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A Compendium of Brazed Microstructures For Fission Power Systems Applications

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Abstract

NASA has been supporting design studies and technology development for fission-based power systems that could provide power to an outpost on the Moon, Mars, or an asteroid. Technology development efforts have included fabrication and evaluation of components used in a Stirling engine power conversion system. This investigation is part of the development of several braze joints crucial for the heat exchanger transfer path from a hot-side heat exchanger to a Stirling engine heat acceptor. Dissimilar metal joints are required to impart both mechanical strength and thermal path integrity for a heater head of interest. Preliminary design work for the heat exchanger involved joints between low carbon stainless steel to Inconel 718, where the 316L stainless steel would contain flowing liquid metal NaK while Inconel 718, a stronger alloy, would be used as structural reinforcement. This paper addressed the long-term microstructural stability of various braze alloys used to join 316L stainless steel heater head to the high conductivity oxygen-free copper acceptor to ensure the endurance of the critical metallic components of this sophisticated heat exchanger. The bonding of the 316L stainless steel heater head material to a copper heat acceptor is required to increase the heat-transfer surface area in contact with flowing He, which is the Stirling engine working fluid.

Introduction

NASA is maintaining the option for fission surface power for the Moon, Mars and beyond by developing and demonstrating technology for affordable fission surface power systems (FSPS). The generation of stable and continuous power is critical in the exploration of the surfaces of any planet or even an asteroid. Nuclear power is an important option, especially for locations in the solar system where sunlight is limited in availability or intensity. The baseline power conversion system technology chosen for the FSPS concept is Stirling dynamic power conversion (Refs. 1, 2, and 3). Stirling power conversion underwent extensive development for space mission applications during the 1980s. The Space Power Demonstration Engine, consisting of two thermodynamically-coupled 12.5 kWe Stirling engines, was built to demonstrate 25 kWe Stirling conversion for use with the SP-100 space power reactor.

A cornerstone of the design architecture for an affordable FSPS was the decision to constrain the reactor coolant outlet temperature to less than 900 K (627 °C) (Ref. 4). This represents a relatively low temperature compared to 1375 K (1100 °C) for the SP-100 reactor designed during the 1980s. Constraining the reactor operating temperature allows the use of stainless steel and other non-refractory materials for the power system structure. Under the sponsorship of NASA, Sunpower, Inc. began the design and development of the Power Conversion Unit (PCU) for the fission power Technology Demonstration Unit (TDU) in April 2009, and is on schedule for delivery of a 12 kWe PCU consisting of two thermodynamically-coupled 6 kWe Stirling engines in 2012 (Ref. 5). A preliminary graphic of the PCU is shown in Figure 1 (Ref. 2). Dissimilar metal joints are required to impart both mechanical strength and thermal path integrity for the Stirling power conversion system hot-side heat exchanger (HX) components for a Fission Power System (FPS). Brazing is frequently a good approach for joining dissimilar metals in net-shape or near-net-shape configurations. Prototype designs include joints between 316L stainless steel, IN718, and copper in various combinations. A GRC-led research task was undertaken to evaluate and develop brazing methods to join various prototypical designs using nickel-, gold- and copper-based braze materials. Microstructural evaluation of the brazed joints was used to down-select braze alloys and methods for each material combination. This paper reports the results of electron microscopy (EM) and energy dispersive spectroscopy (EDS) microstructural evaluations, used to explore the varying degrees of chemical interactions of the brazed and the long-term-aged brazed joints, between 316L stainless steel (SS316L) and high conductivity oxygen-free copper (OFHC Cu). The long-term durability exposure consisted of heat treating the brazed samples at 823 K (550 °C) for 1000 hr in vacuum. A successful dissimilar-metal joint in this component application must provide mechanical

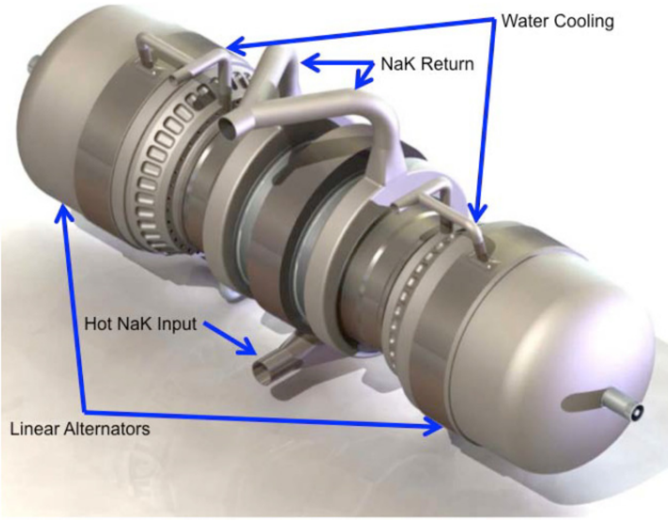

Figure 1.—TDU power conversion unit design (Sunpower, Inc.).

integrity, thermal conductivity, and stability for the expected mission life at elevated temperatures. Promising joints have been produced and a "toolbox" of options is described to respond to numerous potential designs or material selection options in the heat exchanger assembly.

Experimental Procedure

The braze alloys selected were originally identified to produce a physically sound joint. The various braze alloys evaluated followed the recommended literature and manufacturers' practices (Refs. 6 and 7). Two similar nickel-based brazes produced by Sulzer Metco (Braze Materials Guide, 2006) were used to investigate the effect of the presence of boron and the possible formation of borides in the joining systems. The presence of boron (B) in AMDRY 936 braze reduces the liquidus temperature to 1261 K, about 22° lower than AMDRY 930 to which B is not added. Both braze alloys have successfully been used in the past to join superalloys, stainless steels in complex assemblies, honeycomb, heat exchangers and wire screens. Technical interactions with the Morgan Advanced Ceramics, Westgo Metals Division resulted in the recommendation of the 35% gold – 65% copper braze (in wt.%). Since the liquidus temperature of the 35% Au – 65% Cu braze is approximately 1283 K, a second group of Au-Cu brazes (40% Au – 60% Cu and 50 % Au - 50 % Cu) with lower liquidus temperatures (1273 and 1243 K, respectively) were also included in the study. The gold-nickel braze alloy Nioro, successfully used to join SS316L to Inconel 718, was added to the study. A summary of these commercially available braze alloys used to join the SS316L with an approximate nominal composition in wt.% of Fe-12Ni-17Cr-2.5Mo-2Mn-0.035max C and the OFHC Cu in the heat exchanger is presented in Table 1.

TABLE 1.—Ni- AND AuCu-BASED BRAZE ALLOYS USED IN THIS STUDY TO JOIN 316L STAINLESS STEEL TO OFHC COPPER

	Alloy/braze	Composition, wt.%	Form	Braze temperature, K (°C)
Ni-based	AMDRY 936	Ni-19Mn-4Cu-6Si-1B0.03Re	Paste	1270 (997)
	AMDRY 930	Ni-22Mn-5Cu-7Si	Tape	1293 (1020)
Au-based	Nioro	82Au-19Ni	Foil (0.001 in.)	1253 (980)
	35%Au-65%Cu	Au34.70-Cu65.30	Foil (0.001 in.)	1293 (1020)
	40%Au-60%Cu	39.58Au-60.42Cu	Foil (0.001 in.)	1283 (1010)
	50%Au-50%Cu	49.67Au-50.33Cu	Foil (0.001 in.)	1253 (980)

The experimental brazing trials were done in vacuum at approximately 6×10^{-3} Pa at the indicated braze temperatures shown in Table 1. The brazing hold time was approximately 10 min and a small load was applied on top of the joining samples. Optical microcopy revealed that in most bonded couples a continuous bond was produced indicating that the copper and stainless steel alloys were wetted by the braze alloys studied with only one exception, the Nioro braze alloy, as described later. Occasionally, small regions with discontinuous porosity were detected in the metallographic cross sections. Despite the good bonding with both sides, careful metallurgical evaluation of the joints revealed that varying degrees of chemical and diffusion reactions occurred at or near the joint interfaces. Some of these interactions, such as second phase precipitation, could affect the long-term structural stability of the stainless steel or copper brazed materials. This paper describes the brazing options explored to join SS316L to OFHC Cu, the resultant microstructure produced and the microstructure stability of the brazed joints after exposure for 1000 hr at 823 K (550 °C) in vacuum. The group of brazed samples that were exposed over 1 month (1000 hr) in vacuum at 823 K (550 °C) is exhibited in Figure 2. The samples were metallographically mounted, polished and observed in optical and electron microscopes. Chemical analysis, by EDS, of the main features observed near the braze/alloy interface was used to identify the chemistry of new phases that may have formed during the brazing and aging process.

Results and Discussion

Ni-Based Brazes

The first two brazes presented in Table 1, AMDRY 936 and AMDRY 930, are very similar in composition with the exception of the presence of boron in AMDRY 936. They both contain similar levels of manganese (Mn), silicon (Si) and comparable amounts of copper (Cu). Figure 3 shows optical micrographs of the brazed SS316L to Cu with AMDRY 936 (Figure 3a) and AMDRY 930 (Figure 3b). Good braze coverage and bonding was achieved with both alloys. Figure 4a and b show field emission scanning electron microscopy

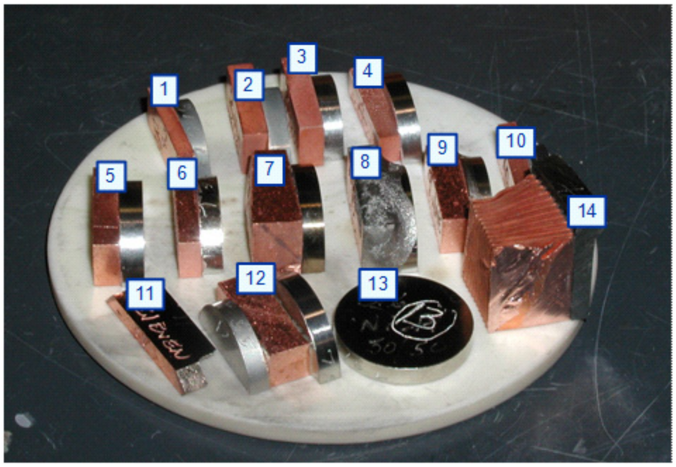

Figure 2.—Group of brazed samples aged at 823 K (550 ºC) for 1000 hr in vacuum.

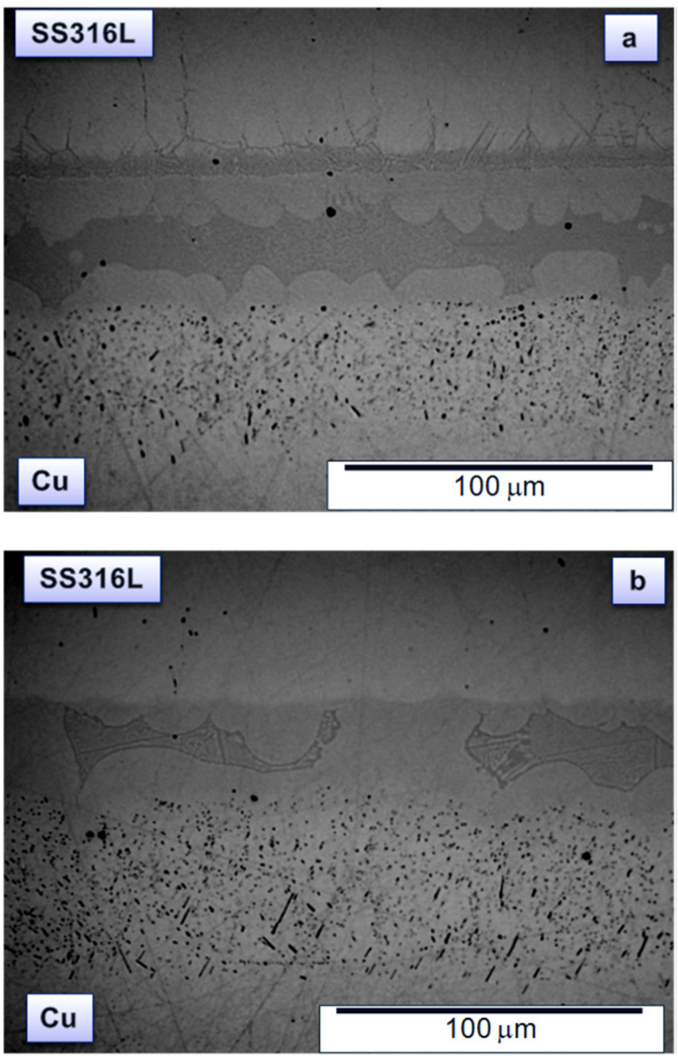

Figure 3.—Optical micrographs of 316L stainless steel brazed to copper with (a) AMDRY 936 and (b) AMDRY 930.

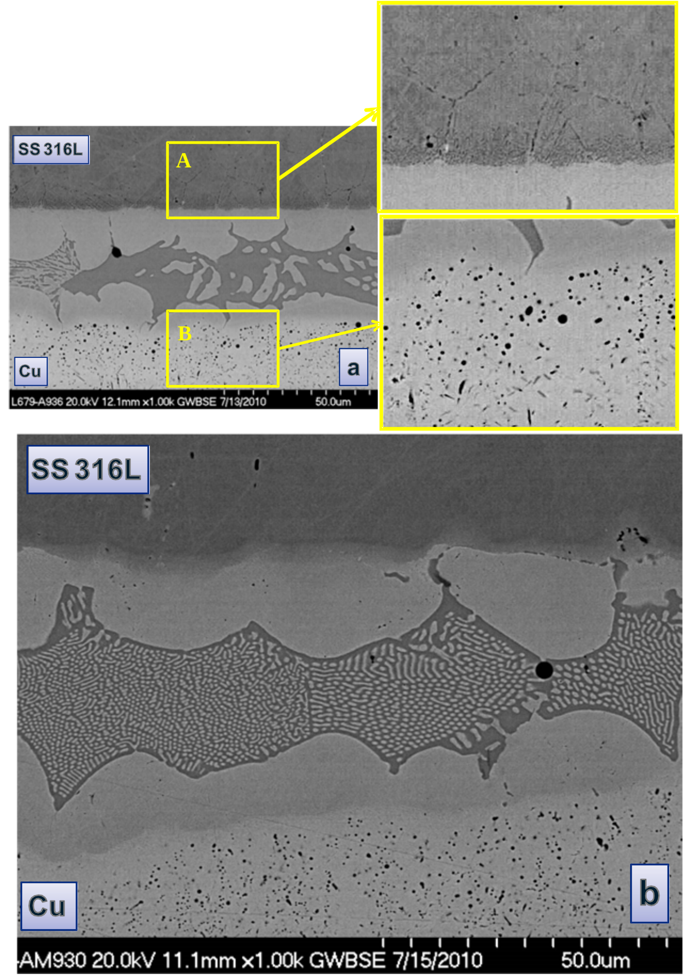

Figure 4.—FESEM micrographs of 316L stainless steel brazed to copper with (a) AMDRY 936 and (b) AMDRY 930. Eutectic phases formed at the center. Inserts in Figure 4a show (A) formation of chrome borides (CrB_2) and (B) Manganese-copper (Mn-Cu) enriched particulates. No second phase precipitation occurred near the stainless steel / AMDRY 930 braze interface.

(FESEM) cross section images of the features observed in the as-brazed SS316L to Cu samples with AMDRY 936 and AMDRY 930, respectively. Both alloys tend to form a eutectic-like microstructure in the center of the brazed regions, with one phase, darker in contrast (Figure 4a and b), enriched in Mn and Si with Ni, while the other, lighter in contrast, is enriched Cu with Ni with lower levels of Si and Mn. One of the main concerns with the presence of B in alloy AMDRY 936 is the possibility of B diffusing into the stainless steel and combining with iron (Fe) or chrome (Cr) to form some type of boride. This would deplete the stainless steel alloy of critical elements in regions near the braze, which could result in changes in the corrosion resistance and general properties of the alloy. Figure 4a (insert A) reveals the SS316L grain boundaries near the interface which are visible due to the presence of boride precipitates decorating the boundaries.

Evidence of Cr tied with B at the grain boundaries near the brazing interface was confirmed by electron microprobe analysis (Figure 5). Figure 5b and c show microprobe wavelength dispersive spectroscopy (WDS) quantitative maps of Cr and B in the region shown in Figure 5a. Boron was one of the elements contained in the braze alloy AMDRY 936, while Cr was available from the stainless steel alloy. The diffusion reaction that resulted in the formation of the borides seems to extend ~ 30 µm into the stainless steel side and occurs mainly at the stainless steel/braze interface in the as-brazed condition. The use of this braze alloy raises the concern that segregation of the boride precipitates could extend further in the SS316L alloy during long-term exposures. In contrast, the braze alloy AMDRY 930 that does not contain B, resulted in no second phase precipitation at or near the stainless steel/AMDRY 930 braze interface (Figure 4b). However, as mentioned earlier, AMDRY 930 requires a much higher brazing temperature than AMDRY 936. Multiple Mn-Cu rich particulates and strings were observed near the braze/copper interface in both braze alloys investigated, as revealed in the insert B of Figure 4a for AMDRY 936 and Figure 4b for AMDRY 930. Figure 6 shows a typical EDS spectrum of the Mn-Cu particulates present on the copper side of the couple brazed with AMDRY 930. The Mn is present in the braze alloy, and since copper and manganese have complete solubility in each other, diffusion of Mn during the brazing process must have occurred to form the fine Mn-Cu particulates inside the Cu matrix alloy. Figure 7 contrasts the extent of the Mn-Cu particulates found on the copper side in the as-brazed versus the 823 K (550 °C), 1000 hr aged condition. The aging exposure had more than doubled the Mn-Cu formation depth of approximately 40 to 100 µm as measured from the braze/copper interface into the copper side.

The aging at 823 K (550 °C) for 1000 hr of the SS316L to Cu joint brazed with AMDRY 930 resulted in several other unique microstructures. Figure 8a and b show low and high magnification FESEM images of samples in the brazed plus aged condition. Besides the common eutectic microstructure observed in the middle of the brazed section for both as-brazed and aged conditions, fine platelets have also formed in the vicinity of either SS316L or Cu sides during the aging treatment. These platelets were not present in the as-brazed microstructure. Clearly some elements have a sufficient driving force to diffuse and combine into more stable compounds. As-shown in Figure 8, Spectrum EDS-A corresponds to a Cr (Mo, Ni) – silicide phase that seems to have formed near the interface boundary between the SS316L and the brazed region. Spectrum EDS-B indicates that the platelet precipitates are mostly enriched in Ni, Si and Mn, while Spectra EDS-C and EDS-D correspond to the composition typically observed in the eutectic region. One is enriched in Ni, Si and Mn (dark region) and the other is enriched with Cu, Ni and Mn (bright region).

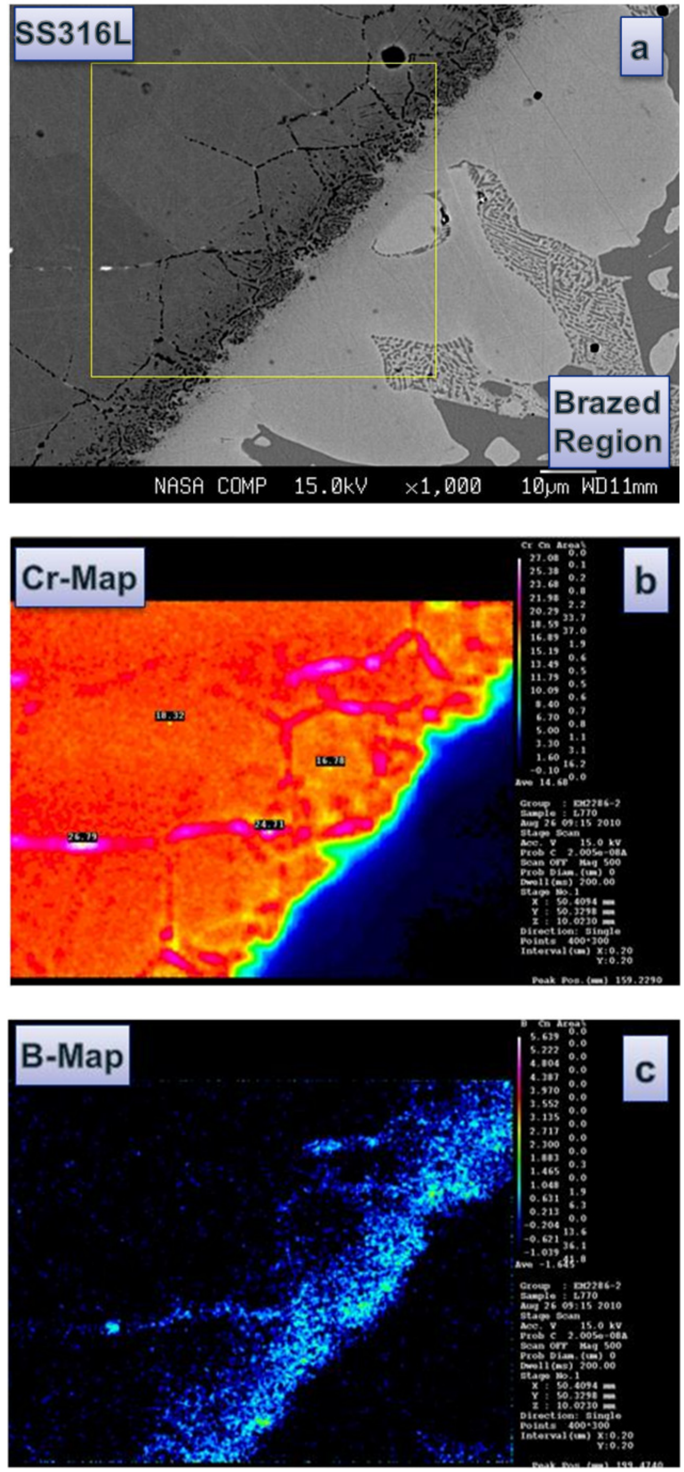

Figure 5.—Brazed SS316L to Cu with AMDRY 936 (a) Electron microprobe image and (b) and (c) Wavelength dispersive Spectroscopy (WDS) quantitative maps for Cr and B, respectively.

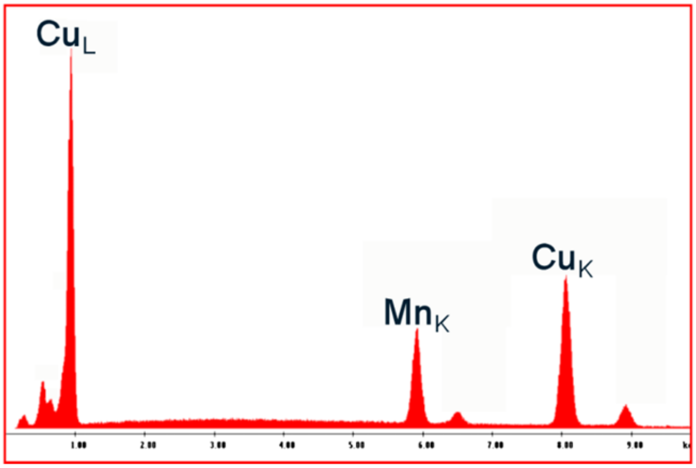

Figure 6.—Typical EDS spectrum of the fine Mn-Cu particulates formed on the Cu side of the as-brazed SS316L/Cu with AMDRY 930.

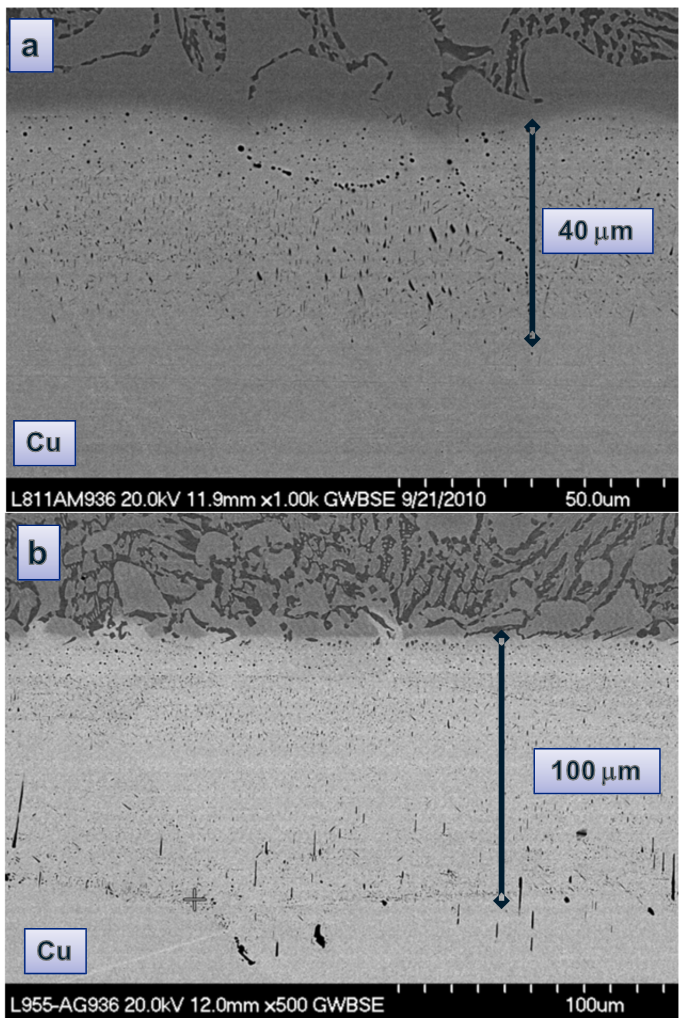

Figure 7.—FESEM Micrographs of the AM936 braze/copper interfaces in (a) as-brazed and (b) aged to 823 K (550 °C) for 1000 hr. Aging exposure resulted in an expansion of the region with Mn-Cu particulates.

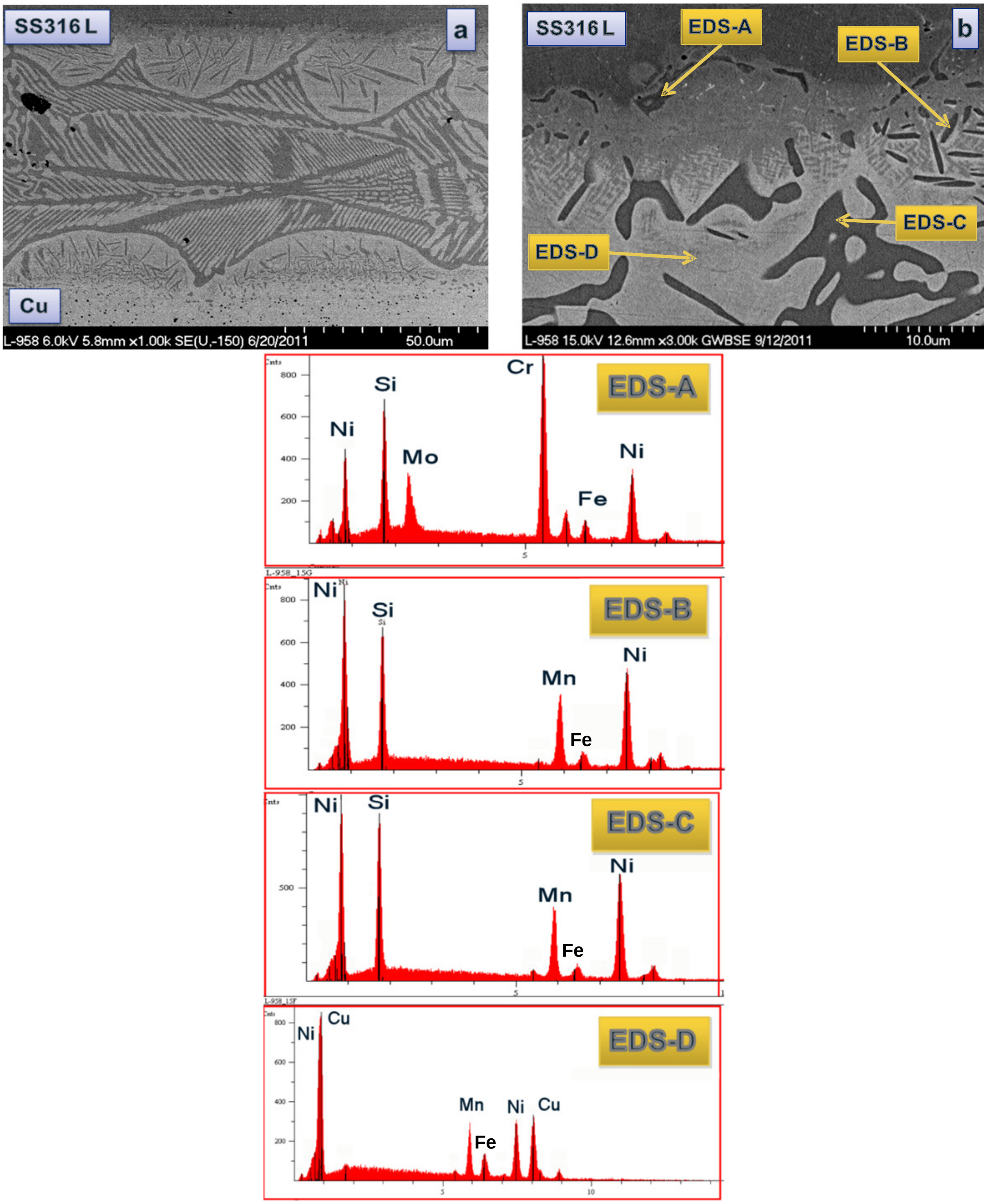

Figure 8.—(a) Low and (b) high magnification FESEM micrographs for sample aged for 1000 hr at 823 K (550 °C). Brazing alloy used is AMDRY 930. EDS analyses of second phases present next to SS/braze interface eutectic region.

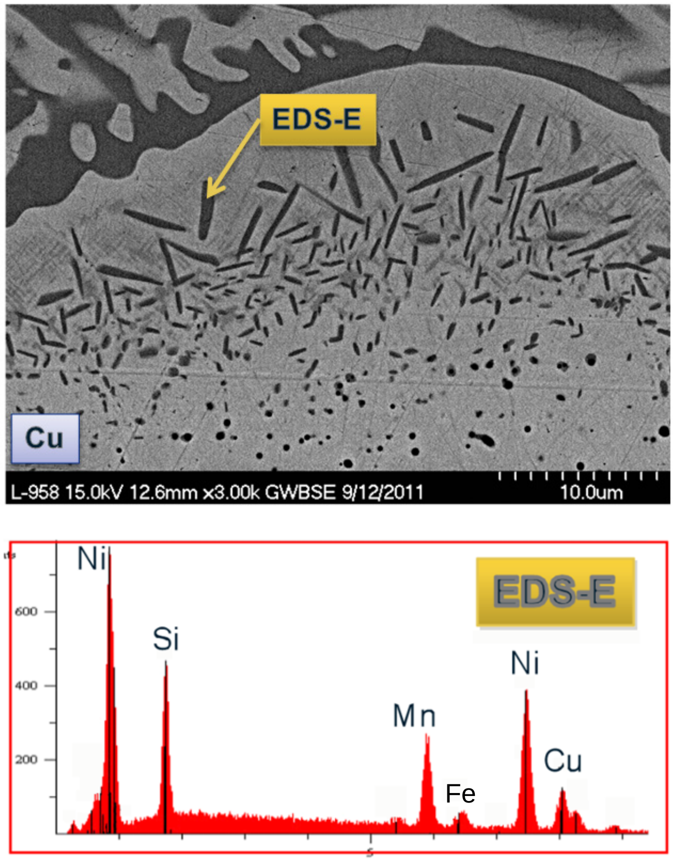

Figure 9.—Platelets formed inside the AMDRY 930 brazed section, near the braze/Cu interface showing similar composition as the one near the SS/raze interface EDS-B of Figure 8.

The platelets formed inside the braze section, near the Cu/braze interface have similar composition as the one found near the stainless steel/braze interface and are presented in Figure 9. The EDS-E spectrum confirms that the platelets are again enriched in Ni, Si, Mn, and Cu. The Cu found came from the surrounding matrix.

Au-Based Brazes

A group of gold-copper (Cu/Au) brazes and an additional gold-nickel (Nioro) braze were explored as an option to join the stainless steel component to the copper. The Cu/Au braze alloys were selected to minimize possible reactions with the components to be joined and also because their different Au to Cu composition levels control their melting points and therefore, their brazing temperatures. Figure 10 shows optical micrographs of the three Au-Cu braze alloys. Each braze wetted and spread evenly on both alloys, which resulted in good bonds.

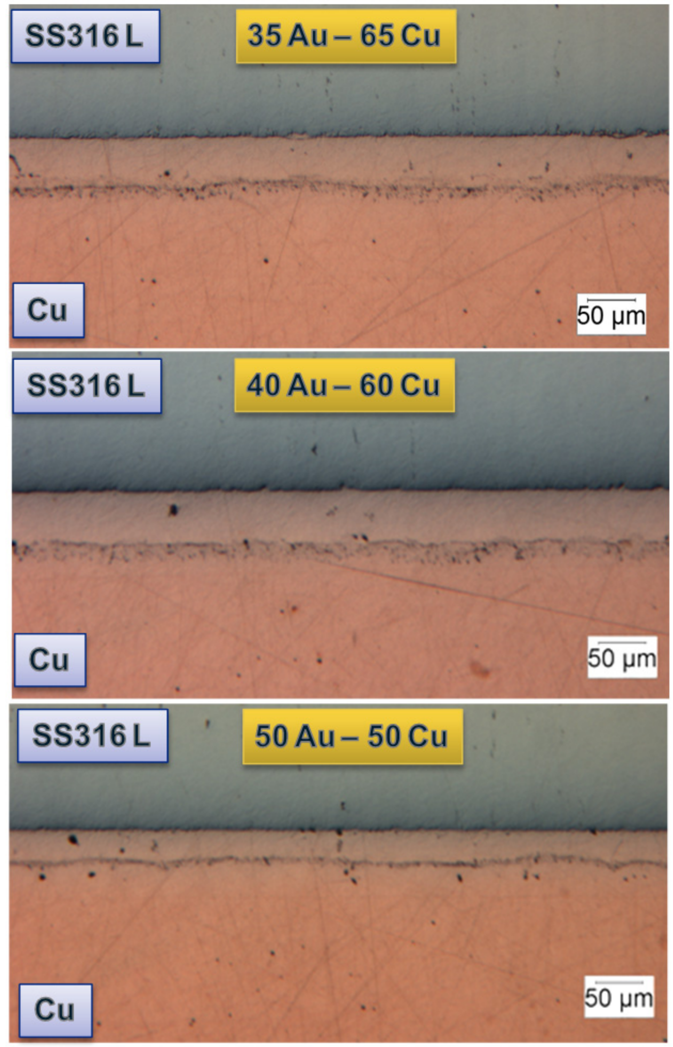

Figure 10.—Optical micrographs of stainless steel brazed to copper using gold-copper braze alloys showing relatively good bonding.

Figure 11 shows optical micrographs of the brazed alloys after the 1000 hr aging heat treatment at 823 K (550 °C). Although difficult to resolve in the optical micrographs, some increased reactions have occurred at the braze/copper side interface. Figure 12 shows low and high magnification FESEM images of the two SS316L and Cu alloys brazed with the 50% Au – 50% Cu braze alloy. The high magnification image shows clearly the particulates that have precipitated near the braze/copper interface. EDS analyses (Figure 13) of the region selected in Figure 12, indicate that the precipitates observed on the Cu side are enriched in Fe (with some Cr) in addition to Cu and Au. Diffusion of Fe and Cr from the stainless side is, therefore, possible in this system through the Au-Cu braze. Similar features were observed in the other two

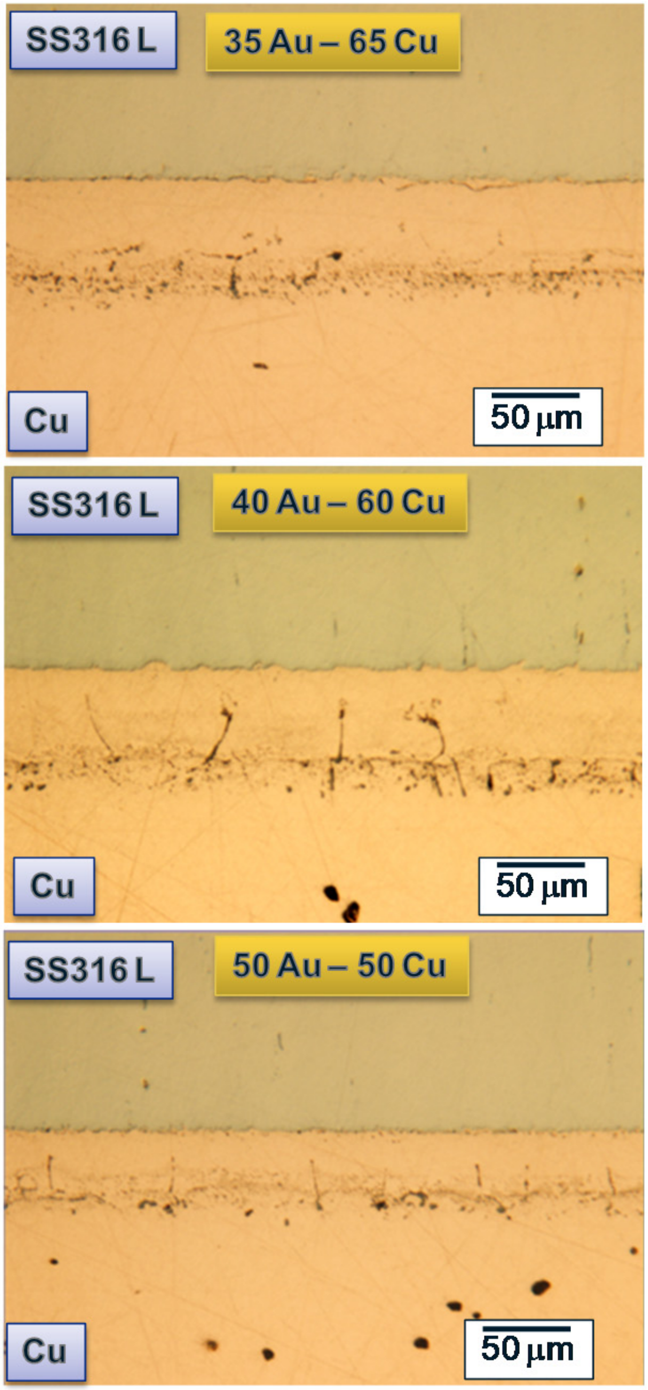

Figure 11.—Optical micrographs of stainless steel brazed to copper using gold-copper braze alloys after an exposure for 1000 hr at 550 ºC showing some reactions at the braze/copper interface.

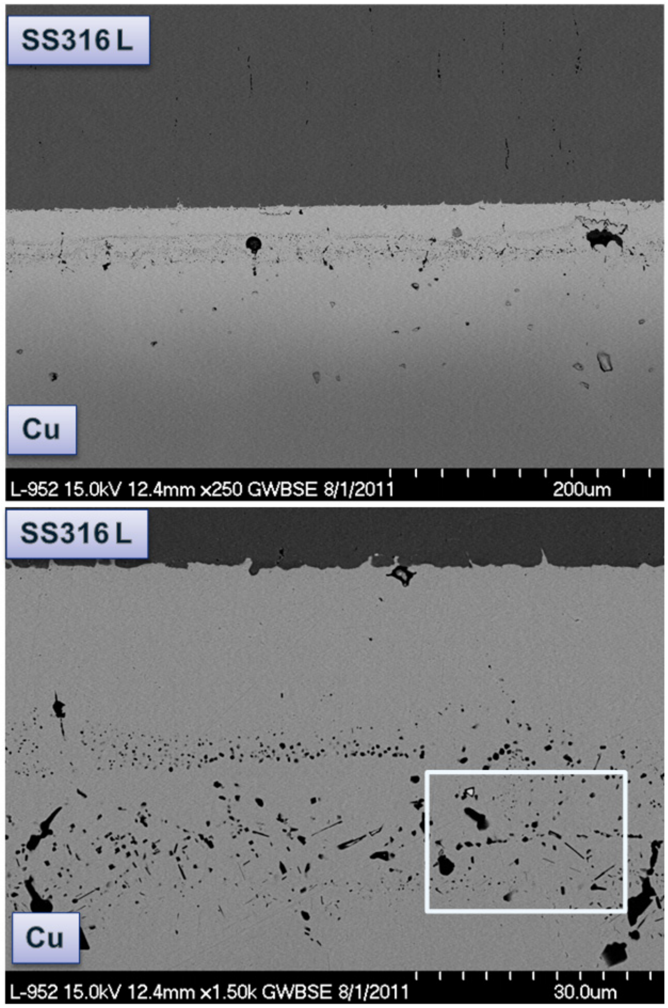

Figure 12.—Low and high magnification FESEM images of stainless steel brazed to Cu using a 50 Au – 50 Cu braze alloy after the aging heat treatment at 550 ºC showing the formation of secondary phases at the braze/Cu interface.

Au – Cu braze compositions. The extent of this reaction is limited to approximately 30 μm deep in the copper side, even after aging for 1000 hr.

The Nioro braze resulted in limited wetting of the copper surface and discontinuous bonding. Figure 14 shows an example of the poor braze bond observed when Nioro was used to bond SS316L to OFHC copper. Nioro was removed from the list of potential braze alloy candidates for SS316L to Cu bonding.

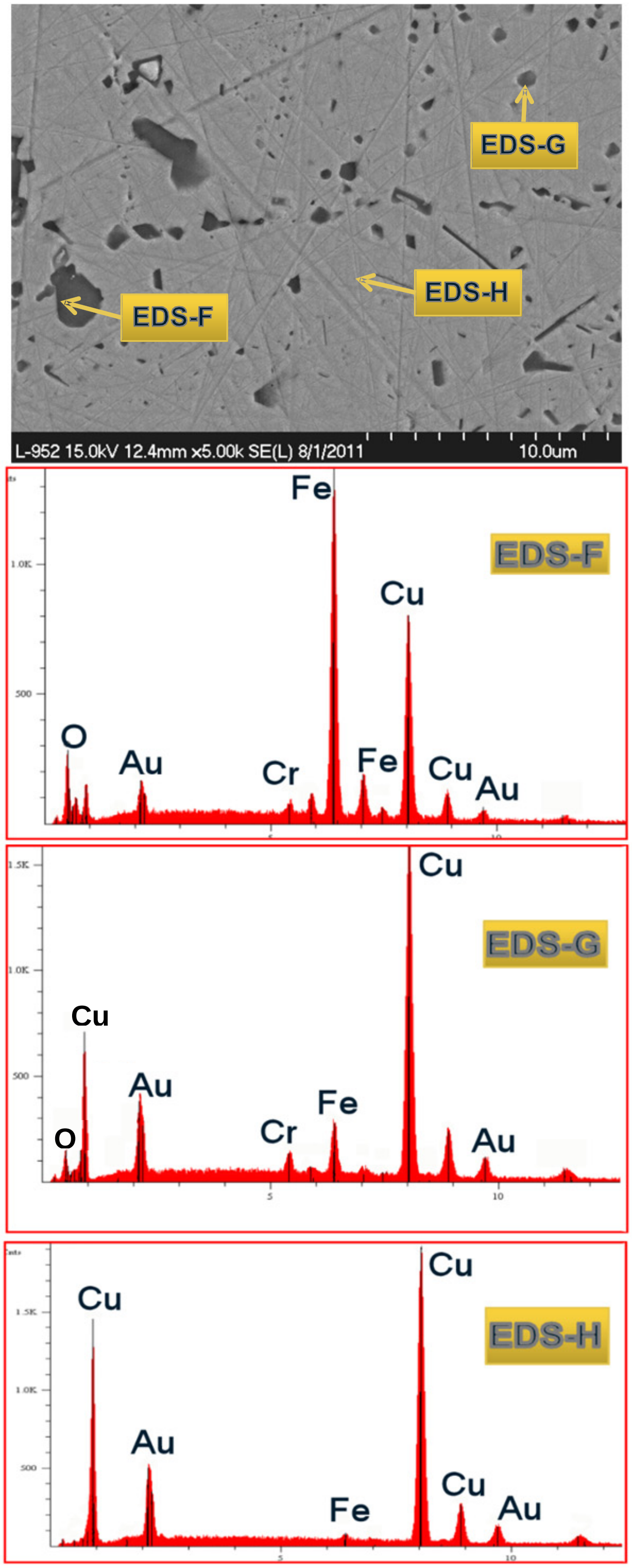

Figure 13.—Energy dispersive spectroscopy (EDS) spectra of main particulates and matrix detected in the selected region shown in Figure 12. Most precipitates are enriched in Fe (with some Cr) in addition to the presence of Cu and Au.

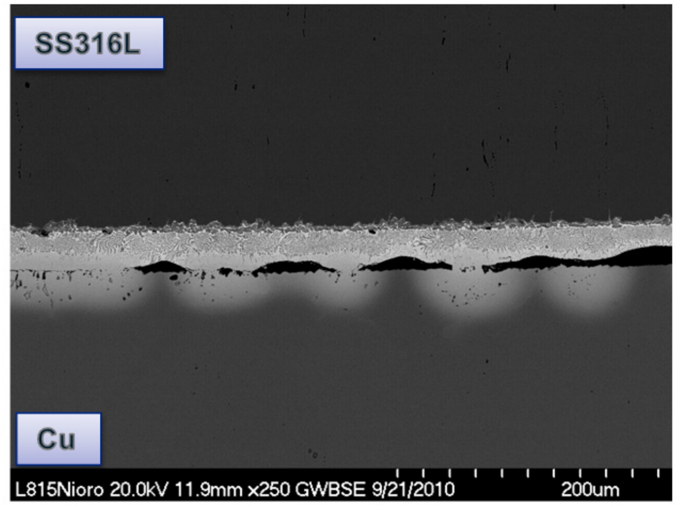

Figure 14.—Stainless steel brazed to copper using Nioro braze alloy showing poor bonding.

Summary

NASA GRC, collaboratively with industry, examined both paste and foil brazing materials to develop procedures, and brazing methods to join various samples and prototypical Stirling heat exchanger concentric ring configurations. Careful metallurgical evaluation of brazed joints between 316L stainless and copper had revealed varying degrees of chemical interactions in the as-brazed condition and after the long-term aging exposure. The microstructure stability and chemical interactions of the various brazes with the component alloys were explored by electron microscopy and energy dispersive spectroscopy after brazing and aging. The Ni-based brazes, with and without B, and the three gold-copper brazes were down-selected for the aging study at 823 K (550 °C). Despite some chemical interaction, the present study suggests that these alloys are adequate for joining 316L stainless steel to copper for use at 823 K (550 °C) for a non-load-carrying component. Conversely, the poor wetting and bonding of the Nioro braze to copper resulted in its elimination as a candidate braze alloy to join SS316L to Cu.

Table 2 summarizes the findings and recommendations after the long-term aging study. All of the alloys have, up to some degree, reacted with the surrounding matrices and formed secondary small phases. These precipitates, or secondary phase formations, do not seem to create a major concern at the 823 K (550 °C) application temperature. In the case of AMDRY 936, the B reaction with Cr zone did not expand beyond 30 μm from the alloy/braze interface after the 550 °C exposure for 1000 hr. Due to the relatively limited reaction, this SS316L/AMDRY936/Cu combination is deemed acceptable and comparable to AMDRY 930. However, the presence of B might be of concern if the brazed alloys are to

TABLE 2.—POST-BRAZE OBSERVATIONS AFTER AGING EXPOSURE AT 823 K (550 °C) FOR 1000 hr

Metal 1	Metal 2	Braze alloy	Wetting	Phase formation with Metal 1	Phase formation with Metal 2	Compatibility with base materials	Recommended
SS316L	Cu	AMDRY 936	Good	Cr-Boride at G. Bds	Mn-Cu	Comparable to as-brazed microstructure	Acceptable
SS316L	Cu	AMDRY 930	Good	Minimal	Mn-Cu	Formation of platelets in brazed section	Acceptable
SS316L	Cu	Nioro	Poor	------------------------	----------------	---	No
SS316L	Cu	35Au-65Cu	Good	Minimal	Fe(Cr) Cu, Au	Diffusion of Cr, Fe to Cu	Acceptable
SS316L	Cu	40Au-60Cu	Good	Minimal	Fe(Cr) Cu, Au	Diffusion of Cr, Fe to Cu	Acceptable
SS316L	Cu	50Au-50Cu	Good	Minimal	Fe(Cr) Cu, Au	Diffusion of Cr, Fe to Cu	Acceptable

be exposed to a corrosive fluid or to significant radiation fluence. Higher use-temperatures beyond those used in this study, could result in more extensive inter-diffusion of some key elements found in the stainless steel or braze alloys and, therefore, the usefulness of these alloy systems would have to be reassessed.

Conclusions

All brazing alloys based on Ni or Au-Cu have, to some degree, reacted with the surrounding matrices and formed secondary small phases after the long-term aging exposure. However, these reactions are limited and not a major concern for an application at 823 K (550 °C). The Nioro braze alloy is the only braze that is not recommended due to its poor wetting of the copper substrate.

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14. ABSTRACT
NASA has been supporting design studies and technology development for fission-based power systems that could provide power to an outpost on the Moon, Mars, or an asteroid. Technology development efforts have included fabrication and evaluation of components used in a Stirling engine power conversion system. This investigation is part of the development of several braze joints crucial for the heat exchanger transfer path from a hot-side heat exchanger to a Stirling engine heat acceptor. Dissimilar metal joints are required to impart both mechanical strength and thermal path integrity for a heater head of interest. Preliminary design work for the heat exchanger involved joints between low carbon stainless steel to Inconel 718, where the 316L stainless steel would contain flowing liquid metal NaK while Inconel 718, a stronger alloy, would be used as structural reinforcement. This paper addressed the long-term microstructural stability of various braze alloys used to join 316L stainless steel heater head to the high conductivity oxygen-free copper acceptor to ensure the endurance of the critical metallic components of this sophisticated heat exchanger. The bonding of the 316L stainless steel heater head material to a copper heat acceptor is required to increase the heat-transfer surface area in contact with flowing He, which is the Stirling engine working fluid.

15. SUBJECT TERMS
Stirling engines; Heat exchangers; Stainless steel; Power reactors; Nickel alloys; Brazing; Fabrication; Metal bonding; Metal joints; Microstructure

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