Dissimilar Laser Brazing of Boron Nitride and Tungsten Carbide

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Abstract

Dissimilar laser brazing of h-BN and tungsten carbide has been successfully carried out using Ag-Cu-Ti braze with good bond strength. In order to investigate the characteristic of joint, observation and structural analysis of its interface by the electron probe micro-analysis (EPMA), the scanning acoustic microscopy and shear strength tests were performed. The wetting property between h-BN and Ag-Cu-Ti braze was excellent. At the interface between h-BN and Ag-Cu-Ti braze, concentration of Ti near the interface with the thickness of about 2μm was observed. It is presumed that Ti-N compound layer was formed by the reaction of Ti with h-BN. From non-destructive observation of the interface, it seemed to be no large voids or cracks in the braze area were existed. It was found that joints formed by 71.07mass%Ag-27.68mass%Cu-1.25mass%Ti could obtain shear strength of 8.7MPa and fracture was made at h-BN side of the interface of brazing joint.

Keywords: Laser brazing, Boron nitride, Tungsten carbide.

1. Introduction

The brazing technology is used in many industrial fields such as engineering structures and electronic devices. This technology has a lot of advantages as suitable for a difficult material and shape to connect in other methods and precisely suitable for mass production. And the bonding of ceramic to metals is a common requirement for their successful application. Moreover, the approach of a new combination on the joint is a common requirement for high functionality to the products in recent years. However, there are some problems such as causing the joint defect that due to thermal stress in the joint field and material deterioration by heating in these dissimilar joining.

Boron nitride has a various functional characteristics. Especially h-BN has a good thermal resistibility and solid-lubrication (Biddulph, 1983) and tungsten carbide alloy made by powder metallurgy has low thermal expansion coefficient and high rigidity good for structural material.

Laser brazing has good characteristics for dissimilar joining process because of possibility of short heating time and small heating area, and suppression of damage to the base materials. So, this study described dissimilar laser brazing of boron nitride and tungsten carbide and in order to investigate the characteristic of joint, cross-sectional observation, elemental and structural analysis of its interface and adhesion evaluation, and shearing strength measurement were performed.

2. Experimental Procedures

Tungsten carbide and h-BN were used in this work. Tungsten carbide purchased from Mitsubishi Materials Corporation (Tokyo, Japan) was equivalent material classified with ISO K10 grade. h-BN was bought from Kojundo Chemical Laboratory Co.,Ltd (Saitama, Japan), Ag-Cu-Ti alloy brazes were made at Tanaka Kikinzoku International (Tokyo, Japan) as sheet, which include Ti as major active ingredient for direct ceramic brazing. Nominal compositions and properties were summarized in Table 1 and Table 2. And the size of braze sheet was determined to 80% for the joint area of h-BN prevent to molten braze
flow out of the joint.

Before being used, h-BN blocks, brases and tungsten carbide plates were degreased by ultrasonic agitation for 10 min in acetone and dried in air. Sample configuration was top hat shape. Braze sandwiched with h-BN from top side and tungsten carbide from bottom side in a vacuum chamber. Small pressure of 1.2 MPa was used to prevent the workpiece from moving when braze was melted. And no gap adjustments for the joint were done. A vacuum chamber with a 100 mm diameter was used which was evacuated to less than 10⁻¹ Pa after samples had been loaded. And substitution to the atmospheric pressure with 99.999% purity Ar gas was done after the evacuation. This evacuation and substitution cycles were done at least three times before brazing. During brazing, Ar gas continued to flow, which flow rate was about 5 L/min. Laser brazing condition was summarized in Table 3. Scanning of laser was done on the tungsten carbide substrate around the h-BN.

Some of the samples were subsequently cross-sectioned by the low-speed diamond saw cooling with water, and mounted by epoxy resin, curing at room temperature about 8 – 10 hours, and grinded by SiC paper #120 – #1200, polished by 3μm – 1μm polycrystalline diamond to provide microstructural information.

Cross sectional observation and elemental analysis of the interface were performed using electron probe micro-analyzer (JEOL Co. Ltd. JXA-8621MX) and interfacial observation and estimation of interface area were performed using scanning acoustic microscope (Hitachi Kenki FineTech Co., Ltd. HSAM220). Some of the samples were placed in a shearing jig as shown in Fig. 1 and stressed to destruction in precision universal tester (Shimadzu

### Table 1 Materials used in this work.

<table>
<thead>
<tr>
<th>Material</th>
<th>Grade</th>
<th>Nominal composition (mass %)</th>
<th>Bend strength at room temperature (MPa)</th>
<th>Density ($\times 10^3$ kg/m³)</th>
<th>Relative Density (%)</th>
<th>Size (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tungsten</td>
<td>HTI</td>
<td>WC: 94, Co: 6</td>
<td>32000</td>
<td>14.9</td>
<td>-</td>
<td>20<em>20</em>2</td>
</tr>
<tr>
<td>Carbide</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>h-BN</td>
<td>99%</td>
<td>h-BN &gt; 99.993</td>
<td>32.5</td>
<td>1.93</td>
<td>82.5</td>
<td>5.5<em>5</em>3.5</td>
</tr>
</tbody>
</table>

### Table 2 Braze used in this work.

<table>
<thead>
<tr>
<th>Grade</th>
<th>Nominal composition (mass %)</th>
<th>Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TKC-711</td>
<td>Ag 71.07, Cu 27.68, Ti 1.25</td>
<td>0.1</td>
</tr>
</tbody>
</table>

### Table 3 Laser brazing condition.

<table>
<thead>
<tr>
<th></th>
<th>Pulsed YAG Output (kW)</th>
<th>CW LD Output (kW)</th>
<th>Pulse frequency (Hz)</th>
<th>Scanning speed (mm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1.7</td>
<td>0.02</td>
<td>100</td>
<td>0.1 ~ 1.6</td>
</tr>
</tbody>
</table>

![Fig. 1 Schematic diagram of shear strength test.](image_url)

Cooperation Autograph AGS-5kNB) operating at a crosshead speed of 0.5 mm/min. And shear strength was calculated from maximum load divided by interface area estimated from scanning acoustic microscopy. And its average shear strength was calculated using the Weibull distribution function:

$$\ln \ln (1-F)^{-1} = m \ln \sigma - m \ln \sigma_0$$  \ (1)

where: $F$ is the cumulative failure probability, $m$ is the Weibull modulus, $\sigma_0$ is the characteristic strength. In this paper, the median rank method was used for computing cumulative failure probability $F$ in Eq. (1) because of its good reliability despite the small number of samples:

$$F = (i - 0.3) / (n + 0.4)$$ \ (2)

where: $i$ is the rank of the observation and $n$ is the number of samples. Average shear strength $\mu$ is as follows:

$$\mu = \sigma_0 \Gamma(1+1/m)$$ \ (3)

where: the $\Gamma$ function is expressed as follows.

$$\Gamma(x) = \int_0^\infty e^{-t} t^{x-1} dt \ (x>0)$$ \ (4)
3. Results

Fig. 2 shows cross sectional SEM observation of a h-BN / Ag - Cu - Ti braze / WC - Co interface with low magnification. The top side of the picture is h-BN, the bottom side of the picture is WC-Co plate. The center layer is Ag-Cu-Ti braze. Though it is not a measurement according to sessile drop method, the contact angle between WC-Co and braze is at an acute angle from Fig. 2. The contact angle between h-BN and Ag - Cu - Ti braze is also at an acute angle although h-BN surface is not flat. And each interfaces spread to the left side.

Fig. 3 shows map analysis of a h-BN / Ag - Cu - Ti braze interface. Fig. 3 a) shows enlargement of h-BN / Ag - Cu - Ti braze interface, which was complicated. Fig. 3 b) - d) shows distribution of Ag, N and Ti near the interface in the same area of Fig. 3 a), respectively. From these distributions of elements, concentration of Ti near the interface was observed, which thickness was about 2μm.

Fig. 4 shows the interface observation of h-BN / Ag - Cu - Ti braze / WC - Co interface. Fig. 4 a) shows external appearance of the specimen. Fig. 4 b) shows the image of scanning acoustic microscopy. In Fig. 4 a), white part of square in the center is h-BN, a peripheral metallic luster part is WC-Co plate. The black area in the center of Fig. 4 b) is Ag - Cu - Ti braze which was melt in the joint. In the white part around the black area, no brazes exist. There was no big void in the black area where braze was melt in the joint.

After these examinations and estimations, shear strength test of the dissimilar joint was done. In each specimen, fracture occurred at h-BN side of the specimen near the interface.

Fig. 5 shows the Weibull probability plot of shear strength test. All points were along the approximation line plotted as Eq (1). And the distribution of shear stress was narrow around 10 MPa. From Eq. (1), the estimated Weibull modulus m was computed to 4.1 and estimated characteristic
strength $\sigma_0$ was also calculated to 9.6MPa, respectively. Its average shear strength $\mu$ was 8.7MPa calculated from Eq. (3).

4. Discussions

From Fig. 2, the contact angle between WC-Co and braze and between h-BN and braze were at an acute angle. Judging from each interface, wettability of Ag-Cu-Ti braze on each interface seemed to be good.

From Fig. 3 a), the interface between h-BN / Ag-Cu-Ti was complicated in detail. It is presumed that molten Ag-Cu-Ti braze infiltrated into open pore part of h-BN surface because the wettability between braze and h-BN was excellent. The addition of Ti as an active element to braze seemed to be sufficient to induce wetting of h-BN (Nicholas, et al, 1990; Nicholas, et al, 1992; Benko, 1995 and Benko, et al, 1997) even with small Ti content of 1.25%, though optimum Ti content was not discussed in this paper.

Ti and N near the interface concentrated as shown in Fig. 3 b) ~ d). Standard free energies of formations of nitrides describes that standard free energies of formations of TiN is about 140 [kJ / mol N$_2$], which is lower than that of BN at 1173K (Elliott and Gleiser, 1960). This means that TiN is more stable then BN in all temperature ranges. From these results, the formation of reacted layer was presumed, which existed near interface of h-BN / braze and its thickness was about 2µm and main component was TiN (Nicholas, et al 1990 and Peteves, 1996).

As shown in Fig. 4 b), it seemed to be no large void or crack in the brazed area due to good wettability of braze with h-BN and WC-Co. In addition, laser beam heating at the selected area with low heat input enabled the crack free joint between h-BN and WC-Co.

In all shear strength tests of the joints, every fracture was made at h-BN side of the interface of brazing joint, and this suggests that the interface of brazing joint has the strength enough to connect h-BN and WC-Co.

From Fig. 5, all of the point was plotted as linear along the approximation line. It suggested that this fracture of dissimilar joint followed a Weibull distribution. So the assumption of its average shear strength $\mu$ was reasonable. From the narrow range of its shear strength distribution, it seemed that each dissimilar joining was done in stable.

5. Concluding Remarks

Dissimilar laser brazing of Boron Nitride and Tungsten Carbide was done. Observation and structural analysis of its interface and shear strength test revealed as follows.

(1) Wettability of both Ag-Cu-Ti braze to both h-BN and WC-Co was considered to be good.
(2) At the interface of h-BN / Ag – Cu – Ti, braze was observed to infiltrate into micro open pores of h-BN.
(3) At the interface of h-BN / Ag – Cu – Ti braze, concentration of Ti near the interface which thickness was about 2µm was observed. It was presumed that the formation of reacted phase existed at the interface.
(4) No large voids or cracks in the braze area were existed from non-destructive observation of the interface using Scanning Acoustic Microscopy.
(5) On shear strength test of the dissimilar joint, fracture occurred at h-BN side near the interface in each specimen. And this suggests that the interface of brazing joint has the strength enough to connect h-BN and WC-Co.
(6) The distribution of the shear strength followed a Weibull distribution. And its average shear strength $\mu$ was 8.7MPa.

Acknowledgment

This work was supported by Joint Research system at the Joining and Welding Research Institute, Osaka University, and Grant-in-Aid for Cooperative Research Project of Nationwide Joint-Use Research Institutes on Development Base of Joining Technology for New Metallic Glasses and Inorganic Materials from The Ministry of Education, Culture, Sports, Science and Technology, Japan.

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