Residual Stresses Measurement of Near Interface Region in Silicon Nitride and Stainless Steel Brazing Joint

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Abstract

High residual stresses are known to exist in the ceramic part near the ceramic-metal joint. However, in carrying out their measurement, we sometimes find it very difficult to get a tendency in stress distribution. One of the reasons for the difficulty seems likely to be the presence of residual stresses produced when ceramic material is sintered. These initial stresses are considered to affect the residual stresses caused by brazing.

In this study we measured both residual stresses before and after brazing and then examined if the measured stress values are proper as such. For this purpose, we compared the result of computer simulation, that of a 4-point bending test, and the measured stress values with one another.

As a result, we found out the following. (1) Over 180 MPa of residual stress was measured in the surface layer of the as-sintered silicon nitride. The area of the stress concentration was as small as below 1.0 mm in diameter. (2) The stress distribution obtained by subtracting the initial stresses before brazing from the residual stresses measured after brazing agreed with the simulation result qualitatively. (3) The tensile residual stress measured with a micro X-ray beam was about 270 MPa at maximum at a point 0.2 mm away from the interface. The strength of the silicon nitride decreased by 335 MPa due to brazing. (4) The cracking position roughly agreed with the point where the maximum principal stress was measured. The direction of cracking also agreed with the normal to the direction of the maximum principal stress.

Key words: Silicon nitride, Stainless steel, Brazing, Residual stress, Micro X-ray beam, X-ray stress measurement, Imaging plate, 4-point bending test

1. Introduction

It is generally known that when a bending test is conducted on a ceramic and metal brazing sample, a crack occurs in the ceramic part near the ceramic-metal joint. To investigate the stress distribution in the ceramic part near the interface, we measured the residual stresses there on earlier occasions and reported on the results. The results showed that in some cases it was very difficult to get the distribution of stresses caused by brazing because the measured values irregularly varied. In view of no machining work applied to the ceramics, we consider that one of the reasons is the pre-existing residual stress in ceramics due to sintering. Unless therefore such a pre-existing residual stress in ceramics is taken into account, it seems unlikely that a real stress due to brazing can be measured accurately.

In this study, we evaluated the residual stress due to brazing from this viewpoint by subtracting the pre-existing residual stress before brazing from the residual stress measured after brazing. The X-ray (photographic) method was used to measure the residual stresses along with an imaging plate (IP) as a record medium for a diffraction ring.

2. Experiment

2.1 Procedure

(i) Before the main experiment, we measured the residual stresses existing in the surface of ceramics sintered under the atmospheric pressure and investigated the stresses existing in the ceramic surfaces.

Then we carried out the main experiment through the following procedures.

(ii) ① Pre-existing stress in ceramics before brazing was measured. Next, a residual stress which occurred after brazing was measured at the same point on the same sample again.

② We subtracted the pre-existing stress before brazing from the residual stress measured after brazing and calculated the real residual stresses due to brazing.

③ We compared the result of ② with the result of computer simulation by using the FEM and examined the comparison result.

(ii) We also carried out 4-point bending tests of silicon nitride samples and silicon nitride-304 stainless steel brazing samples. After that, we investigated the relations between the residual stresses and the fracture strength.

2.2 Sample

Three kinds of samples were used. They are described in Figure 1 (sample a, b and c).

2.2.1 Sample a: Many reports are available on residual stress of ceramic surface after grinding and polishing. But there is little to be known about the stresses which exist in as-sintered ceramics themselves. Therefore we measured stresses existing in the surface of ceramics sintered under the atmospheric pressure. The samples used for this purpose are called the sample a.
The following two kinds of samples were used. They were silicon nitrides manufactured by A and B companies. The ceramic sizes of A and B products were $37 \times 90 \times 4.5$ mm$^3$ and $107 \times 52 \times 6.0$ mm$^3$, respectively. We cut the ceramics to $25 \times 25 \times 4.5$ (or $6.0$) mm$^3$ with a diamond cutter and made the samples for stress measurement. The samples of A and B companies are called the samples a-I and a-II (Figure 1). No machining work was applied to the measurement surfaces of the samples a-I and a-II.

2.2.2 Sample b: Using the sample b shown in Figure 1, we measured the residual stresses which occurred by brazing. All of the silicon nitride surface before brazing was ground by using #120, #240 and #600 diamond wheels. The grinding quantity by each diamond wheel was 40 microns. Sample b was chamfered before brazing. The position of stress measurement was 0.2 mm inside the ceramic edge, on the A line (Figure 1). We knew the typical broken form in the ceramic part near the interface from the past experimental results. Therefore, we measured this part because we considered that it was closely related with a break point. As with the samples for the 4-point bending test, pieces of silicon nitride and 304 stainless steel were directly brazed with a 100 microns thick silver alloy foil (Ag-27.5Cu-2Ti) in a vacuum furnace. Figure 2 shows the temperature history for brazing. After brazing, no machining work was applied to the samples.

2.2.3 Sample c: The samples used for the 4-point bending test are called the sample c (Figure 1). Two kinds of samples were used. One is a brazed sample and the other is silicon nitride itself. The 4-point bending test was carried out according to JIS R1601. The sample edges also were chamfered.

2.3 Experimental equipment

The X-ray optical system and the X-ray conditions for stress measurement are shown in Figure 3. We used Microflex D-1 X-ray generator made by RIGAKU Corporation. The position of stress measurement on the sample was checked with a microscope. The actual method for sample setting is as follows (Figure 4). The sample must be rotated on the center of an X-ray irradiation part in order to measure stresses by the $\sin^2 \psi$ method. For that reason, the sample holder is equipped with fine adjustments for the $x$, $y$ and $z$ axes respectively on the upper and lower side of the rotatory axis. First,
a 10 microns diameter pinhole is set at a position on the sample surface. Then, with a mirror and a microscope, the pinhole position is checked for a shift to right or left when the pinhole is rotated. If the pinhole position shifts, we make the position of the rotatory center agree with the pinhole position by using the fine adjustment on the upper side of the rotatory axis. And using the fine adjustment on the lower side of the rotatory axis, the X-ray beam is led to pass through the pinhole. After completing these operations, the pinhole is removed, and with the mirror and microscope, the sample is set at the pinhole position that has just been searched. By this method the sample can be rotated on the center of the X-ray irradiation part.

2.4 X-ray beam and diffraction ring

For the application to small areas, we prepared about 90 microns diameter X-ray beam (in full width at half maximum) specifically on the sample surface and irradiated the measurement point with that X-ray beam. The measured diameter of the X-ray beam on the sample surface is shown in Figure 5. The diameter of the X-ray beam was about 90 microns in full width at half maximum in both directions, vertical and horizontal. A diffraction pattern recorded on the Imaging plate is shown in Figure 6. Because the X-ray irradiation area was small, the pattern of diffraction ring became

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(X-ray conditions for stress measurement)

<table>
<thead>
<tr>
<th>Characteristic X-rays</th>
<th>Y-Kα</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffraction plane</td>
<td>212</td>
</tr>
<tr>
<td>Diffraction angle 2θ₀ (deg)</td>
<td>170.83</td>
</tr>
<tr>
<td>Filter</td>
<td>Ti</td>
</tr>
<tr>
<td>Tube voltage (kV)</td>
<td>30</td>
</tr>
<tr>
<td>Tube current (μA)</td>
<td>300</td>
</tr>
<tr>
<td>Irradiated area (mm²)</td>
<td>6×10⁻⁶</td>
</tr>
<tr>
<td>X-ray Young’s modulus E (GPa)</td>
<td>340</td>
</tr>
<tr>
<td>X-ray Poisson’s ratio ν</td>
<td>0.27</td>
</tr>
</tbody>
</table>

Fig. 3 X-ray optical system for stress measurement.

Fig. 4 Arrangement of the equipments for X-ray stress measurement.

Fig. 5 Measured diameter of X-ray beam.

Fig. 6 Measurement of Debye ring diameter.
spotty. If such a spotty diffraction ring is measured by the counter method for X-ray stress measurement, it is practically impossible to measure the stress because the measured values largely vary. For this reason, "Measurement of the diffraction angle" described below was carried out.

2.5 Measurement of the diffraction angle

Diffraction data stored on the IP (imaging plate) was read out with a laser scanner (FUJI-BAS2000). Then the diffraction data was converted to the corresponding X-ray intensities on the computer software. The resultant data was divided into concentric circular rings of a 100 microns pitch. The computer integrated the X-ray intensity on each concentric circular ring of the 100 microns pitch (Figure 6). The computer made out a graph in which the abscissa and the ordinate respectively represent the radius and the integrated X-ray intensity. Next, we subtracted a background from the integrated data and approximated a peak part of the integrated data to a parabola by the least squares method. Then we fixed the position of the diffraction line to the center of the parabola. Using this method, we decided the position of the diffraction line.

Described below are measurement errors when this method was used (Figure 7 and 8). As the diffraction angle $2\theta$ comes closer to 180 degrees, the complementary angle $\eta(90^\circ - \theta)$ becomes smaller. Namely, the normal to the diffracting plane approaches the incident X-ray direction. Accordingly, if the diffracted X-ray positions are measured over the full circumference of the Debye ring, followed by calculations of strains at each point and those of the average strain, then we believe this average strain will come quite close to the strain in the incident X-ray direction. As shown in Figure 7(b), the incident X-rays are given from A and B directions, the full circle of the diffraction rings is measured and the average strain is calculated. We regard the values as approximate strains respectively in the A and B directions.

In the general method, $\psi$ is equal to $\psi_0(90^\circ - \theta)$ and the stress $\sigma_\phi$ in the $\phi$ direction is given as follows (Figure 7(a) and 8).

$$\sigma_\phi = E/(1+\nu) \times \varepsilon_{\phi_0}/\sin^2\psi,$$

where $E$ is the young's modulus, $\nu$ is the poisson's ratio and $\varepsilon_{\phi_0}$ is a strain in the normal direction fixed with $\phi$ and $\psi$. In our method, as $\psi_0 = \eta_0$, $\alpha$ is nearly equal to 5 degrees. The error in this assumption is 2 percent at the maximum. The simulation result is shown in Figure 8.

3. Result and Consideration

3.1 Stress measurement in the surface of as-sintered ceramics

Figure 9 shows the result of residual stress measurements on the samples a-I and a-II. Three measurement positions were taken respectively. We measured the stresses in three directions at the same point on the same sample and showed the results as the principal stresses. The values measured on the samples a-I and a-II of as-sintered silicon nitride were about -70 to +70 MPa and about -40 to +180 MPa respectively. To investigate a stress distribution near the point A where the maximum value 183 MPa was measured on the
sample a-II, we also measured stresses near the point A. The measured values near the point A are shown in Figure 9. If we take notice of the measured stresses in the X-direction and estimate the stress distribution by the full width at half maximum, the value is about 400 microns. The principal stress is not used for the estimation because the anisotropy of the principal stress is large.

In the case of measuring such local stresses, if the X-ray beam for large irradiation area is used, it is impossible to measure the stresses in this way because the stresses are averaged in the X-ray irradiation area.

The existence of over 180 MPa tensile stress was much higher than the value generally expected. For this kind of welded ceramics, the following are considered. A pre-existing residual stress will much affect the stress state after brazing, leading to a considerable influence on the brazing strength, too.

The main values of the measured stresses are given in Table 1.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Measurement method</th>
<th>Measured stresses or strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-I</td>
<td>Micro X-rays</td>
<td>Pre-existing stresses -70~+70</td>
</tr>
<tr>
<td>a-II</td>
<td>Micro X-rays</td>
<td>Residual stresses due to brazing +270(max)</td>
</tr>
<tr>
<td>b</td>
<td>Bending test</td>
<td>Brazing samples 314±27</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Silicon nitrides 649±14</td>
</tr>
</tbody>
</table>

![Figure 9](image9.png) Measured residual stress.

![Figure 10](image10.png) Measured residual stress.

![Figure 11](image11.png) Residual stress distribution near the interface between silicon nitride and 304 stainless steel.

3.2 Silicon nitride–304 stainless steel brazing sample

3.2.1 Residual stress measurement

The stresses in three directions are measured at the same point and the results are shown in terms of the principal stresses. Figure 10(a) and (b) show the stresses measured on silicon nitride before brazing and the residual stresses measured after brazing respectively. Figure 10(c) shows the stress state obtained by subtracting the initial stress before brazing from the residual stress measured after brazing. From Figure 10(b) it is very difficult to grasp the tendency of the residual stresses which occurred by brazing. But it seems that the tendency does appear in Figure 10(c). A tendency toward high tensile stress is shown in a direction nearly normal to the brazing interface. Also, near the interface, high tensile stress having a slight inclination toward the center of the joint is recognized. The tensile residual stress obtained was about 270 MPa at maximum at a point 0.2 mm away from the interface. Figure 11 shows the residual stresses measured near the brazing interface by simplification. Judging from the inclination of the measured value shown in Figure 11, a residual stress nearer than 0.2 mm from the interface seems to be a higher tensile stress. But a certain paper describes that the residual stress decreases near the joints. We have yet to confirm the phenomenon. We therefore intend to...
measure the stresses in further detail and examine the stress state. For the reason, we think we need to arrange thinner X-ray beams.

3.2.2 Analysis by using the FEM

In order to examine this experimental result, we conducted thermal stress analysis of two dimensions by using the FEM. Properly speaking, three dimensional analysis must be needed. Here, for simplicity, we just investigated the stress state so as to get the basic tendency of residual stresses due to brazing. For simplification, the analysis conditions assumed are as follows: (1) The temperature distribution is uniform. (2) The cooling speed of the temperature is very slow. (3) Young's modulus $E$ and coefficient of thermal expansion $\alpha$ in each material aren't changed by the temperature. (4) The temperature is changed from 500 K to 300 K in consideration of the yield strength of austenitic stainless steel at an elevated temperature. The thermal stress under this condition almost corresponds to the stress within the elastic limit of austenitic stainless steel. (5) The number of elements is 428.

Part of the result is shown in Figure 12. We took notice of a tensile thermal stress near the interface. When Figure 12 and 10(b) were compared, it was difficult to find a similar tendency. By comparing Figure 12 with 10(c), however, we found that the stress distribution by computer simulation agrees qualitatively with that measured by using X-rays.

3.3 4-point bending test\(^{12,13}\)

The result of the 4-point bending test is shown in Figure 13 and Table 1. The numbers of silicon nitride samples and silicon nitride-304 stainless steel brazing samples were 5 and 7, respectively. The average strength of silicon nitride was 649 MPa and the standard deviation was 14 MPa. The average strength of a silicon nitride-304 stainless steel brazing sample was 314 MPa and the standard deviation was 27 MPa. Originally, we considered that we must check whether high stresses exist in the samples for bending. But, as the strength variations of silicon nitrides themselves were small, we concluded that the existing stresses are small in the silicon nitrides. So stress measurement of each sample for bending was omitted. Silicon nitrides used for the bending test of the silicon nitrides themselves and the silicon nitride-304 stainless steel brazing samples were made from the same silicon nitride plate.

We found that the strength of the silicon nitrides decreased by 335 MPa after brazing. As the influential factors, the following are considered. (1) The pre-existing residual stress in ceramics before brazing. (2) The crack which occurred by machining work before brazing. (3) The stress which occurred by brazing. Regarding (1), we believe that the influence of the pre-existing residual stress is small because the strength variations of silicon nitrides themselves were small. Regarding (2), since the ceramics are very sensitive to cracks, if the cracks happen in the silicon nitride surface, the bending strength seems to be extremely lower and the strength variations seem to be larger. So, we think that the influence of the crack is little. Therefore, we believe that the stress which occurred by brazing influences the bending strength very strongly.

For six out of the seven samples, the break position was located in the ceramic part near the joint (Figure 14(b)) and the break position of the other sample ranged from part of the interface to the ceramic part near the joint. The interface part in the latter case was toward the side where tensile stresses do not work in the bending test. On the contrary, the interface part was near...
the edge where compressive stresses work (Figure 14 (a)). Accordingly, we thought the starting point of crack was in the ceramic and the crack advanced from the ceramic toward the interface. We could not recognize an appreciable difference in the bending strength by the difference in the break position. The break direction agreed with the normal to the direction of the maximum principal stress.

4. Conclusions

(1) Over 180 MPa of residual stress was measured in the surface layer of the as-sintered silicon nitride. The area of the stress concentration was as small as below 1.0 mm in diameter.

(2) The stress state obtained by subtracting the initial stresses before brazing from the residual stresses measured after brazing agreed with the computer simulation result qualitatively.

(3) The tensile residual stress obtained with the micro X-ray beam was about 270 MPa at maximum at a point 0.2 mm away from the interface. The result of the 4-point bending test shows that the strength of the silicon nitride decreased by 335 MPa due to brazing.

(4) The break position agreed approximately with the point where the maximum principal stress was measured. The break direction also agreed with the normal to the direction of the maximum principal stress.

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