \cdot 10^{-4}$ in./s ($8.38 \cdot 10^{-6}$ m/s). The tensile strengths shown in the tables are averages of samples tested. Variations of ± 2 ksi (14 MPa) from the average tensile strengths were observed. Formulations of brazing filler metals are displayed in wt-%.

A scanning electron microscope (SEM) image of a cross-sectioned brazed metal-ceramic assembly, utilizing moly-manganese metallization and nickel plating is shown in Fig. 2. The ceramic is 94% alumina, and the metal member is Fe-29Ni-17Co. Notice the 25–35- μ m-thick reaction zone where the moly-manganese metallization diffuses and reacts with the glassy phases of the alumina ceramic. The clearly defined nickel plating layer shown has been sufficiently wetted by the braz-

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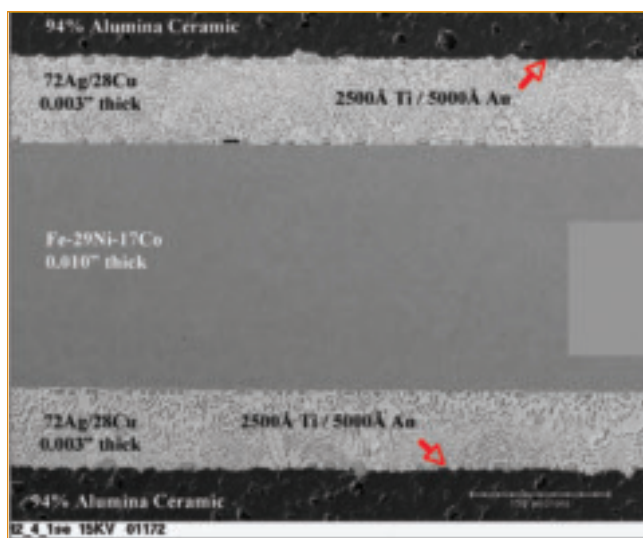


Fig. 4 — Ag/Cu brazed metal-to-ceramic sample made using thin-film metallization.

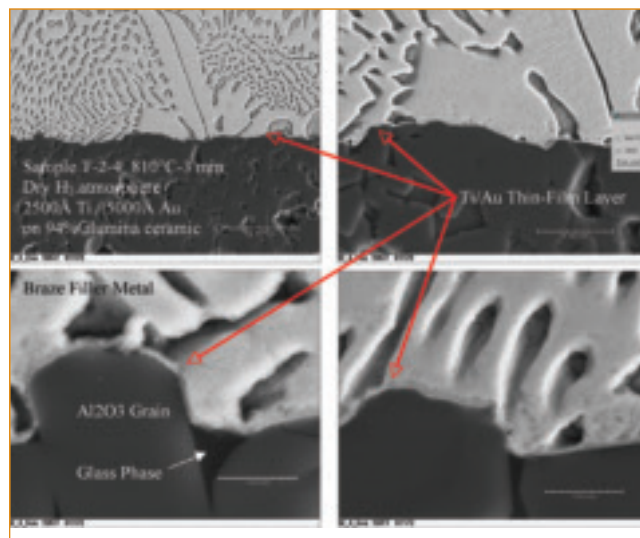


Fig. 5 — Ti/Au thin-film deposition layer on Ag/Cu brazed metal-ceramic sample.

Table 2 — Thin-Film Metallization ASTM-F19 Tensile Button Test Results

Filler Metal	Substrates	Thin Films	Brazing Temperature/Time	Furnace Atmosphere	Average Tensile Strength ^(a)
50 Au/50 Cu	94% Alumina Fe-29Ni-17Co	0.25 μm Ti/ 0.5 μm Au	1000°C/3 min	Dry Hydrogen	15.1 ksi/102 MPa
50 Au/50 Cu	94% Alumina Fe-29Ni-17Co	0.25 μm Ti/ 0.5 μm Au	1020°C/10 min	Dry Hydrogen	12.9 ksi/89 MPa
50 Au/50 Cu	94% Alumina Fe-29Ni-17Co	0.25 μm Ti/ 0.02 μm Pd/ 0.5 μm Au	1000°C/3 min	Dry Hydrogen	16.1 ksi/111 MPa
50 Au/50 Cu	94% Alumina Fe-29Ni-17Co	0.25 μm Ti/ 0.02 μm Pd/ 0.5 μm Au	1020°C/10 min	Dry Hydrogen	11.8 ksi/81 MPa
72 Ag-28 Cu	94% Alumina Fe-29Ni-17Co	0.25 μm Ti 0.5 μm Au	810°C/3 min	Dry Hydrogen	13.0 ksi/90 MPa
63 Ag-27 Cu-10 In	94 % Alumina Fe-29Ni-17Co	0.25 μm Ti/ 0.5 μm Pt	755°C/2 min	UHP Argon	13.2 ksi/91 MPa
63 Ag-27 Cu-10 In	951 LTCC Fe-29Ni-17Co	0.25 μm Ti/ 0.5 μm Pt	755°C/2 min	UHP Argon	8.2 ksi/57 MPa
63 Ag-27 Cu-10 In	951 LTCC Fe-29Ni-17Co	0.5 μm Ti/ 0.5 μm Au	755°C/5 min	UHP Argon	6.5 ksi/45 MPa
63 Ag-27 Cu-10 In	951 LTCC Fe-29Ni-17Co	0.5 μm Ti/ 0.5 μm Pd	755°C/5 min	UHP Argon	3.8 ksi/26 MPa

(a) Tensile strength averages are ± 2 ksi/14 MPa.

ing filler metal to provide high joint strength and hermeticity. The light and dark areas within the brazed joint are the silver-rich and copper-rich regions.

Thin-Film Deposition

Depicted in Fig. 1B, thin-film deposition is another commonly used (Refs. 2, 3) method to apply a metallization layer

to a ceramic substrate so that it may be joined using conventional braze filler metals. A combination of materials, usually two or three, are deposited onto the nonmetallic surface using a physical vapor deposition (PVD) method such as evaporation or sputtering. The first layer deposited, often titanium, is typically 0.05–0.25 μm thick. Other strong oxide-forming elements such as hafnium, zirconium, chromium, niobium, etc. may be

chosen depending on the application and service temperature. Occasionally, an intermediate layer or layers are deposited to prevent unwanted metallurgical reactions between the initial metal layer and the braze filler metal. The top, or outer, layer is normally a noble metal such as gold, platinum, or palladium that is 0.25–1.0 μm thick. A noble metal is cho-

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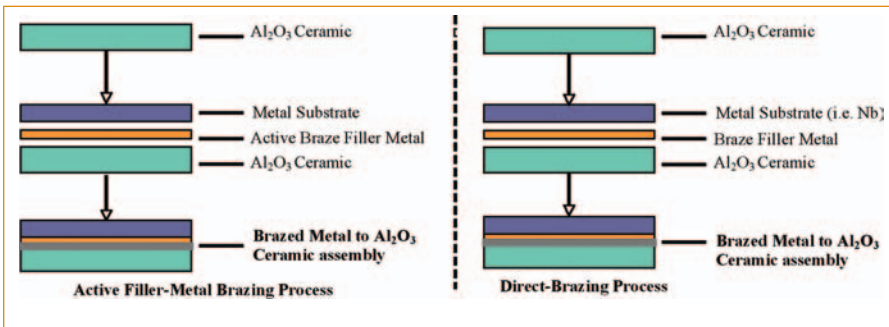


Fig. 6 — Direct metal-to-ceramic brazing processes.

sen in order to prevent the underlying layer from oxidizing and subsequently preventing proper braze filler metal wetting and flow. Detailed brazing-related concerns when using thin-film metallization coatings for ceramic assemblies have been published (Ref. 3).

The following are a few of the advantages for using thin-film metallization coatings:

- They have a proven brazing practice history and are forgiving when used with standard filler metals.
- Versatility. A wide range of metal choices exist for the engineer or designer that can be deposited to address special applications or environments.
- Another important advantage is the speed, which can often be less than a

few hours total, that simple geometries can be prepared for brazing.

The primary disadvantages of thin-film metallization coatings are as follows:

- Specialized equipment is required to apply the coatings.
- Intricate masking may become necessary to prevent the deposition of metal in unwanted locations.
- Ceramic geometric constraints, which may prohibit the proper positioning of the ceramic member or hinder the application of uniform coating thicknesses, of most thin-film deposition chambers.

Tensile button strengths obtained using various thin-film metallization schemes are shown in Table 2. Captured in Table 2 (compare lines 1 and 2, then lines 3 and 4) is the increased size of the

brazing process window (peak temperature and time ranges) that can be obtained without the drastic decline in tensile strength usually witnessed when using the moly-manganese metallization method. This is because the thin-film metallization method, in contrast to moly-manganese metallization, does not use nickel plating, which readily dissolves into the braze filler metal at higher temperatures and longer peak soak times. Also shown in Table 2 are the tensile strengths of brazed Fe-29Ni-17Co tensile buttons to Low-Temperature Co-Fired Ceramic (LTCC) interlayers. When using a 63Ag-27Cu-10In braze filler metal, the tensile strengths varied by a factor of two, depending on which thin-film metallization scheme was chosen (Ref. 5). This strength loss is due to the formation of brittle intermetallic compounds within the braze joints or at the braze joint interfaces.

Shown in Fig. 3 is an SEM image of a cross-sectioned ceramic-metal-ceramic brazed sample utilizing a thin-film metallization scheme of 2500 Å (0.25 μm) titanium and 5000 Å (0.50 μm) gold. A 50Au-50Cu brazing filler metal was utilized for the joining operation. The same substrate materials and geometry were joined with a silver-based braze filler metal, 72 Ag-28Cu, and shown in Fig. 4. In both SEM images, the samples exhibit excellent wetting and flow onto the irregular alumina ceramic surface with little or no base

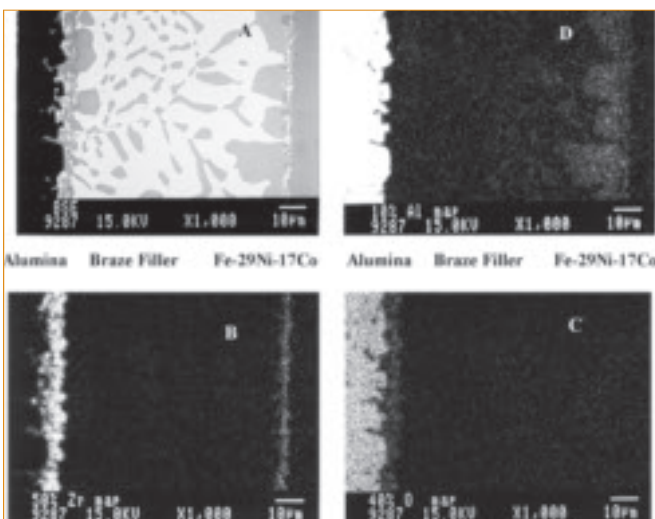


Fig. 7 — SEM backscattered image (BSE) with energy-dispersive spectroscopy (EDS) maps showing postbrazed zirconium, oxygen, and aluminum (counterclockwise from upper left) concentrations in an active brazed specimen.

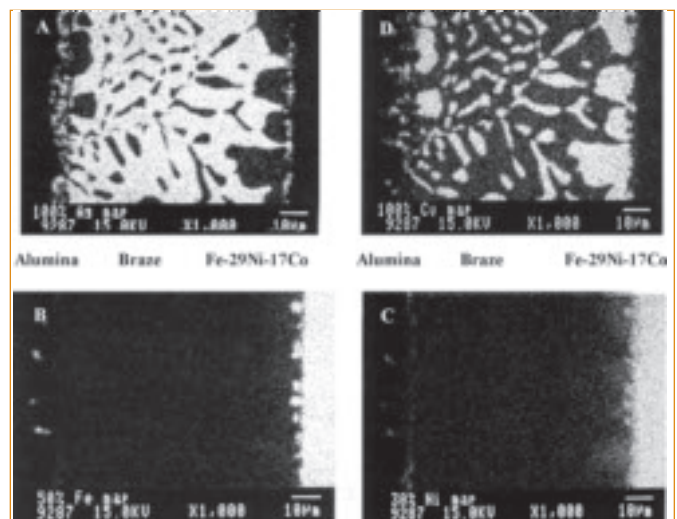


Fig. 8 — Energy-dispersive spectroscopy maps showing postbrazed silver, iron, nickel, and copper concentrations (counterclockwise from upper left) in an active brazed specimen.

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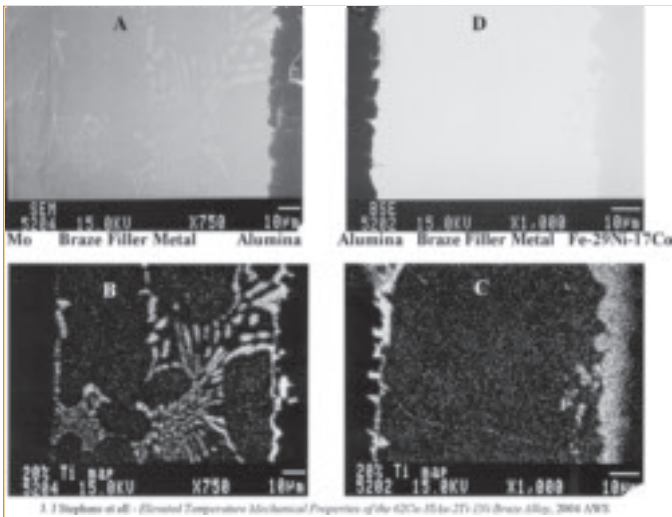


Fig. 9 — Active brazed molybdenum to 94% alumina ceramic sample (A and B) and active brazed Fe-29Ni-17Co to 94% alumina ceramic (C and D).

metal erosion. Figure 5 shows higher magnification images of the thin-film/ceramic interface from Fig. 4. Easily seen in these images is the continuous thin-film metallization layer along the alumina grain boundaries. The dark regions seen along the interface and between the alumina grains are the glassy phase of the 94% alumina ceramic.

Active Filler Metal Brazing

Active filler metal brazing is an area of high growth within the metal-ceramic brazing community. A primary reason for this growth is that unlike the moly-manganese metallization that is very material dependent, active filler metals display good wetting with most ceramic materials (Refs. 6, 7). Active filler metal brazing is a metal-ceramic joining method that permits the use of standard brazing techniques when making metal-to-ceramic brazements without the need to apply any metallization to the ceramic substrate. As shown in the left-hand portion of Fig. 6, the metal and nonmetal substrates are cleaned, and the active filler metal preform or paste (Ref. 8) is positioned or applied between the faying surfaces of the brazement. The brazing operation is usually performed in an inert or ultrahigh vacuum environment. For certain applications and component geometries, the transfer from a conventional brazing process to an active brazing process is accomplished quite readily. Many times,

however, the braze joint might require a redesign to accommodate the preplacement of brazing filler metal between the faying surfaces of the brazement. Capillary flow is inhibited by the bare oxide ceramic surface that exhibits limited spreading and flow of the brazing filler metal. High-vacuum or inert atmospheres are required because excessive oxygen in the atmosphere can react with the active element in the active braze filler metal and compromise joint strength and integrity (Refs. 9, 10).

Apart from these limitations, there are many advantages to using an active filler metal brazing process for certain brazing applications. These include the following:

- 1) The number of required steps to make metal-ceramic brazes are reduced and greatly simplified;
- 2) There are a variety of commercially available filler metal compositions for use in a wide range of processing temperatures and service conditions;
- 3) Specialized metallization equipment and the associated time-consuming metallization processes are eliminated.

There are, however, several disadvantages of using an active brazing process over a conventional metallization and

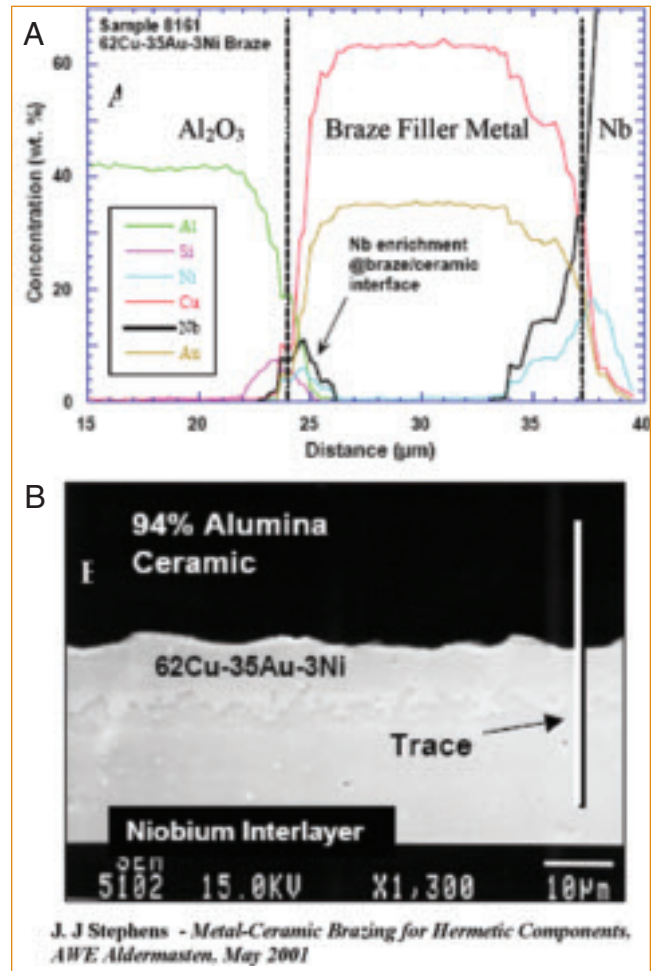


Fig. 10 — A — Electron microprobe analysis, and B — SEM image of a direct brazed niobium-94% alumina ceramic sample.

subsequent standard brazing process. The primary disadvantages are as follows:

- 1) Active brazing processes require more stringent atmospheric control;
- 2) Not all braze joint geometries are compatible with active brazing processes;
- 3) Processing equipment capable of adequate atmospheric control can be a limiting factor, placing size constraints on brazed assemblies.

Figures 7 and 8 illustrate the distribution of elements in an active braze filler metal following a brazing process. Figure 7A is a backscattered SEM image showing a portion of a 94% alumina ceramic that has been brazed using a 97Ag-1Cu-2Zr active braze filler metal. The sample was brazed at a temperature of 950°C, with a peak soak time of 5 min in a 12-torr ultrahigh-purity (UHP) argon partial pressure atmosphere. Figure 7B

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Table 3 — Active Filler Metal Brazed ASTM-F19 Tensile Button Test Results

Filler Metal	Nonmetal Substrate	Metal Substrate	Brazing Temperature/Time	Furnace Atmosphere	Average Tensile Strength ^(a)
62 Cu-35 Au-2Ti-1Ni	94% Alumina	Fe-29Ni-17Co	1006°–1026°C 6–8 min	Vacuum/Partial pressure Ar	11–14 ksi/76–97 MPa
97 Ag-1Cu-2Zr	94% Alumina	Fe-29Ni-17Co	990°C/5 min	UHV/Dry Hydrogen	15.4 ksi/106 MPa
97 Ag-1Cu-2Zr	94% Alumina	Fe-29Ni-17Co	963°C, 3 min above liquidus	Partial pressure Ar	21.3 ksi/147 MPa
63.00 Ag-35.24Cu-1.75Ti	94% Alumina	Fe-29Ni-17Co	1040°C/2 min	Dry Hydrogen	14.5 ksi/100 MPa
63.00 Ag-35.25Cu-1.75Ti	94% Alumina	Fe-29Ni-17Co	825°–1040°C/2–10 min	Partial Pressure Ar	11–14 ksi/76–97 MPa
63.00 Ag-35.25Cu-1.75Ti	94% Alumina	Fe-29Ni-17Co	825°–1040°C/2–10 min	Vacuum	11–16 ksi/76–110 MPa
59.00 Ag-27.25Cu-12.5In-1.25 Ti	94% Alumina	Fe-29Ni-17Co	755°C/5 min	Vacuum	14.5 ksi/99 MPa
59.00 Ag-27.25Cu-12.5In-1.25 Ti	DuPont 951 LTCC	Fe-29Ni-17Co	755°C/5 min	Vacuum	8 ksi/55 MPa

(a) Tensile strength averages are ± 2 ksi/14 MPa.

shows the migration of the elemental zirconium to the ceramic surface where it reacts with available oxygen and forms the layer that the primary filler metal element, silver, will wet and adhere to. A trace amount of zirconium can also be seen in the same image bound to the surface of the Fe-29Ni-17Co. Figure 7C shows a small concentration of oxygen that has dissolved into the zirconium-rich region of the solidified braze filler metal. Notice in Fig. 7D that a slight amount of aluminum from the ceramic material, having been replaced by zirconium, has diffused through the molten braze filler metal toward the Fe-29Ni-17Co surface. Figure 8 A–D are companion energy dispersive spectroscopy (EDS) maps that show the silver- and copper-rich phases of the resolidified brazing filler metal along with limited dissolved Fe-29Ni-17Co base metal.

The choice of the base metal substrate and active filler metal element can have a substantial impact on the end product as reported by Stephens et al. (Ref. 11), and shown in Fig. 9. A and B show a molybdenum substrate brazed to a 94% alumina ceramic using a gold-based active braze filler metal, 62Cu-35Au-2Ti-1Ni. C and D show the results when the molybdenum is replaced with Fe-29Ni-17Co. 9B and 9C are EDS maps showing the resulting titanium concentrations in the brazed samples. Figure 9B demonstrates that a

minimal amount of the active element, titanium, has reacted with the molybdenum allowing for the majority of the titanium metal to react with the ceramic substrate. Figure 9C reveals that a substantial portion of the titanium has reacted with the Fe-29Ni-17Co substrate to the point of causing some base metal erosion to occur and hindering the ability to make a hermetic seal. This scavenging of the titanium element can be prevented by coating the Fe-29Ni-17Co member with a barrier layer (Refs. 12–14). While some scavenging of titanium does occur, there is sufficient titanium in commercially available active brazing filler metals to make hermetic braze joints to Fe-29Ni-17Co substrates when careful attention is given to surface preparation, fixturing, atmosphere, and the brazing thermal cycle (Refs. 15, 16).

Tensile test results of tensile button samples made with gold- and silver-based active braze filler metals are displayed in Table 3. A comparison of the sample strengths in Tables 1 and 2 to those in Table 3 reveals that the results are very similar for tensile samples brazed with similar composition filler metal families and temperatures. Of particular interest in Table 3 are the high tensile strengths obtained when using the 97Ag-1Cu-2Zr active braze filler metal. Samples averaging more than 21 ksi (147 MPa) were obtained using this recently developed

active braze filler metal. This relatively new (Refs. 17, 18) silver-based filler metal uses zirconium as the active element, but currently has very limited commercial availability.

Direct-Brazing Method

The direct-brazing method is the last method for joining metals to ceramics to be considered. As the name implies, the direct-brazing method allows metals to be directly brazed to ceramics without the need for metallization coatings. Unlike active filler metal brazing, however, the direct-brazing method utilizes standard brazing filler metals to accomplish the metal-to-ceramic braze. The direct-brazing process is illustrated on the right-hand side of Fig. 6. Comparisons of the two brazing methods portrayed in Fig. 6 illustrate how similar these processes are. Similar to the active brazing process, a direct-braze is made by cleaning the ceramic and metal materials, fixturing the assembly with the braze filler metal pre-placed between the metal and ceramic substrates and then brazing the entire assembly in an inert or UHV brazing atmosphere. During the direct-braze process, specific metal substrates and braze filler metal combinations interact to form an adherent metallic oxide layer on the oxide ceramic faying surface.

The dissolution, migration, and inter-

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Table 4 — Direct Brazed ASTM-F19 Tensile Button Test Results

Filler Metal	Nonmetal Substrate	Metal Substrate	Brazing Temperature/Time	Furnace Atmosphere	Average Tensile Strength
92 Au-8Pd	94% Alumina	Niobium	1270°C/4 min	Vacuum	12.8 ksi ± 1.8 ksi/ 88 MPa ± 12 MPa
62 Cu-35Au-3Ni	94% Alumina	Niobium	1058°C/3 min	Vacuum	8.8 ksi ± 1.0 ksi/ 61 MPa ± 7 MPa
50 Au-50Cu	94% Alumina	Niobium	1000°C/3 min	Vacuum	8.8 ksi ± 1.0 ksi/ 61 MPa ± 7 MPa

action of the base metal with the filler metal and ceramic surface are shown in Fig. 10 (Ref. 19). Electron microprobe analysis (EMPA) of a niobium-94% alumina ceramic sample brazed with 62Cu-35Au-3Ni (BAu-3) braze filler metal shows how the niobium base metal is enriched at the alumina ceramic surface, where it forms a relatively stable oxide. To perform a successful direct-braze, candidate metal substrates must contain an element or elements able to form thermally stable oxides and have sufficient solubility within the chosen liquid braze filler metal. As shown in Table 4, the direct-braze method was used to produce tensile button assemblies having average tensile strengths ranging from 9 to 13 ksi (61–88 MPa). For these assemblies, niobium base metal provided the active element required to react with the alumina ceramic.

There is a host of benefits for the designer or engineer to use the direct-brazing method. Some of these advantages are

1) Ease of use and lower expense, compared to other metal-ceramic brazing methods;

2) No metallization equipment or associated processes and process development is required;

3) A variety of conventional braze filler metals can be utilized covering a wide range of temperatures;

4) The direct-brazing method has been successfully used to hermetically join metal-ceramic components used in high-reliability long-term applications.

There are several disadvantages to using the direct-braze method. Among these are

1) Not all joint designs are viable. Similar to active brazing in this regard, the braze filler metal must be preplaced between the faying surfaces because the filler metal is unable to be drawn by capillary forces along the bare ceramic surface.

2) Good atmospheric control, while not as stringent as that required when active brazing, is also necessary when using the direct-brazing method.

3) The strengths obtained using the direct-braze method are slightly inferior to those obtained using the other discussed metal-ceramic brazing methods, as seen when comparing the strength data in

Active braze filler metals can be used as a replacement system for most metal-to-ceramic brazed assemblies with no loss of mechanical properties.

Table 4 to that shown in Tables 1–3. Transmission electron microscopy (TEM) analysis results on niobium-94% alumina ceramic direct brazed samples (Ref. 20) showed the niobium bonded with the glass-phase only. Though not yet evaluated, it is anticipated that a metal with the ability to form more thermally stable oxides than those of niobium will be required to adequately join high-purity alumina ceramics using the direct braze method.

In conclusion, high-strength, hermetically sealed metal-ceramic assemblies can be successfully brazed using a variety of methods, some requiring metallization of the ceramic member and others allowing the direct brazing of metals to ceramics.

The designer, engineer, or user can choose from a traditional metallization method such as moly-manganese/nickel plating or from a variety of thin-film coatings applied using PVD methods, which are specifically tailored to meet the needs of the application. Active braze filler metals can be used as a replacement system for most metal-to-ceramic brazed assemblies with no loss of mechanical properties. Whether choosing to use metallized ceramics or the direct-braze process, conventional braze filler metals can be used for the brazing operation. The direct-braze process has been demonstrated with a limited set of conventional filler metals to have adequate bond strength when used in conjunction with niobium metal substrates. Premetallized substrates may be used without joint geometry restrictions; however, active and direct-brazing techniques work best with butt or lap-style braze joint geometries where the brazing filler metal may be preplaced between the faying surfaces. ♦

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BRAZING & SOLDERING TODAY

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