Active Metal Brazing and Characterization of Silicon Nitride-to-Metal Joints

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Abstract

Active metal brazing of Inconel and titanium to turbine grade silicon nitride, widely acknowledged to be one of the most difficult ceramics to join to metals, was demonstrated for NASA's Subsonic Rotary Wing project. A titanium-containing active braze, Cu-ABA (brazing temperature ~1297 K) was used in foil form together with judiciously arranged interlayers of Ni, W, Mo, Ta, Cu, and Kovar to manage residual stresses in joints. The joints were vacuum brazed and examined for microstructure, composition, and microhardness using optical microscopy, scanning electron microscopy, energy dispersive spectroscopy, and microhardness testing. Interlayers of Ta and W; Ni and W; Kovar and W; and Ni, W, Ni led to sound joints of Si_3N_4 with Inconel-625 and Ti; however, micro-cracking occurred within Si_3N_4 in some systems without loss of joint integrity. The formation of interfacial reaction layers enriched in Ti and Si (possibly a titanium silicide phase) suggested chemical reaction-induced bonding. Self-joined Si_3N_4 displayed the best joint characteristics.

Introduction

During 2009 summer, over a ten-week period, research on joining of silicon nitride (Si_3N_4) to metals was carried out in the Ceramics Branch at the NASA Glenn Research Center at Cleveland, OH. The research was done with support from Lewis' Educational and Research Collaborative Internship Program (LERCIP). This paper presents the objectives and the main findings of the research project.

The research focused on joining of silicon nitride to titanium metal and Inconel-625[1], a nickel-base superalloy used in turbine engines, as a part of the joining subtask for NASA's Subsonic Rotary Wing project. Silicon nitride is a light-weight ceramic with excellent high-temperature strength, creep-resistance, low thermal expansion, and excellent resistance to thermal shock, wear, and oxidation. This ceramic material is currently used in reciprocating engine components, turbochargers, auxiliary power unit components for aircraft, bearings, and metal cutting tools.

There is interest in using silicon nitride in turbine components of the next-generation turbo-shaft engines because it can be used without the need for extensive cooling that is required when using metallic parts. This is projected to significantly raise engine efficiency and performance. Additionally, Si_3N_4 components may be easier to fabricate than silicon carbide fiber-reinforced silicon carbide matrix composites (another contender for such applications), because of the inherent complexity in weaving cooling channels and sharp edges using the SiC fibers. In an ongoing research effort at NASA Glenn, joining and integration of Si_3N_4 ceramics with metallic, ceramic, and composite materials using braze interlayers with the liquidus temperature in the range 1023-1513 K is being investigated. One way to integrate Si_3N_4 to metallic parts is by making the entire turbine or rotor out of ceramic instead of the individual blades, and inserting compliant

1. Inconel-625 is a product of Inco Specialty Metals, and has a nominal composition (in wt%) of 58Ni-21.5Cr-9Mo-5Fe-1Co-0.5Si-0.5Mn-0.4Al-0.4Ti -3.7% (Nb, Ta).

layers into the slot. These rotors have to be connected to metal rods or shafts. This is where the joining of Si_3N_4 to high-temperature metals and alloys comes in.
Because of its brittle nature, silicon nitride, like other ceramics, is less amenable to shaping via machining and conventional manufacturing techniques. As a result, robust joining techniques that are capable of integrating geometrically simpler silicon nitride parts into complex components play a critical enabling role. Additionally, in advanced technology applications such as turbine rotors, silicon nitride needs to be integrated with other types of materials such as metals and alloys. Thus, one key aspect of utilization of silicon nitride is its joining response to diverse materials.

Silicon nitride is acknowledged to be one of the most difficult ceramics to join to metallic materials (Suganuma et al (1988)) in spite of its useful properties. This difficulty arises mainly from the relatively small coefficient of thermal expansion (CTE) of silicon nitride (~$3x10^{-6}$ K^{-1}), even compared with other engineering ceramics. A structural alloy such as steel (CTE: ~$14x10^{-6}$ K^{-1}) and high-temperature alloys such as Inconel (CTE: ~$16x10^{-6}$ K^{-1}) have appreciably larger CTE which causes large residual stresses during cooling from the joining temperature and often lead to the fracture of the ceramic. Being extremely brittle, ceramics are less forgiving than metals which fail more gracefully than ceramics. Among prominent applications of Si_3N_4-to-metal joints, turbo-charger rotors with Si_3N_4 blades joined to a steel shaft with laminated interlayers via active metal brazing is probably the most well-known product. It has a soft metal/low expansion and hard metal/soft metal interlayer structure (e.g., laminate interlayer of Fe/W). However, the total thickness of joints that utilize interlayers is rather large; for example, in the turbo charger rotor, the total thickness of interlayers is 2 mm.

Among the filler metals that have been used to join Si_3N_4 ceramics (Brochu et al (2004); Gopal et al (2001); Hadian and Drew (1996); Liu et al (2006); Loehman et al (1990); Peteves et al (1996); Suganuma et al (1988); Zhang et al (2008)), Ag-Cu eutectic alloys containing Ti have been most widely used. Other notable fillers include Ag-Cu-In and Ag-Cu-Sn containing Ti. Besides Ti, active elements such as Hf, Zr, Nb and Ta also have been evaluated for joining Si_3N_4. In addition, active brazes such as Pd-Ni-Ti, Au-Pd-Ti, and Cu-Pt-Ti/Nb as well as non-reactive brazes Pd-Ni and Au-Pd-Ni in conjunction with premetallized Si_3N_4 also have been used. Chromium has been used as an active metal in Ni-Pd and Ni-Si eutectic braze alloys. Many other brazes have been used to join Si_3N_4 ceramics. The Ag-Cu-Ti fillers are known to produce the highest levels of joint integrity and are most commonly employed to join silicon nitride ceramics. There also is interest in evaluating brazes with melting points higher than that of the Ag-Cu-Ti fillers to join Si_3N_4.

In this study, an active braze alloy (ABA), called Cu-ABA, with liquidus temperature (T_L~1297 K) higher than that of commonly used Ag-Cu eutectic brazes containing Ti, was evaluated. In particular, joints were created using vacuum brazing, and joint microstructure, composition, and microhardness at room temperature were evaluated with the help of optical microscopy (OM), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and Knoop microhardness testing. The research outcomes were included in the Subsonic Rotary Wing (SRW) project report, and shall be utilized in developing recommendations for continued joining research on Si_3N_4 ceramics.

Experimental Procedure

Silicon nitride containing 4 wt% Y_2O_3 as a sintering aid was used for joining. The material, designated as NT-154, was obtained from St. Gobain Ceramics. Silicon nitride was brazed to silicon nitride and metallic substrates using a commercial Cu-Si-Al-Ti braze alloy, Cu-ABA (from Morgan Advanced Ceramics, Hayward, CA). The alloy has a nominal composition (in weight %) of 92.75Cu-3Si-2Al-2.25Ti, and with the solidus and liquidus temperatures of T_S~1231 K and T_L~1297 K, respectively. Cu-ABA has high ductility (42%) and was obtained in foil form (thickness ~50 μm). The elastic modulus, yield strength and tensile strength of Cu-ABA are 96 GPa, 278 MPa, and 520 MPa, respectively, and its coefficient of thermal expansion (CTE) is 19.5×10^{-6}/K.

For multilayer joints with Inconel-625, an amorphous braze alloy MBF-20 (Ni-Cr-B-Si) from Honeywell, Inc., was used in combination with Cu-ABA (with MBF-20 contacting the Inconel and Cu-ABA contacting the Si_3N_4 in the joint). The superalloy, Inconel 625 was obtained from Inco Specialty Metals, and had a nominal composition (in wt%) of 58Ni-21.5Cr-9Mo-5Fe-1Co-0.5Si-0.5Mn-0.4Al-0.4Ti (with the remaining 3.7% of the composition consisting of Nb and Ta). Commercially pure titanium metal substrates were obtained from Ti Metal Inc., MO. In order to create multilayer joints, single or multiple layers of Ni, W, Mo, Ta, and Cu obtained from GoodFellow, MA, were used. Selected physical and mechanical properties of these interlayer materials are shown in Table 1. A total of 26 separate joints were created using various combinations of substrates and interlayer materials.

The substrates and braze foils were sliced into 2.54 cm x 1.25 cm x 0.25 cm pieces using either diamond saw (SN) or ceramic saw (Inconel and Ti). All materials were ultrasonically cleaned in acetone for 15 min. Two braze foils were sandwiched between the substrates, and a load of ~1 to 2 N (~3.5-7.2 kPa pressure) was applied during brazing. The assembly was heated in a furnace to ~15-20 K above braze T_L) under vacuum (~10^{-6} torr), soaked for 30 min. at the brazing temperature, and slowly cooled to room temperature. The brazed joints were mounted in epoxy and prepared for metallurgical examination using grinding and polishing on a Buehler automatic polishing unit, and examined using optical microscopy (Olympus DP 71 system) and SEM coupled with EDS on a JEOL 840A unit. The polished joints were subjected to microhardness test with a Knoop micro-indenter on Struers Duramin-A300 machine under a load of 200 g and loading time of 10 s. Multiple hardness scans were accessed across representative regions of joined samples to check the reproducibility and consistency of the data.

Results and Discussion

This research investigated the use of metallic interlayers of graded strength and expansion properties to join Si_3N_4 to Ti and to Inconel 625. The graded interlayer approach utilizes a variety of materials that have coefficients of thermal expansion (CTE) within the range of the materials that are being bonded together. With this approach, the goal is to lessen the internal stress of the bond between Si_3N_4 and Inconel-625 (or Ti) by layering the other materials so that the CTE transitions from the Si_3N_4 to the Inconel-625 (or Ti). In the study, the materials used as interlayers were tungsten (CTE: 4.5x10^{-6} K^{-1}), molybdenum (CTE: 4.8x10^{-6} K^{-1}), tantalum (CTE: 6.5 x10^{-6} K^{-1}), niobium (CTE: 7.1x10^{-6} K^{-1}), copper (CTE: 16.5 x10^{-6} K^{-1}), Kovar (CTE: 5.5- 6.2 x10^{-6} K^{-1}), titanium (CTE: 8.6 x10^{-6} K^{-1}), and nickel (CTE: 13.4 x10^{-6} K^{-1}).

Theoretical models (Park et al (2002)) of residual stress in ceramic/metal joints indicate that such stresses might be effectively accommodated by a judicious arrangement of ductile interlayers of graded yield strength and thermal expansion coefficient within the joint region. Furthermore, calculations also show that multiple interlayers accommodate residual stresses more effectively than single interlayers. One concern is that multiple interlayers also increase the number of interfaces within the joint and the probability of defects besides increasing the joint thickness. It also can be a challenge to keep the many layers from shifting around when brazing. Brazing of multilayer joints must, therefore, be done under carefully controlled conditions (e.g., high vacuum) to avoid formation of defects in multilayer joints.

Figure 1 shows Si_3N_4 joined to itself using Cu-ABA but without interlayers. The joint is devoid of interfacial defects such as voids and micro-cracks, and metallurgically sound. In the present study, the main criterion for acceptability of joints was taken to be defect-free interface structure as revealed via optical microscopy and scanning electron microscopy. EDS scans indicated that the interface is enriched in Ti and Si, possibly indicating reactive formation of titanium silicide that may have facilitated bond formation.

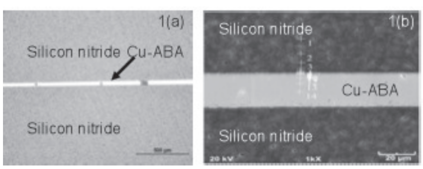


Fig. 1(a) Fig. 1(b)

Fig. 1 (a) An optical photomicrograph and (b) a secondary electron SEM image of a Si_3N_4/Cu-ABA/Si_3N_4 joint.

The general procedure used in the study for brazing multilayer joints is schematically shown in Fig. 2(a). Figures 2(b)-(e) show the secondary electron images and EDS composition map for a multilayer Si_3N_4/Ti joint with the following interlayer and braze foil arrangement:

Si_3N_4/Cu-ABA/W/Cu-ABA/Ta/Cu-ABA/Ti. Based on microstructure observation, the joint is sound and well-bonded; however, a curved crack has developed within the Si_3N_4 (Fig. 2b) possibly due to the residual stresses. The titanium substrate shows a diffused interface with the braze region. The Knoop hardness scans across the joints display a consistent pattern with the highest hardness in the ceramic followed by tungsten, tantalum, and Inconel-625. The braze and the neighboring interaction zone reveals a relatively low hardness.

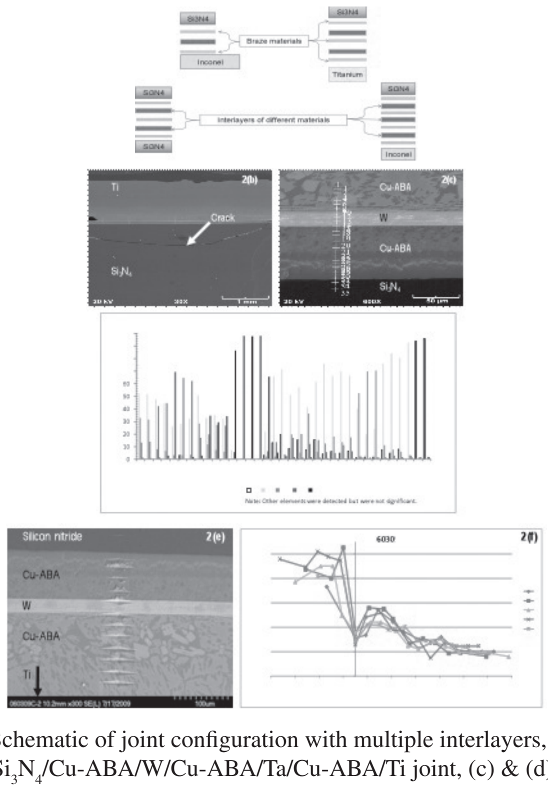
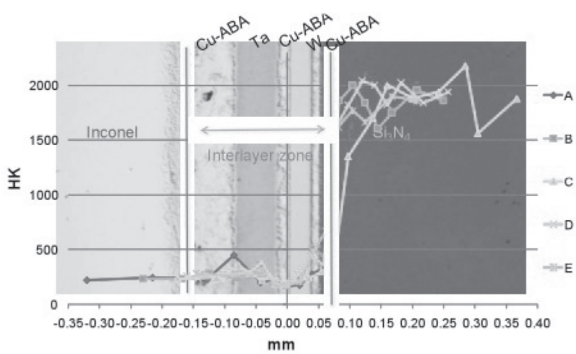


Fig. 2(a) Schematic of joint configuration with multiple interlayers, (b) overall view of a Si_3N_4/Cu-ABA/W/Cu-ABA/Ta/Cu-ABA/Ti joint, (c) & (d) secondary electron SEM image and EDS composition plot across point markers shown in (c), (e) SEM view of the joint near Si_3N_4/Cu-ABA/W side showing Knoop indentation marks, and (f) microhardness profile across the joint (multiple traces are marked as A, B, …).

A Si_3N_4/Inconel-625 joint with W and Ta interlayers with the following arrangement Si_3N_4/Cu-ABA/W/Cu-ABA/Ta/Cu-ABA/Inconel, is shown in Fig. 3. The microstructure of the joint showing the physical location of the braze and W and Ta interlayers is shown in Fig. 3(a) together with superimposed Knoop microhardness profiles. The assembly is well-bonded at each interface within the joint, and EDS scans across the Si_3N_4/Cu-ABA interface reveal a reaction layer enriched in Ti and Si (point markers 6-12 in Fig. 3b & c). This reaction layer could possibly be a titanium silicide reaction layer.

Figures 4(a)–(d) show a Si_3N_4/Inconel-625 multilayer joint containing W and Mo interlayers with the following arrangement: Si_3N_4/Cu-ABA/W/Cu-ABA/Mo/Cu-ABA/Inconel. Although the joint visibly displayed integrity and appeared to be well-bonded, a few micro-voids were observed within the joint region. Additionally, a crack that emanates from the reaction zone propagates through the Si_3N_4 parallel to the joint region (Fig. 4a). At the Si_3N_4/Cu-ABA interface (Fig. 4b) Ti and Si enrichments are noted (Fig. 4c). The microhardness scans (Fig. 4d) across the joint are reproducible, and display the highest hardness within the Si_3N_4 and two



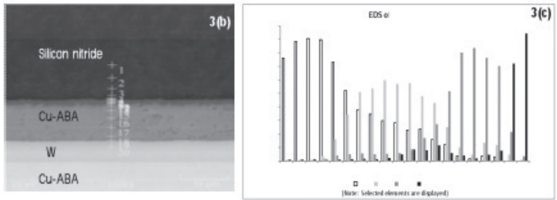


Fig. 3 (a) An optical photomicrograph of a multilayer Si_3N_4/Cu-ABA/W/Cu-ABA/Ta/Cu-ABA/Inconel joint showing interlayers and superimposed microhardness profiles, and (b) & (c) show the Si_3N_4/Cu-ABA interface together with the elemental composition at point markers of Fig. 3b.

smaller peaks within the hard W and Mo interlayers. The braze (plus interaction zone) region displays the lowest hardness. Overall, the joint may be considered of acceptable quality.

The secondary electron SEM images of a Si_3N_4/Inconel-625 joint made using W and Kovar intelayers in conjunction with two types of braze foils, Cu-ABA and MBF-20, is shown in Fig. 5. The joint configuration is as follows: Si_3N_4/Cu-ABA/Kovar/Cu-ABA/W/Cu-ABA/Kovar/MBF-20/Inconel. The joint contains two Kovar interlayers each of 390 µm thickness (similar joints with thicker, 500 µm, Kovar interlayers yielded inferior joints). The interfaces across the different regions of the joint (Fig. 5a & b) are defect-free and well-bonded, and display reaction layer formation. No evidence of cracking was observed in the joint. Identical joints with Ta in place of W led to unacceptable joints that exhibited defective interfaces and cracks.

Figure 5 shows two other multilayer joints: Si_3N_4/Cu-ABA/Ni/Cu-ABA/W/Cu-ABA/Si_3N_4 (Figs. 5c) and Si_3N_4/Cu-ABA/Ni/Cu-ABA/W/Cu-ABA/Inconel (Fig. 5d). The first joint, Si_3N_4/Si_3N_4, with single layers of Cu-ABA, Ni and W yields an excellent joint with no evidence of interfacial defects and/or cracks. The second joint, Si_3N_4/

Inconel-625, is also of acceptable quality although there is evidence of a few voids in the joint region.

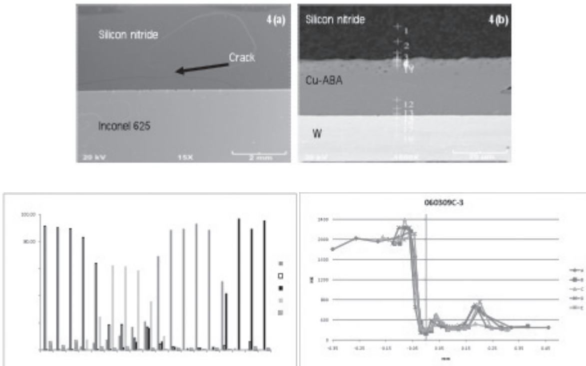


Fig. 4(a) Overview of a Si_3N_4/Cu-ABA/W/Cu-ABA/Mo/Cu-ABA/Inconel joint, (b) & (c) Si_3N_4/Cu-ABA interface and the corresponding elemental composition, and (d) Knoop microhardness distribution across the joint.

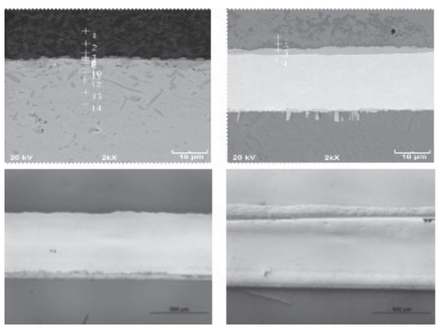


Fig. 5(a) & (b) Si_3N_4/Cu-ABA/Kovar/Cu-ABA/W/Cu-ABA/Kovar/MBF-20/Inconel joint showing (a) Si_3N_4/Cu-ABA and (b)W/Cu-ABA interfaces, (c) & (d) optical photomicrographs of a (c) Si_3N_4/Cu-ABA/Ni/Cu-ABA/W/Cu-ABA/Si_3N_4 joint, and (d) a Si_3N_4/Cu-ABA/Ni/Cu-ABA/W/Cu-ABA/Inconel joint.

Interestingly, increasing the number of Cu-ABA braze foils from one to two at each location within the joint of Fig. 5d yielded poor quality joints. Likewise, adding a Ni interlayer toward the Inconel side in the following arrangement also yielded a microstructurally poor-quality joint: Si_3N_4/

Cu-ABA/Ni/Cu-ABA/W/Cu-ABA/Ni/Cu-ABA/Inconel. However, the same joint produced excellent results when the number of Cu-ABA layers was increased from one to two; the joint was well-bonded and defect-free (although a small crack formed in the silicon nitride). These empirical results suggest that braze layer thickness must be controlled in relation to the type, number and arrangement of the ductile metal interlayers in order to achieve sound bond quality.

Overall, interlayers of Ta and W; Ni and W; Kovar (390 μm thick) with W; and of Ni, W, Ni all joined with the substrates of Si_3N_4 and Inconel-625. The two that were composed of Si_3N_4 bonded to Si_3N_4 were without cracking and had excellent bonding. The combinations that had a substrate of Ti did not bond well except when W and Ta were used in conjunction with two Cu-ABA foils. Other possible combinations of the same interlayer materials should be examined (e.g., Ni and Ta). All such empirical observations should be i) organized and evaluated to identify interlayer configurations that yield acceptable joining response, and ii) examined in light of the theoretical models of ceramic/metal bonding behavior. In addition, brazing runs should be made in progressively more complex (e.g., non planar) configurations to test the effectiveness of selected configurations to joining real components. Finally, extensive testing for joint strength and other properties (fracture toughness, corrosion resistance) should be carried out at room- and elevated temperatures to provide the designer reliable processing-properties database.

Conclusions

A CuSiAlTi active braze was used to join the Ni-base superalloy Inconel-625 and titanium to St. Gobain silicon nitride (NT-154) for NASA's Subsonic Rotary Wing project. An attempt was made to manage residual stresses during joining by judiciously arranging interlayers of Ni, W, Mo, Ta, Cu, and Kovar within the joint. The vacuum brazed joints were evaluated for metallurgical structure, elemental distribution, and microhardness. The results showed that interlayers of Ta and W; Ni and W; Kovar and W; and Ni, W, Ni led to sound joints between Si_3N_4 and Inconel-625 or Ti. Micro-cracking within Si_3N_4 occurred in some systems without impairment of joint integrity. Evidence of interfacial reaction layers with relatively large Ti and Si concentrations points toward chemical bonding due presumably to titanium silicide formation which bonded well to both the alloys and Si_3N_4.

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