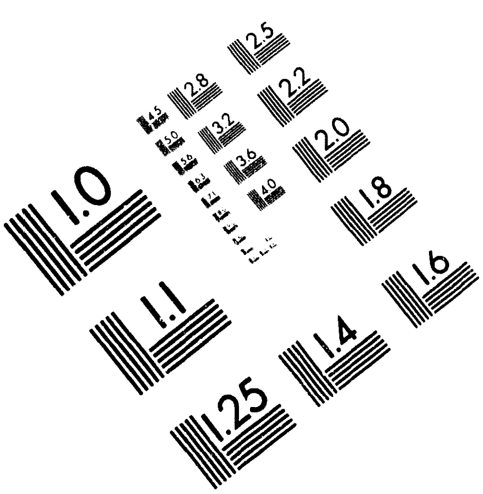

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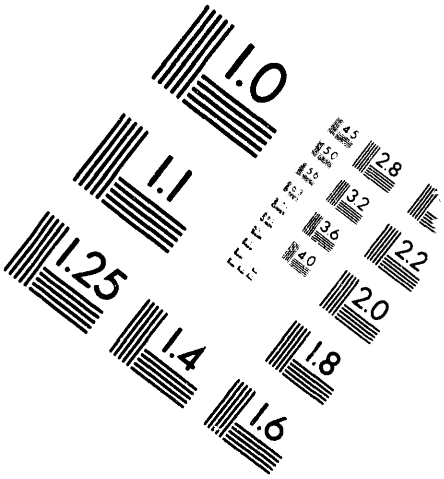

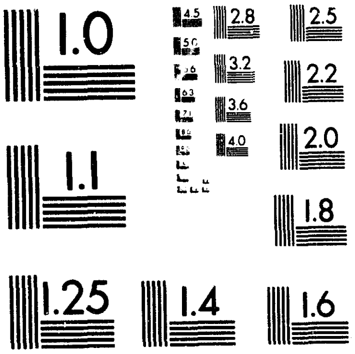

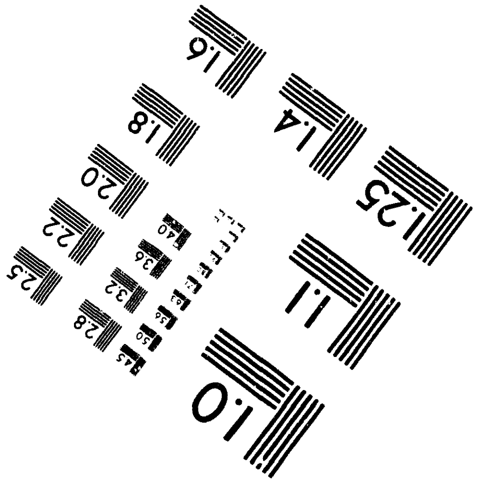

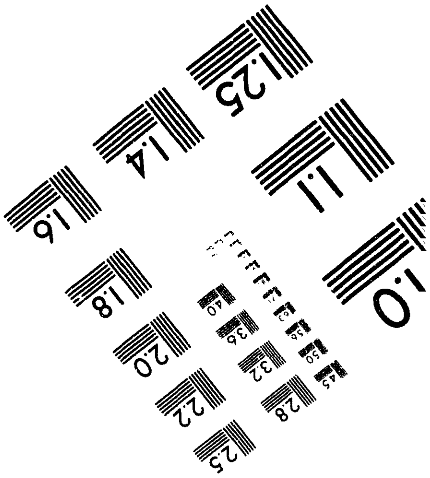

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STRENGTH TESTING OF Ti-VAPOR-COATED SILICON NITRIDE BRAZE JOINTS

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ABSTRACT

Sintered silicon nitride was vacuum brazed to itself at 1130°C with a Au-25Ni-25Pd wt% filler metal. Wetting was obtained by coating the Si_3N_4 surfaces with titanium prior to brazing by electron beam evaporation. The brazed joints were virtually free of porosity. Metallographic analysis showed that Ti reacted with the Si_3N_4 to form a TiN reaction layer during brazing. Small amounts of Si and Ti dissolved in the filler metal layers but they did not appear to influence the mechanical properties of the braze layer. Flexure bars were made from the brazed coupons and tested at room temperature, 600°C, 700°C, and 800°C in air. At 700°C and below, fracture of the test bars occurred in the Si_3N_4, either near the brazed surfaces or at some distance into the monolithic material. The measured strength of joint specimens decreased slightly with increasing test temperature, and generally exceeded the intrinsic braze filler metal strength in this temperature range. It was also found that lapping the Si_3N_4 prior to Ti coating reduced the number of near-surface flaws and produced joints with higher average strength and lower scatter than those left in a ground condition. Specimens tested at 800°C had very low strengths, and this behavior was related to the microstructure at the brazed Si_3N_4 surfaces.

INTRODUCTION

Ceramic joining, especially ceramic-to-metal joining, has been the subject of much developmental research over the years. However, with the current interest in using ceramics as structural components in such demanding applications as internal combustion engines, turbine engines, and heat exhangers, there has come a heightened interest in ceramic joining technologies. The reasons for the interest in joining ceramics are the same as those for joining metals, however, the development of effective ceramic joining techniques could have a much greater impact on their use in mass produced components. One of the most important functions of joining techniques is to provide the means for economic fabrication of complex, multi-component structures. Development of effective ceramic joining techniques will be especially significant because of limitations imposed on component manufacturing by ceramic processing techniques and by the materials themselves. For example, deformation of densified ceramics to form complex shapes is practically impossible owing to the fact that most ceramic materials are brittle even at elevated temperatures. Also, in some development programs like those for advanced heat engines some complex parts are being made as monoliths by difficult processing schemes or by extensive machining of densified billets. While, this approach to component manufacturing is acceptable for development purposes, it is undesireable for mass production because of high costs. Ceramics are also difficult, and therefore costly, to machine, and by reducing the complexity of individual parts significant

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savings in machining cost can be expected. The ability to effectively join ceramics may eliminate machining altogether in some cases.

Effective ceramic joining techniques can also play an important role in improving the reliability of ceramic structures. Because ceramics are brittle materials they are very sensitive to flaws due to the quality of raw materials used in their production, and to the characteristics of various processing techniques, including machining. A single flaw can cause the rejection, or, if undetected, the failure of a ceramic part. Rather than dealing with complicated monolithic parts, it is easier to inspect and detect flaws in simple-shaped components before they are joined to form complex structures.

Of the many techniques available for joining ceramics,[1-4] brazing was selected for the present study. Brazing is a liquid-phase bonding process with characteristics that: (1) only the filler material, but not the parts being joined, is melted during bonding; and, (2) the filler material must wet the base materials and be drawn into or held in the joint by capillary forces. Traditionally, the term "brazing" is used for processes where the filler material is metallic, but, in general, non-metallic materials such as glasses can also be used as filler materials. Ideally, the process is accomplished by assembling the component and filler materials, and then uniformly raising the temperature of the assembly to a point where the filler material becomes molten. If wetting conditions are favorable, the liquid filler material will be drawn into an intentionally made gap (i.e., the joint gap) between the parts being bonded, or it will remain in the joint gap if preplaced. Generally, some chemical reaction or mass transport occurs between the liquid filler material and the base materials, and, upon cooling, the filler material solidifies to produce a bonded part.

The brazing process has some significant advantages compared to other joining techniques. For instance, because melting of the base materials is avoided joining can be accomplished over a wide temperature range depending on filler material composition. This characteristic also makes it possible to join ceramics by brazing. Also, the use of a liquid bonding phase means that minor variations in surface flatness, surface topology, and part fit-up tolerances can be accommodated with relative ease. Other desireable features of the process are that: thermally induced stresses can be minimized by heating components uniformly in a furnace; and, close assembly and finish tolerances can be maintained which can reduce the need for costly secondary finishing operations. Lastly, the equipment and processing requirements for brazing are usually simple and straightforward, e.g., furnace heating in vacuum or controlled atmosphere.

Of course, brazing ceramics with metallic filler materials has drawbacks. One is that the full high temperature and strength capabilities of some ceramic components may by compromised by the presence of metallic braze layers. Another is that many ceramics are wetted only with difficulty by liquid metals including most commercially produced braze filler metals. This means that identifying or developing filler metals suitable for particular ceramics and their expected service conditions can often be problematic.

Generally speaking, the problem of wetting ceramic surfaces can be overcome by two general methods: (1) alloying braze filler metals with elements that activate wetting; and, (2) applying coatings that promote wetting to the oxide surfaces prior to brazing. The ability of Ti alloying additions to promote wetting of ceramics by liquid braze filler metals is well documented and effective.[5-12] This approach, often referred to as active metal brazing, has the advantage of being a relatively straight forward process, but it is not widely used because of the limited commercial availability of active braze filler metals. In contrast, the use of metallization to promote wetting of ceramics is a widely-used, well established practice that has several variations.[1] Metallized ceramic surfaces can be brazed with standard commercial braze filler metals so that this approach offers considerable flexibility for selecting joining materials which are compatible with processing or joint property constraints. Although it is not used often as a pretreatment for brazing, a common method of metallizing ceramic surfaces is vapor coating. The success of vapor coatings as a means of promoting the wetting of oxide surfaces by liquid metals was demonstrated some time ago,[13,14] but was not exploited for brazing of ceramics until more recently.[15-20]

This report summarizes the initial results of a program of strength testing of sintered Si_3N_4 braze joints. Wetting of the Si_3N_4 was accomplished by vapor coating its joint surfaces with Ti prior to brazing. Titanium was selected for a coating material because it was previously found to be effect for producing zirconia braze joints. Details of preliminary Si_3N_4 brazing studies are reported elsewhere.[19,20]

MATERIALS AND EXPERIMENTAL DETAILS

Sintered Si_3N_4 was used for this work. It was procured from Kyocera Corporation, and its commercial designation is SN-220. Selected properties of SN-220, as quoted by the manufacturer, are given in Table 1. The chemical composition of the SN-220 was not available from the manufacturer and was not determined, but it reportedly[21] relies on sintering aids of Al_2O_3 and Y_2O_3, and also contains particles of WC which are artifacts from a milling operation. This material was selected because it was judged to be representative of sintered silicon nitrides being evaluated for various heat engine applications, and because it was readily available and relatively inexpensive. The SN-220 material was supplied in billets with dimensions of either 150 x 10 x 5 mm or 25 x 15 x 3 mm. The 150-mm-long bars were cut into 10 x 10 mm coupons for brazing trials and metallographic specimens. The 25 x 15 billets were used for making coupons for braze joint flexure testing.

Because other studies[22] have shown that the surface condition of ceramics can influence joint properties, brazed coupons for room temperature testing were prepared with three different finishes on the joint surfaces: as-ground, 30-µm-finish, and lapped to a 1-µm-diamond finish. The as-ground condition was the finish supplied by the manufacturer. The 30-µm-finish was produced by manually diamond grinding joint surfaces on a metal disc impregnated with diamond particles 30 µm in size. The lapped finish was prepared by metallographic polishing techniques.

Prior to brazing, all Si_3N_4 joint surfaces were coated with a 1-µm-thick layer of Ti by vacuum evaporation. The Ti vapor was produced by melting a pure Ti source held in a water-cooled Cu crucible with an electron beam melting system operating at 7 kV and 100 mA. Typically, the pressure inside the evaporation chamber was 400 µPa at the time melting of the Ti commenced, and it eventually dropped to near 50 µPa during evaporation. The Si_3N_4 specimens were shielded from the Ti vapor until the pressure inside the chamber stabilized at the lower level. A standard quartz oscillation technique was used to monitor coating thickness during evaporation, and thickness was verified by the weighing of reference tabs. A typical time to produce a 1-µm-thick coating was 30 min. The Si_3N_4 specimens also were heated with quartz lamps to the range of 250-300°C prior to initiating the coating process. The Si_3N_4 specimens were rinsed with acetone followed by ethyl alcohol and then air dried before being placed in the evaporation chamber.

The filler metal used for making the braze joints has a nominal composition of Au-25Ni-25Pd wt%. This alloy is commercially available in a variety of forms (AWS standard designation: BVAu-7 Gr1). A 25-µm-thick foil was used for brazing the Si_3N_4 coupons. Typically, a piece of filler metal foil was cut to match the interfacial area of the braze joint and preplaced between the Si_3N_4 coupons before heating to the brazing temperature. The liquidus temperature of the filler metal is near 1120°C, and a brazing temperature of 1130°C was used for all experiments.

All brazing was done in vacuum. Joint assemblies were placed in an alumina tube which was subsequently sealed and evacuated to a pressure of about 130 µPa. Afterwards, the tube was inserted into an air muffle furnace preset and equilibrated at the brazing temperature. A thermocouple positioned near each joint assembly was used to continuously monitor temperature during the brazing thermal cycle. The holding time at the brazing temperature was normally 20 min, after which the alumina tube was withdrawn from the furnace and cooled to room temperature. Joint assemblies for metallographic examination were secured together with Mo wire. The assemblies made for flexure testing were held on edge in a graphite fixture with the joint interface in the horizontal plane. No load was applied to any of the assemblies during brazing.

Coupons used for flexure testing were made by brazing pairs of 25 x 15 x 3 mm together along the 25 x 3 mm edges. Afterward, the brazed coupons were surface ground on both 30 x 25 mm faces, polished to a 1-µm-diamond finish on one face, and cut into flexure bars with dimensions of 30 x 2.5 x 2 mm. Each bar had a braze layer centrally located along its length and oriented perpendicular to its axis. The long edges on the tensile faces of each bar were chamfered. Testing was done by four point bending at a crosshead displacement rate of 0.25 mm/s with the polished side of each bar being loaded in tension. The flexure test fixture had an outer span of 19.05 mm and an inner span of 6.35 mm. The tests were done in air at temperatures of 25°C, 600°C, 700°C and 800°C. For the elevated temperature tests, the bars were equilibrated for 30 min prior to loading.

Microstructures were examined optically and with a scanning electron microscope (SEM). Standardless, semiquantitative microchemical analyses were carried out in the SEM using an energy-dispersive x-ray spectrometer (EDS) system.

RESULTS

Joint Microstructure: The overall appearance of the Si_3N_4 braze joints is typified by the cross sectional view shown in Fig. 1(a). Generally, the joint gaps were uniformly filled by the filler metal, there were no indications of cracking in the Si_3N_4 near the joints, and there was very little porosity in the braze layers. Microstructural details at the Ti-vapor-coated Si_3N_4 surface are shown in Fig. 1(b). After brazing, the filler metal layer contained two major phases: a Au-rich matrix and irregularly-shaped Ni-rich particles. The presence of the Au-rich and Ni-rich phases is consistent with the solid-state phase separation observed for both the

Au-Ni and Au-Pd binary systems.[23] The overall composition of the filler metal layer, and those of the individual phases are given in Table 2. The overall composition is reasonably close to the nominal composition of Au-25Ni-25Pd wt%, but the presence of Si in the filler metal layer after brazing indicates that some reaction with the Si_3N_4 has occurred. A Vickers indenter under a 100 g load was used to measure the hardness of the filler metal layer at 2.52 GPa, and based on these measurements the yield strength of the braze filler metal was estimated to be 265 MPa at room temperature.

The interface region between the filler metal and the Ti-vapor-coated Si_3N_4 is also shown in Fig. 1(b). After brazing, the layer at the Si_3N_4 surface was still about 1 µm thick and Ti-rich, but reaction of the Ti-vapor-coating was evident and Ti was detected throughout the braze layer. The Ti-rich layer was studied in more detail by dissolving away the filler metal in an acid solution, and examining the exposed reacted surface in the SEM and by x-ray diffraction (XRD). The surface revealed by this approach was composed of fine cuboidal grains and occasional larger smooth regions as shown in Fig. 2. Microchemical analysis indicated that the cuboidal grains contained Ti with trace amounts of Al, Au, Ni, Pd and Si, while the smooth regions were Au-rich indicating they were filler metal fragments. Analysis by XRD showed that the only phase present besides Si_3N_4 was TiN. These observations show that the Ti-vapor-coating reacted with the Si_3N_4 during brazing to form a TiN layer.

Room temperature flexure testing: Data from the room temperature tests are summarized in Table 3, and are plotted in Figs. 3-5 using a Weibull distribution function and assuming the data for each surface condition represent a single population. The straight lines and Weibull modulus values, m, were determined by separate linear regressions.

Comparison of Fig. 3 for the as-ground joint finish and Fig. 5 for the 1-µm-diamond finish indicates that because of a slightly higher average strength and less spread in the strength values, there was an advantage to using the polished joint surfaces over the as-ground surfaces. As noted in Tables 3, many of the specimens from these two groups actually fractured in the Si_3N_4.

Comparing Fig. 4 to Figs. 3 and 5 shows that the 30-µm-diamond joint surface finish resulted in lower joint strengths and more scatter than was found for joints with the other two surface finishes. All of the joints with the 30-µm finish broke near or at the interface between the Si_3N_4 and the braze layer. The erratic behavior of these joints compared to those with the as-ground and 1-µm-diamond joint surface finishes suggests this strength data is spurious, and this behavior was attributed to improper cleaning of the Si_3N_4 surfaces after grinding to the 30-µm finish and before Ti vapor coating.

Flexure tests at 600° C: The results for flexure testing joint specimens at 600° C are given in Table 3, and are plotted in Figs. 6 and 7. The data for specimens with ground joint surfaces, Fig. 6, show that testing at 600° C produced lower average strength and slightly more scatter (σ_f = 376 MPa, m = 6.8) when compared to room temperature test results (σ_f = 405 MPa, m = 7.6). Examination of the test bars also indicated that the higher test temperature resulted in a greater tendency for fracture to occur in the Si_3N_4 near the joint surfaces compared to the room temperature tests.

In the cases where the joint surfaces were lapped prior to Ti vapor coating and brazing, the higher test temperature also resulted in slightly lower average joint strength but more scatter (σ_f = 405 MPa, m = 6.7) compared to room temperature data (σ_f = 418 MPa, m = 11.2). For these joints, however, testing at this temperature did not significantly affect the failure location tendency of the joints. At 600°C, over half of the test bars fractured in the Si_3N_4 away from the joint surfaces.

Flexure tests at 700°C & 800°C: The results for flexure testing at these two temperatures are given in Table 4, and the data from tests at 700°C are plotted in Fig. 8. Because more favorable strength characteristics were associated with lapped joint surfaces, this was the only surface finish condition used at these two temperatures.

Figure 8 shows that the 700°C strength data fit two distributions. The average joint strength values were: σ_f = 308 MPa, m = 15.9 in the lower distribution and σ_f = 410 MPa, m = 9.4 in the upper distribution. The bimodal nature of the data was verfied by grouping it into two sets, one containing 814 & 816 and the other containing 815 & 817, and then applying the *"Student's" t* analysis to the respective mean and standard deviation values. Reference to Table 4 shows that the data sets were differentiated based on braze layer thickness as measured on the tensile faces of the test bars.

As the nominal thickness of the braze filler metal was 25 µm the source of the wide variation in braze layer thickness was at first problematic. No attempt was made to intentionally influence braze layer thickness, and experience had shown that thicknesses near 25 µm were typically obtained. However, examination of untested bars indicated that joint thickness was not uniform. The lapped joint surfaces were misoriented slightly (about 1°) from 90° relative to the 25 x 15 mm surfaces on the individual Si_3N_4 coupons. This in turn resulted in a maximum variation in braze layer thickness from about 10-µm thick on one 30 x 25 mm face of the braze coupons to about 50 µm on the other. The test bars from coupons 814 & 816 had their tensile faces on the side where the increased braze layer thickness was observed, while those from 815 & 817 had their tensile faces on the side of reduced braze layer thickness. Regardless of the source of thickness variation, the 700°C data suggest that braze layer thickness may have a significant influence on joint strength. These data also show that excellent joint strength was maintained at this temperature.

All of the bars heated to 800°C broke under the preloading conditions, so only a limited number of tests were attempted at this temperature as indicated in Table 4.

Fractography: The tensile face of each flexure bar was examined optically after testing to assess where failures occurred, and these results are given in the data tables. Some of the joint specimens clearly fractured entirely in the Si_3N_4, often several millimeters away from their braze layers. Other specimens failed very near, Fig. 9(a) or along, Fig. 9(b), the interface between the filler metal layer and the Si_3N_4, and these are noted as joint failures. Examination of a more select sample of the joint specimens in the SEM showed that, in many cases, failures which initiated near Si_3N_4/braze layer interfaces propagated largely through the ceramic. Testing temperature did not have a strong effect on the overall fracture characteristics at 700°C and below. Also, failure in or significant elongation of the braze filler metal layer was not observed even at 700°C.

Several of the joint specimens tested at 800°C were examined in the SEM, and the micrograph shown in Fig. 10 is representative of their fracture surfaces. The granular appearance suggests that some liquid phase was present at the test temperature, or that the fracture surfaces oxidized extensively during cooling. Microchemical analyses from area scans on the surface shown in Fig. 10 indicated that the average Si concentration was in the range of 11 wt% (23 at%). Significant levels of Ni, Pd and Ti were also found. These observations suggest that liquation occurred between 700°C and 800°C and that this behavior was caused by a Si-rich or a silicide phase.

DISCUSSION

Metallographic examination showed that the Au-25Ni-25Pd wt% filler metal was effective for producing sound, defect-free braze joints of Ti-vapor-coated Si_3N_4. A Ti-rich layer was found at the Si_3N_4 surfaces after brazing, but Ti was also detected throughout the braze layers indicating that some dissolution in the filler metal occurred. Microchemical analysis also detected small concentrations of Si in the braze layer suggesting that reduction of the Si_3N_4 occurred during the brazing operation. Titanium forms a stable nitride and silicides and because the Ti was in direct contact with Si_3N_4 reaction between them was likely. The possible reactions for which thermodynamic data are available[24,25] are:

$$1/5Si_3N_4 + Ti = 1/5Ti_5Si_3 + 2/5(N_2) \qquad \Delta G^0_T = -14.7 \text{ kcal/mole} \qquad [1]$$

$$1/3Si_3N_4 + Ti = 1/3TiSi + 2/3(N_2) \qquad \Delta G^0_T = -16.9 \text{ kcal/mole} \qquad [2]$$

$$2/3Si_3N_4 + Ti = TiSi_2 + 4/3(N_2) \qquad \Delta G^0_T = +13.5 \text{ kcal/mole} \qquad [3]$$

$$1/4Si_3N_4 + Ti = TiN + 3/4Si \qquad \Delta G^0_T = -32.1 \text{ kcal/mole} \qquad [4]$$

These data show that TiN, Ti_5Si_3, and TiSi can all form by reaction of titanium with Si_3N_4 at 1130°C, but that TiN is considerably more stable than the titanium silicides. The XRD data confirmed that a layer of TiN formed preferentially at the Si_3N_4 surfaces during brazing. The lack of porosity in the braze layers also offers indirect evidence for the preferential formation of TiN. If silicide reactions were preferred, they would have released N_2 which would have formed pores in the liquid filler metal.

Other phases such as Ni, Pd, or Ti silicides or Ni_xTi compounds were not directly observed by SEM or XRD. However, their presence cannot be precluded. Quantities of these compounds too small to be detected may have formed in the braze layer microstructures. It is also possible that low-melting compounds incorporated into the reaction layer at the Si_3N_4 surfaces were dissolved by the acid solution used to reveal the TiN.

Microchemical analysis further showed that Si partitioned preferentially to Ni-rich particles found in the braze layer. The unmelted filler metal is ductile, and the absense of cracking in the reacted filler metal layers indicates that the dissolution of the Si and Ti did not seriously compromise braze layer ductility. The lack of cracks around hardness indentations also suggests that ductility was maintained in the braze layers.

The flexure tests illustrate several important points about the strength of the Ti-vapor-coated braze joints. The first is that joint strength was not limited by the braze layer strength at temperatures up to 700°C, even though the intrinsic strength of the metallic layer is much lower than that of the Si_3N_4. Four-point flexure testing imposes a tensile stress on the outer faces of test bars, which in the present study is also normal to the braze filler metal layers. The problem of loading a thin soft layer in tension between two stronger components has been analyzed by Dalgleish el al.[26,27] who showed that substantial hydrostatic stresses develop in the thin layer. The limiting imposed stress σ_L that such an arrangement of materials will support is described by the relation:

$$\sigma_L/Y = 3/4 + 1/4(w/t) \qquad [5]$$

where Y is yield strength of the layer material, w is the layer width, and t is layer thickness between the higher strength components. For the Ti-vapor-coated Si_3N_4 braze joints estimates of w and t are 3 mm and 25 μm, respectively, which means that $w/t \approx 120$ and $\sigma_L/Y \approx 30$. Consequently, the joints may support tensile loads 30 times greater than the yield strength of the braze layer which was estimated at 265 MPa. Clearly, according to this stress analysis, joint strength should not be limited by the braze filler metal strength, and failures in the Si_3N_4 should be expected. This is exactly the situation which was observed. The tendency for failures to occur outside of the braze layer even at 700°C must be taken as an indication that, even at this temperature, the conditions of equation [5] are satisfied and σ_L/Y exceeds the intrinsic strength of the Si_3N_4.

The flexure test data also show that the condition of the Si_3N_4 joint surfaces is an important factor in determining the joint strength characteristics. As compared to a ground finish, lapped joint surfaces resulted in higher average joint strength and greater tendency for failure initiation in the Si_3N_4. At room temperature, lapped joint surfaces also resulted in a higher Weibull modulus. Based on the joint strength data from room temperature and 600°C it is concluded that removing at least 50 μm of material from Si_3N_4 surfaces by a process which involves minimal damage and chemical residues is recommended for improving the overall strength characteristics of the joints.

To learn more about the effects of finish condition, both ground and lapped Si_3N_4 surfaces were examined by an acoustic surface wave technique. Using 50 MHz acoustic surface waves, the defects greater than about 25 μm in size and within about 100 μm from the outer specimen surfaces were detected. The results of this examination, shown in Fig. 11, verified that lapping greatly reduced the population of near-surface flaws in the Si_3N_4. Figure 11(a) shows the result of examining a ground surface where near-surface defects produced by the grinding process are clearly visible as light areas. Lapping removed a thickness of about 50 μm from the Si_3N_4 surfaces. The effect of this treatment on the near-surface flaw population in shown in Fig. 11(b), where a significant decrease in the number of near-surface flaws is evident. Undoubtedly, the lower number of near-surface flaws at the lapped joint surfaces reduced the probability of failure initiation in those regions relative to that for failure in the Si_3N_4.

The data from testing at 700°C also imply there were two flaw populations in the test bars which resulted in two substantially different failure modes. Because all of the joint coupons and test bars were were prepared by the same techniques, the flaw situation for the specimens tested at 700° C should have been similar to that of the lapped-joint-surface specimens tested at lower temperatures. A variable which unintentionally appeared in these specimens, however, was the variation in braze layer thickness on the tensile faces of the test bars. Joint strength is known to increase as braze layer thickness decreases,[26-28] a trend which is attributed to geometric factors and the mechanical properties of the layer material. This general trend is supported by the present data, but Fig. 8 strongly suggests that the braze layer thickness directly influenced the failure mode at 700° C. The analysis of metal-bonded ceramic joints[29] assumes the braze layer is uniform in thickness, and the slight misorientation found in these Si_3N_4 joint specimens may have influenced the distribution and intensity of stresses in the Si_3N_4 enough to activate the different failure modes. More study is required before firm conclusions can be made about this behavior, and such work should be pursued because effects of a nonuniform braze layer can have important practical implications.

More study will also be required before the loss of joint strength at 800° C can be satisfactorily explained. Silicon and Au form a simple eutectic system and are mutually insoluable in the terminal pure solid phases. Nickel, Pd and Ti, also form eutectic systems with Si. However, two or more intermetallic phases form in each of these binary systems, and eutectic temperatures of 964° C and 760° C occur respectively in the Ni-Si and Pd-Si systems. Liquation of phases in the braze layer due to the solution of Si then is a real possibility. The fracture surfaces of flexure bars tested at 800° C suggested that a liquid phase may have been present at this temperature. Also, the high Si concentration on the fracture surfaces supports the possibility of liquation. On the other hand, the true appearance of these surfaces may have been obscured, at least partially, by oxidation. Further examination of these specimens will be required before the cause of failure at 800° C can be determined.

SUMMARY AND CONCLUSIONS

Silicon nitride surfaces were vapor coated with Ti by electron beam evaporation in order to improve their brazing characteristics. Coupons of Ti-vapor-coated Si_3N_4 were then vacuum brazed together with a filler metal of Au-25Ni-2525Pd wt% at 1130° C. The braze joints had good overall appearance and were virtually free of porosity and lack-of-bond type defects. During the brazing operation, the Ti coatings reacted with the Si_3N_4 to form a TiN reaction layer at the original Ti/Si_3N_4 interfaces. Silicon produced by this reaction, as well as some Ti from the vapor coating dissolved in the braze filler metal layer. These reactions did not appear to significantly alter the braze layer microstructure or properties.

Brazed Si_3N_4 coupons were cut into flexure bars and tested at room temperature, 600° C, 700° C, and 800° C. The results of these tests support the following conclusions:

1. Joint strength was not limited by the braze filler metal strength at temperatures of 700° C and below. Joint flexure bars often failed in the monolithic Si_3N_4 at strength values far exceeding the yield strength of the braze filler metal. This behavior apparently was due to the development of a hydrostatic stress state in the braze filler metal layer which substantially increased its effective yield strength.
2. The condition of the Si_3N_4 surfaces which formed the braze joint had a significant effect on joint strength characteristics. A lapping operation was used to remove approximately 50 µm of material from some joint surfaces prior to Ti vapor coating and

brazing. The lapping operation significantly reduced the number of near-surface flaws in the Si_3N_4, and produced joints having higher average strength and reduced scatter compared to those left in a surface ground condition.

3. At 700°C and below, the fracture behavior of the joint specimens generally was consistent with that predicted by stress analyses[20] of ceramics bonded with ductile metal interlayers. Preferred failure initiation sites were in the Si_3N_4 either near or some distance away from the brazed surfaces.

Two other noteable observations were also made. First, joint strength at 800°C was minimal, and appeared to be limited by a phase or phases formed in the braze filler metal layer. Second, the thickness of the braze layer on the tensile face of the test bars appeared to have a significant effect on measured strength. Joint strengths were higher when the apparent braze layer thickness was reduced. Fracture in the Si_3N_4 away from the joint region also was favored under these conditions.

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Table 1. Selected properties of SN-220 Si_3N_4. Room temperature values are given unless otherwise noted.

Bulk Density g/cm^3	Flexure Strength, MPa					Weibull Modulus R.T.
	R.T.	800°C	1000°C	1200°C	1300°C	
3.2	588	598	510	323	118	7-15

Fracture Toughness MPa-√m	Hardness GPa	α_r, µm/m/°C 40-800°C	Thermal Conductivity W/m-K	Thermal Shock Resistance ΔT°C
6.2	14.7	3.2	20.9	500-600

Young's Modulus GPa	Poisson's Ratio	Oxidation Resistance, mg/cm^2 in 24h			
		1200°C	1250°C	1300°C	1400°C
294	0.28	0.2	0.6	0.8	1.9

Table 2. Compositions of phases in braze filler metal layer after brazing at 1130°C.

Braze Layer Analysis Location	Composition in wt%				
	Au	Ni	Pd	[illegible]	Si
Overall	50.43	23.60	23.51	2.07	0.41
Au-rich matrix	51.20	23.43	23.10	1.91	0.36
Ni-rich particles	34.77	43.01	19.20	1.73	1.29

Table 3. Results for flexure testing of Ti-vapor-coated Si_3N_4 braze joints at room temperature and 600°C.

Specimen Series No.	Surface Finish	Test Temp. °C	Number Tested	Strength Range, MPa	Average Strength, MPa	No. Failed at Joint
400	ground	25	18	293-496	405 ± 54	10
403	30 μm	25	18	195-440	340 ± 77	18
407	1 μm	25	14	345-464	418 ± 30	8
451	ground	600	18	260-516	376 ± 56	14
444	1 μm	600	17	292-487	405 ± 50	7

Table 4. Results of flexure testing Ti-vapor-coated Si_3N_4 braze joints at 700°C and 800°C. The 1 μm surface finish was used on all test bars.

Specimen Series No.	Test Temp. °C	Number Tested	Thickness of Braze, μm	Strength Range, MPa	Fracture Location
814	700	6	45-50	296-322	Joint
815	700	4	10-13	375-415	Si_3N_4
815	700	2	16-25	347-378	Joint
816	700	3	41-51	274-339	Joint
816	800	2	41	13-21	Joint
817	700	4	10-16	409-489	Si_3N_4
817	800	2	10-13	22-99	Joint

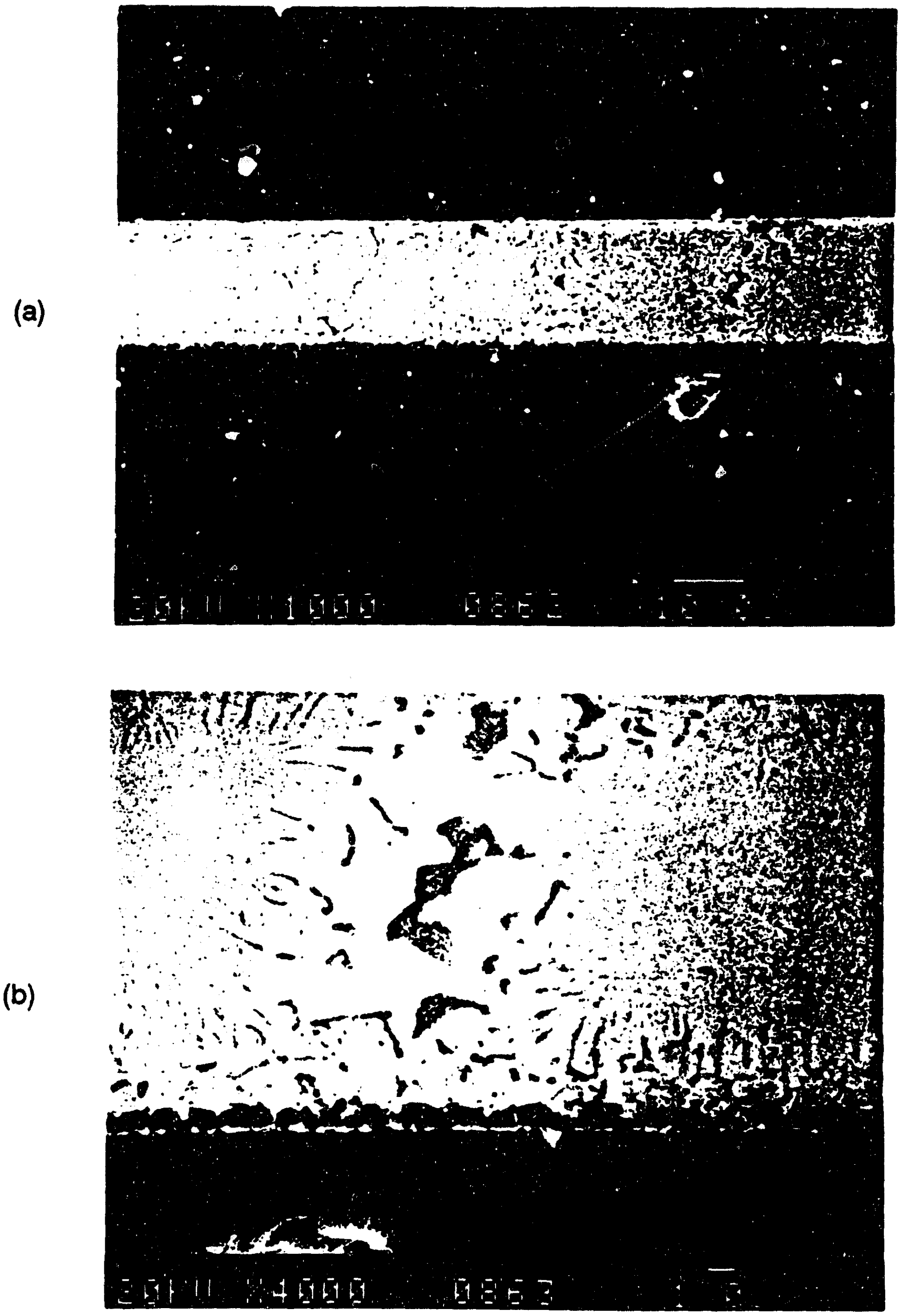

Fig. 1. SEM micrograph from a cross-sectioned joint showing the overall appearance of the Au-Ni-Pd filler metal layer between coupons of Ti-vapor-coated Si_3N_4, (a); and, the microstructure at a Ti-vapor-coated Si_3N_4 surface after brazing.

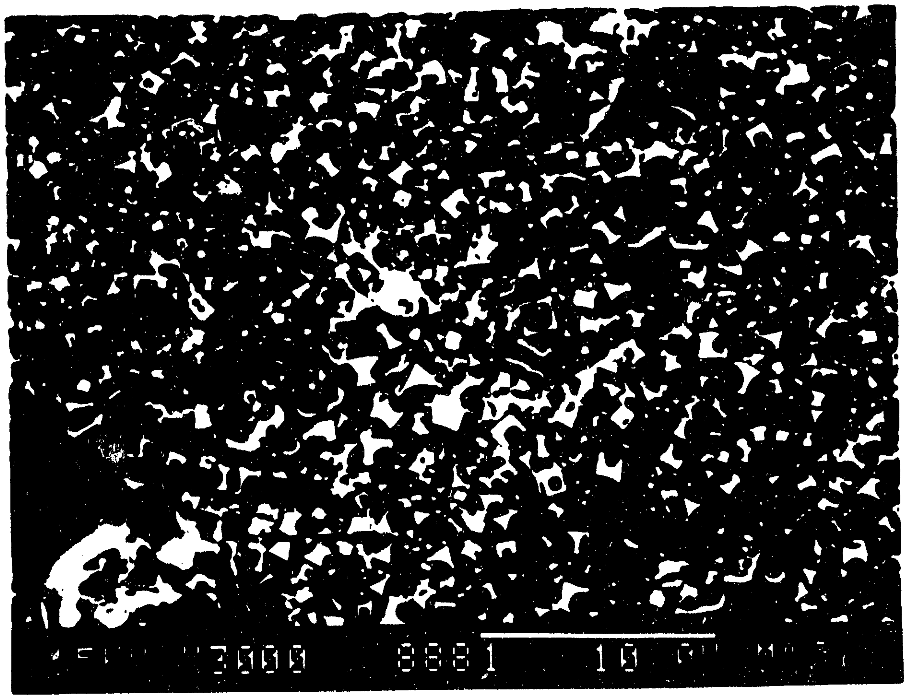

Fig. 2. SEM micrograph on a Si_3N_4 surface after brazing and removal of the braze filler metal by acid dissolution showing grains of TiN.

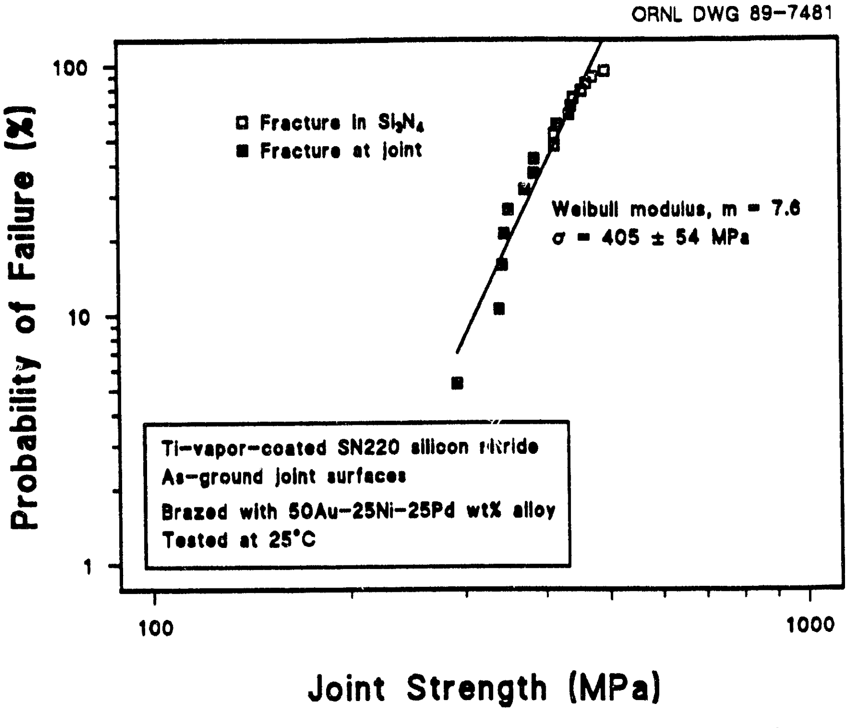

Fig. 3. Room temperature flexure strength of specimens taken from coupons in which the brazed surfaces had a ground finish.

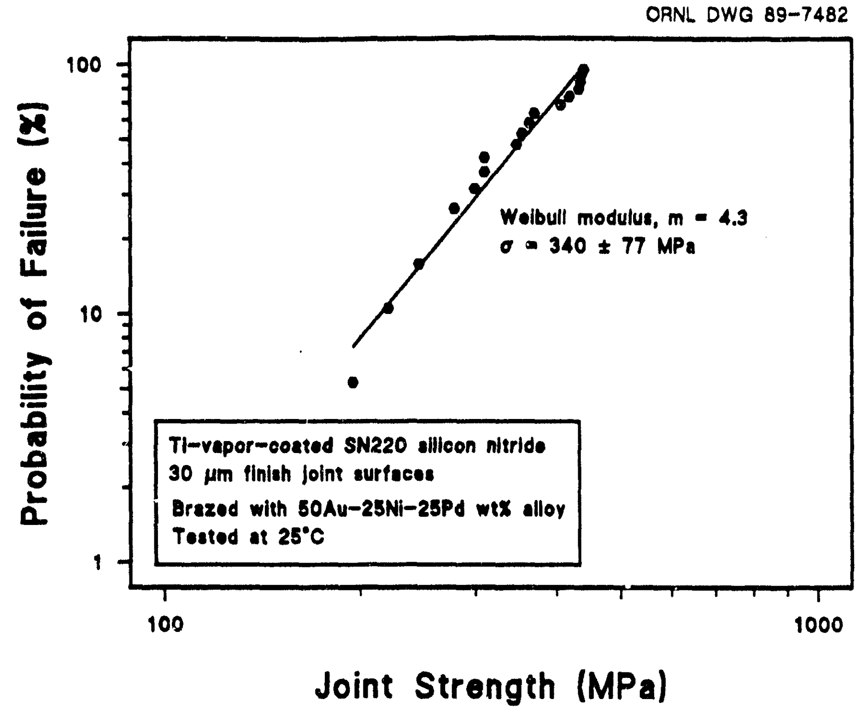

Fig. 4. Room temperature flexure strength of specimens taken from coupons in which the brazed surfaces were lapped to a 30-μm-diamond finish.

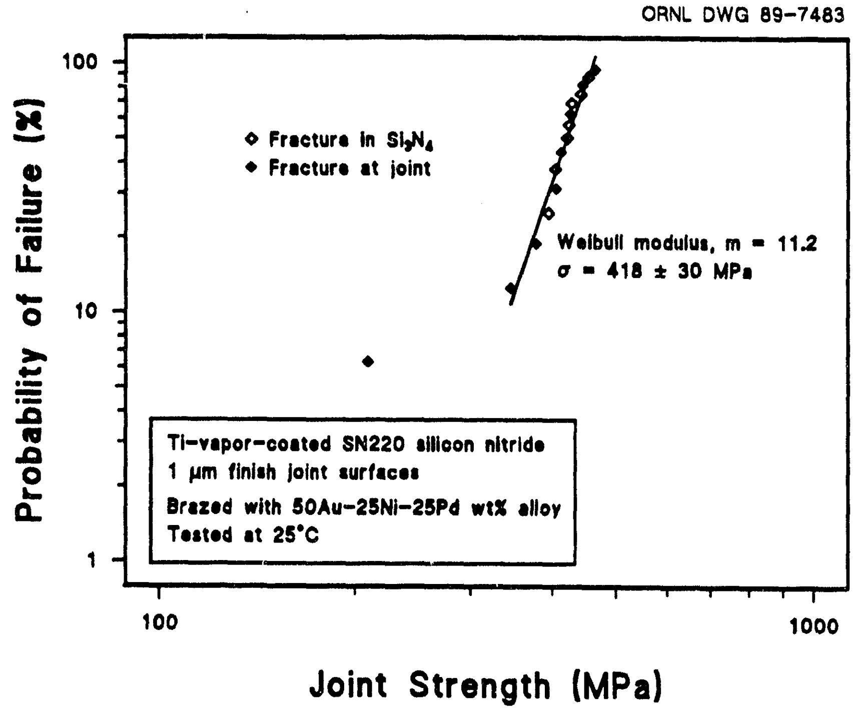

Fig. 5. Room temperature flexure strength of specimens taken from coupons in which the brazed surfaces were lapped to a 1-μm-diamond finish.

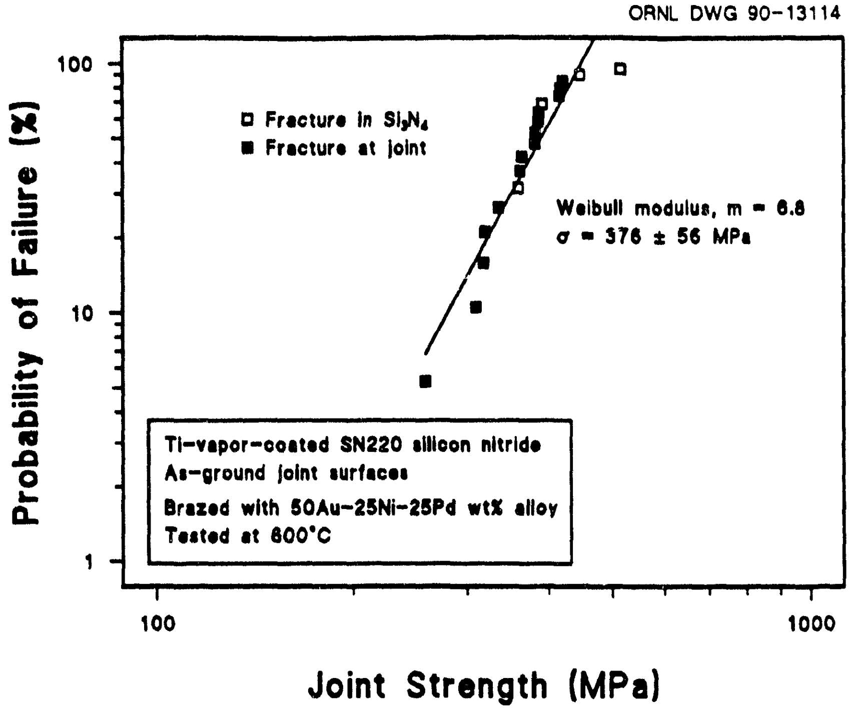

Fig. 6. Flexure strength at 600°C of specimens taken from coupons in which the brazed surfaces had a ground finish.

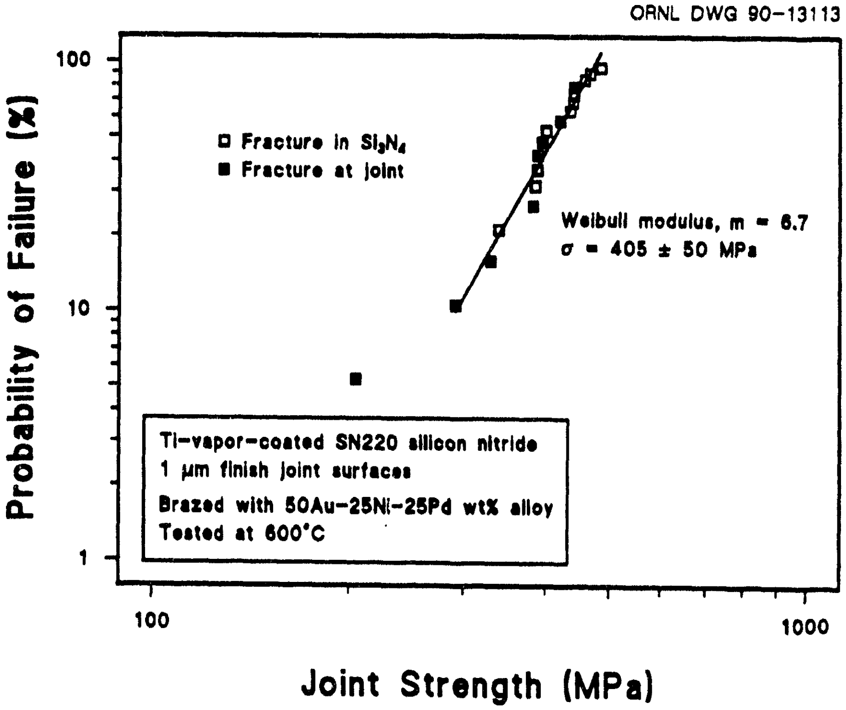

Fig. 7. Flexure strength at 600°C of specimens taken from coupons in which the brazed surfaces were lapped to a 1-µm-diamond finish.

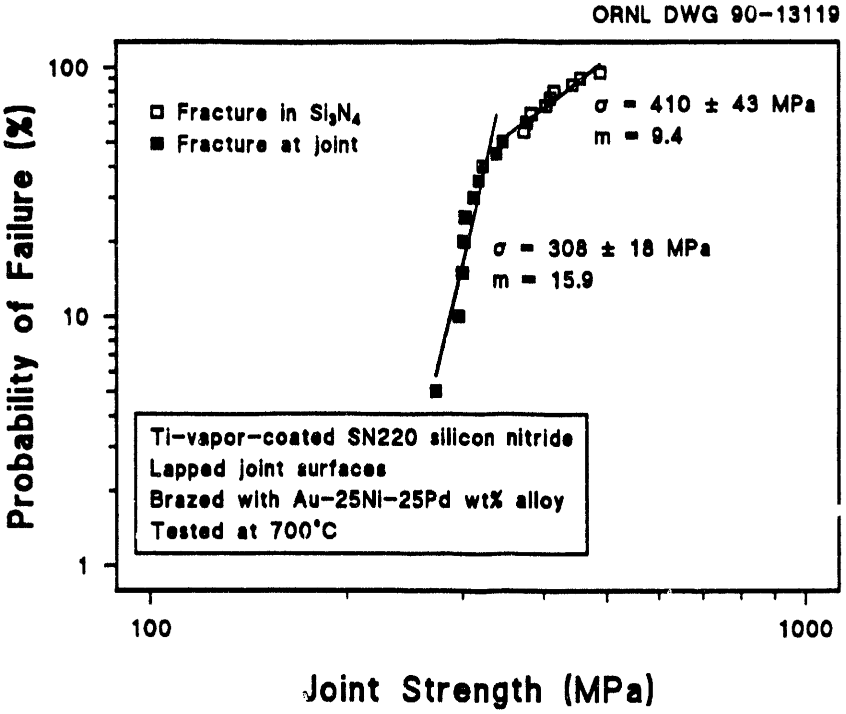

Fig. 8. Flexure strength at 700°C of specimens taken from coupons in which the brazed surfaces were lapped to a 1-μm-diamond finish.

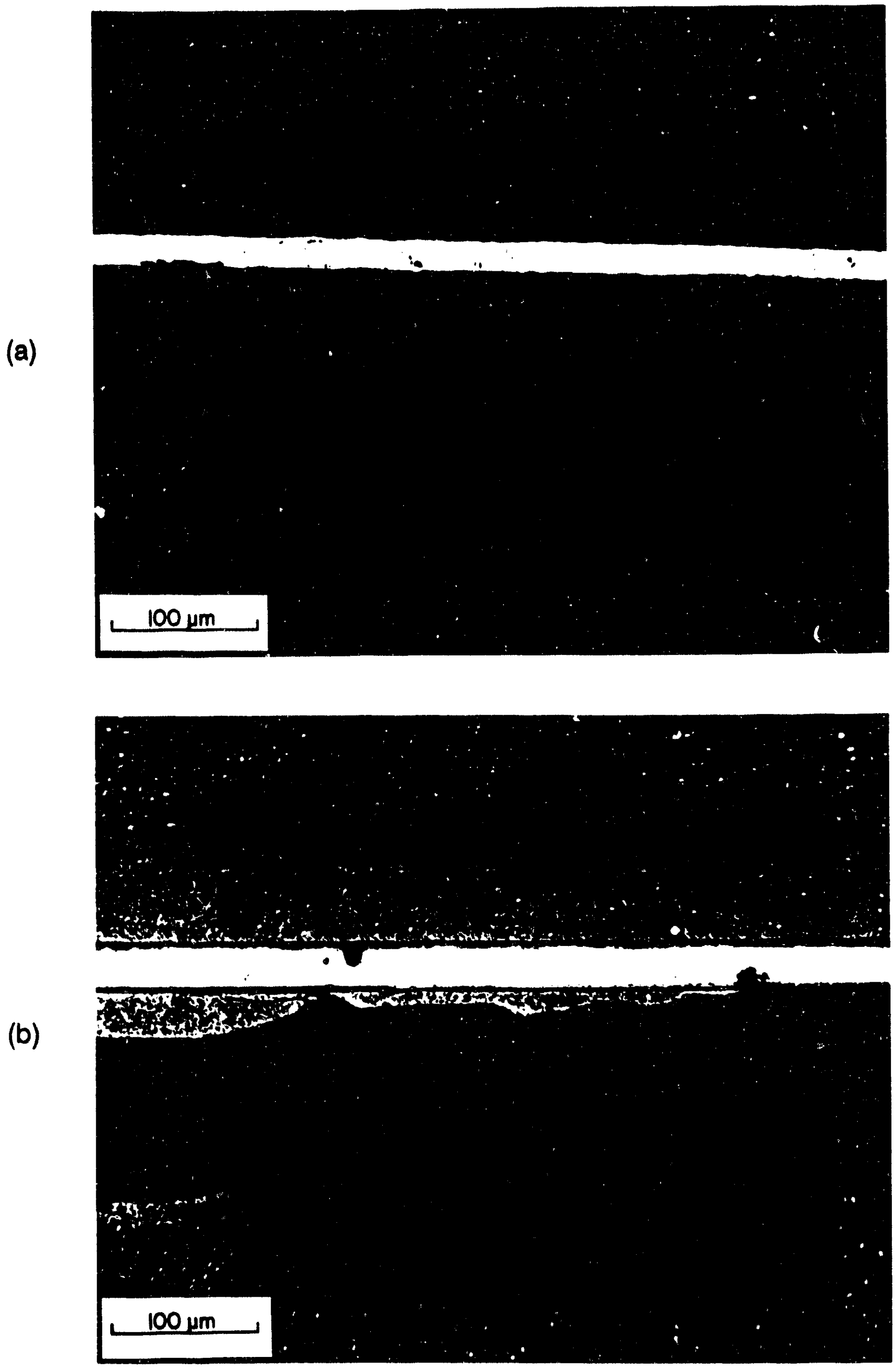

Fig. 9 Optical micrographs on tensile faces of flexure test bars showing failure at (a), and failure near (b) the interface between the Si_3N_4 and the braze filler metal layer.

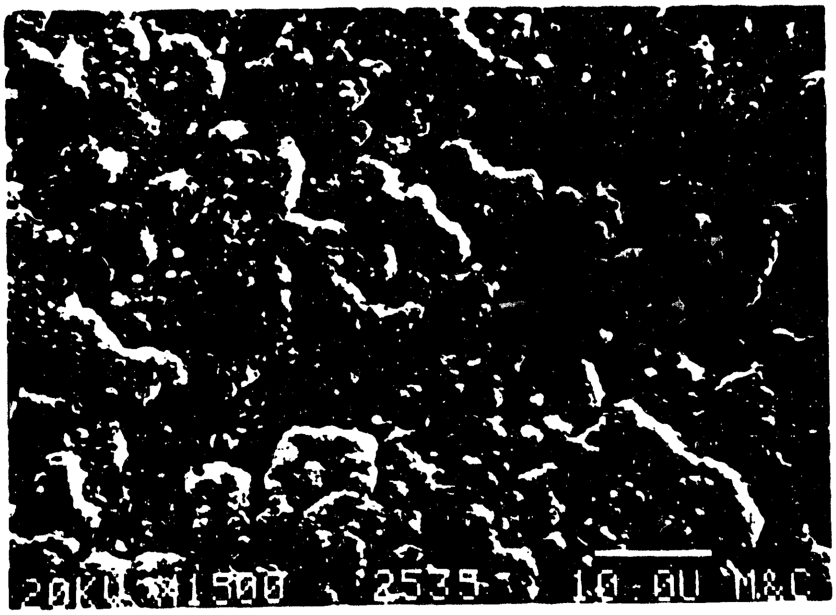

Fig. 10. SEM micrograph from a fracture surface of a braze joint flexure bar tested at 800°C.

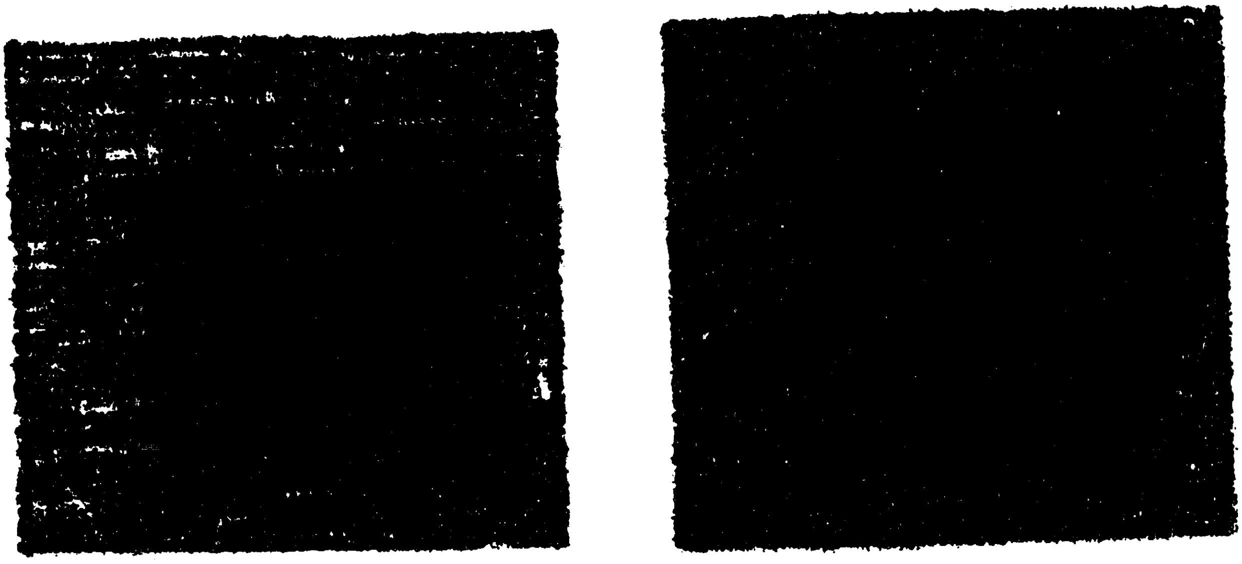
(a) (b)

Fig. 11. Results from examination of a ground (a) and a lapped (b) surface by an acoustic surface wave technique. Light areas represent regions containing near surface flaws.

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