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Short communication

Characteristics of dissimilar laser-brazed joints of isotropic graphite to WC–Co alloy

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ABSTRACT

The effect of Ti serving as an activator in a eutectic Ag–Cu alloy filler metal in dissimilar laser-brazed joints of isotropic graphite and a WC–Co alloy on the joint strength and the interface structure of the joint is investigated in this study. To evaluate the joint characteristics, the Ti content in the filler metal was increased from 0 to 2.8 mass%. The laser brazing was carried out by irradiating a laser beam selectively on the WC–Co alloy plate in Ar atmosphere. The threshold content of Ti required to join isotropic graphite to WC–Co alloy was 0.4 mass%. The shear strength at the brazed joint increased rapidly with increasing Ti content up to 1.7 mass%, and a higher Ti content was found to be likely to saturate the shear strength to a constant value of about 14 MPa. The isotropic graphite blocks also fractured at this content. The concentration of Ti observed at the interface between isotropic graphite and the filler metal indicates the formation of an intermetallic layer of TiC.

1. Introduction

Isotropic graphite has isotropic characteristics of the mechanical properties in addition to the fundamental functional characteristics of graphite, such as a small weight, low thermal expansion coefficient, superior thermal and electrical conductivities, thermal shock resistance and solid lubricity [1–5]. The process of brazing is suitable for joining dissimilar materials, particularly those that cannot be easily joined by a fusion welding process, such as ceramics and metals [6–19]. Similar to ceramics, it is also difficult to braze graphite to other materials without the use of an activated filler metal or some kind of surface treatment, because wettability of graphite and the molten filler metal plays a crucial role in the brazing of these materials [6–16]. Dissimilar brazing of a carbon material to metal using an activated filler metal for application in the energy and aerospace industries [1,5,8–10,20] was investigated in recent studies: for example, brazing of graphite to W–Re alloys [1], Mo [5], Cu [5] and SiC [8]; brazing of carbon–carbon composites to Nimonic alloys [9] and Cu-clad Mo [20]; and brazing of carbon fiver reinforced SiC composite to Ti alloys [10]. The behavior of the activator and molten filler metal in a conventional furnace heating process is well known [1,8–11,21–24].

An activator, e.g., Ti, Zr, or Cr, in a filler metal improves the wettability of graphite and the molten filler metal and reacts with the carbon of graphite to form a stable carbide. This results in the joining of the graphite to the metal [1,9–11,23,24]. Wettability improves with increasing activator content in the filler metal, and increased wettability in turn increases the joint strength [6,16–18]. However, certain problems remain in conventional brazing, such as material deterioration, thermal stress and strain in the joint [8–19,22,25]. Especially, depending on the brazing temperature and time, an increase in the activator content causes a decrease in joint strength because of the high thermal expansion coefficient mismatch between the graphite and the thick reaction layer [12,13,17–19,23,25]. These problems arise because conventional furnace brazing requires a long treatment time in order to ensure heating and cooling of the whole component.

Such problems can potentially be avoided in the laser brazing which is a novel brazing technique, as the heating and cooling times in this technique are short and only the selected part of the component needs to be heated [14–16]. Therefore, this brazing technique has recently been successfully applied to join h-BN to WC–Co alloy [15,16]. The behaviors of the activator and molten filler metal during laser brazing differ from those during conventional brazing because of the short processing time and smaller selected heating area [15,16]. In particular, the short processing

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time affects the diffusion and interfacial reaction processes at the interface of graphite and filler metal [12,13]. With this background, the objective of this study was to determine the effect of Ti serving as an activator in a filler metal of a dissimilar laser brazed joint of isotropic graphite to WC–Co alloy on the joint's shear strength and the interface structure of the joint; WC–Co alloy was selected as a counter material owing to its low thermal coefficient and relatively high heat resistance.

2. Materials and methods

Experiments were carried out using isotropic graphite blocks, WC–Co alloy plates and nine types of tentatively made Ag–Cu–Ti filler metal sheets with different Ti contents. Table 1 lists the chemical compositions of the filler metals. The isotropic graphite blocks, made by a cold isostatic press, were classified as grade IG-11 (Toyo Tanso Co., Ltd.) and measured 5 mm × 5 mm × 3.5 mm. WC–Co alloy plates (ISO K10 grade, Mitsubishi Materials Corporation) measuring 10 mm × 10 mm × 2 mm were used as the substrate plate. The filler metals were eutectic-type Ag–Cu alloy containing additional Ti as an activator, with the Ti content ranging from 0 to 2.8 mass%. The thickness of filler metal was 0.1 mm and its dimensions were fixed to 3 mm × 3 mm to ensure that it does not flow out of the joint interface. Before brazing, all materials were degreased by ultrasonic agitation in acetone for 10 min and dried in air.

Fig. 1 shows a schematic illustration of the laser brazing apparatus. The specimen was shaped liked a "top hat". A filler metal sheet was sandwiched between the isotropic graphite block (above it) and the WC–Co alloy plate (below it) and placed in a vacuum chamber. The top of the specimen was covered with a transparent quartz glass plate, which not only acted as a window of the vacuum chamber for laser beam irradiation, but also fixed the specimen in place. The vacuum chamber was evacuated to less than 10^{-1} Pa, and then, Ar gas of purity 99.999% was pumped into atmospheric pressure.

Table 1
Chemical composition of the filler metal (mass%).

No.	Ag	Cu	Ti
1	72.0	28.0	0
2	71.5	28.2	0.3
3	71.5	28.1	0.4
4	71.3	28.1	0.6
5	71.2	27.9	0.9
6	70.9	27.8	1.3
7	70.2	28.1	1.7
8	70.2	27.5	2.3
9	69.6	27.6	2.8

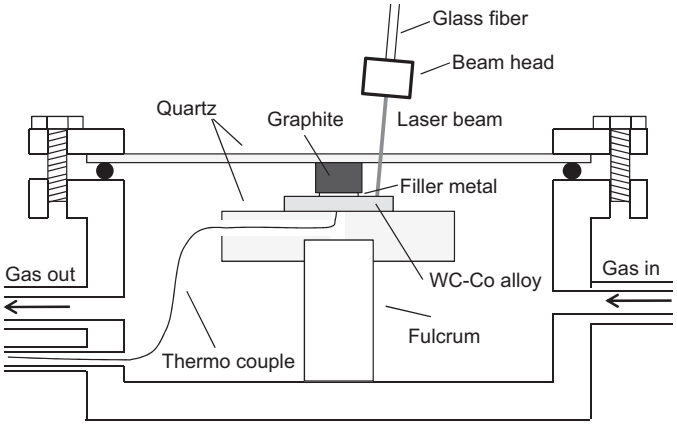

Fig. 1. Schematic illustration of the laser brazing apparatus and specimen arrangement.

Table 2
Laser brazing conditions.

Pulsed YAG average output	0.134 kW
Pulsed YAG wave length	1064 nm
CW LD output	0.02 kW
CW LD wave length	808 nm
Pulse frequency	100 Hz
Scanning time	36 s
Atmosphere	Ar flow (5 L/min)

This evacuation and substitution cycle was performed five times prior to brazing. During brazing, Ar gas continued to flow at a rate of 5 L/min. The YAG and LD lasers were coaxially transferred via optical fiber to the laser head unit and irradiated through the transparent quartz glass plate to the top side of the WC–Co alloy plate at an irradiation angle of 85°. Table 2 lists the laser brazing conditions, which were set according to JIS Z3261 BAg-8 and previous studies [15,16]. The WC–Co alloy plate around the isotropic graphite block was subjected to laser irradiation for 36 s. During laser heating, the temperature of the WC–Co alloy plate was monitored by an R-type thermocouple inserted into a hole beneath the joint on the lower surface of the plate.

After brazing, a shearing test was carried out using a precision universal tester operated at a cross-head speed of 0.5 mm/min. Five joints for each filler metal composition were tested using nine types of filler metals. The shear strength was calculated as the maximum load divided by the joining area, which was estimated from the fractured surface. The brazed joint specimens were cross-sectioned and mounted in epoxy resin, before being ground with SiC paper #220 and polished with diamond paste. Observations of the microstructure of the brazed interface and its elemental analysis were then performed using a scanning electron microscope, a transmission electron microscope and an energy dispersive X-ray analyzer.

3. Results and discussion

3.1. Strength of the brazed joint

Fig. 2 shows a typical temperature profile of the WC–Co plate during laser brazing. The highest brazing temperature was 1102 K, which is above the melting temperature of the filler metal (approximately 1027 K).

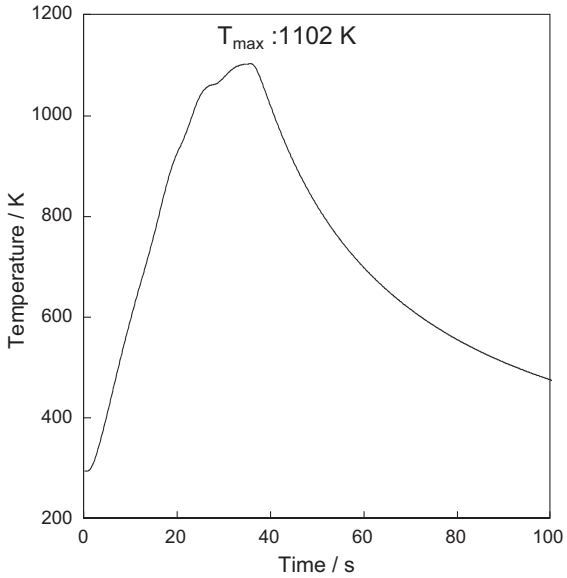

Fig. 2. Temperature profile of the WC–Co plate during laser brazing.

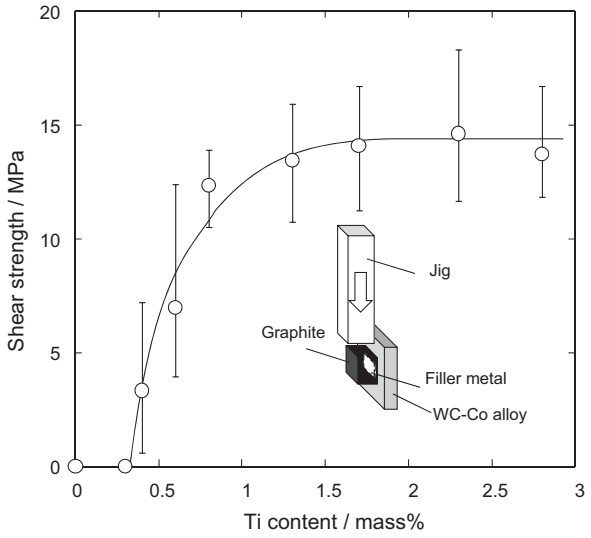

Fig. 3. Effect of Ti content on the shear strength of the joint formed by brazing using Ag–Cu–Ti filler metal with different Ti contents.

Fig. 3 shows the effect of the Ti content on the shear strength of the brazed joint. The threshold Ti content required to join the graphite block to the WC–Co alloy was 0.4 mass%. The shear strength rapidly increased as the Ti content increased up to 1.7 mass%. Higher Ti content appeared to saturate the shear strength to a constant value of about 14 MPa. This corresponded to the occurrence of fracture in the isotropic graphite block near the interface. In contrast, when the Ti content ranged from 0.4 to 1.3 mass%, fracture occurred both at the interface between the graphite block and the filler metal and in the graphite block. The shear strength increased with increasing area fraction of the fractured surface of the isotropic graphite block. This means that the Ti activator increased the joint strength of the interface between the graphite block and the filler metal. A good joint was also formed between the WC–Co alloy and Ag–Cu alloy filler metal without using Ti as an activator. The increase in shear strength with increasing Ti content in the filler metal is possibly a result of the improvement in the wettability of graphite and the molten filler metal and the formation of stable carbides when Ti reacts with the carbon in the graphite [1,9–11,23,24].

The joint strength between ceramics and metal has been reported to decrease with increasing content of Ti as an activator in the filler metal depending on the brazing temperature and time because of the high thermal expansion coefficient mismatch between the ceramics and the thick reaction layer [12,13,17–19,22,25]. However, in this study, because the heating and cooling times were rather short, the thickness of the reaction layer would be below the critical value required for decreasing the joint strength.

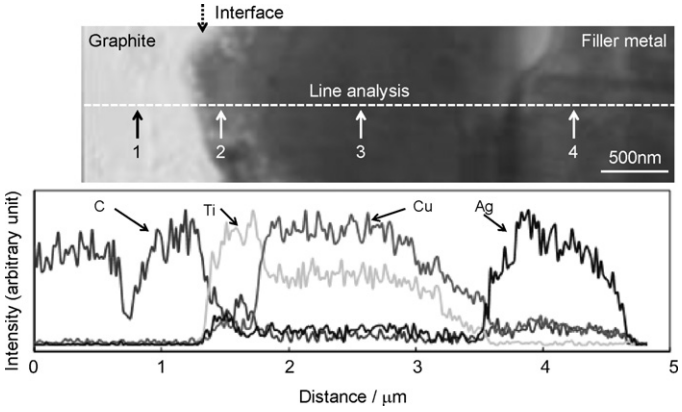

Fig. 5. Bright field image and element distributions at the interface between isotropic graphite and 1.7 mass% Ti filler metal.

The wide distribution of the shear strength of joints can be explained by the wide distribution of the graphite strength itself. Defects such as pores and cracks in the graphite matrix affect the fracture strength, as the graphite is a brittle material. This causes the shear strength of the joint to vary widely.

3.2. Microstructure of the brazed joint

Fig. 4 shows the cross-sectional microstructure and the distributions of elements C, Ag, Cu and Ti of brazed joints of isotropic graphite and the filler metal containing 0.6 and 1.7 mass% Ti (upper and lower panels, respectively). An obvious concentration of Ti was observed at the interface between isotropic graphite and each filler metal. This suggests that the Ti activator diffused from the filler metal to the interface and reacted with the isotropic graphite during laser heating. The formation of Ti-rich layers became more obvious as the Ti content in the filler metal increased, which is reflected in the increase in the thickness and the area of the Ti-rich layer.

Fig. 5 shows the bright field image and element distribution at the interface between the isotropic graphite block and the filler metal containing 1.7 mass% Ti. Positions 1–4 were marked in the bright field image to divide it into areas for determining the primary constitutional elements at each position; these were as follows: C at position 1, Ti and C at position 2, Ti and Cu at position 3, and Ag at position 4. Fig. 6(a)–(c) shows the selected area diffraction patterns of areas corresponding to positions 2, 3 and 4, respectively. These images reveal that the precipitated intermetallic compounds at the interface between the isotropic graphite and the filler metal were identified as TiC at position 2 and Cu_3Ti at position 3. Ag was

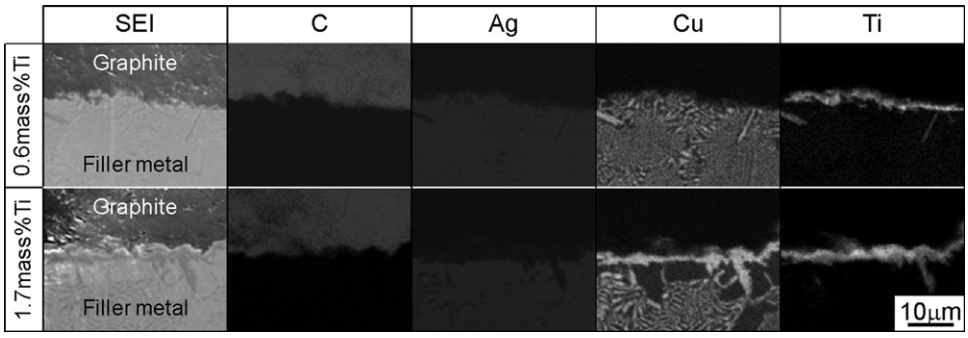

Fig. 4. Cross-sectional microstructure and distributions of elements C, Ag, Cu, and Ti of the joint of the graphite/filler metal having 0.6 and 1.7 mass% Ti.

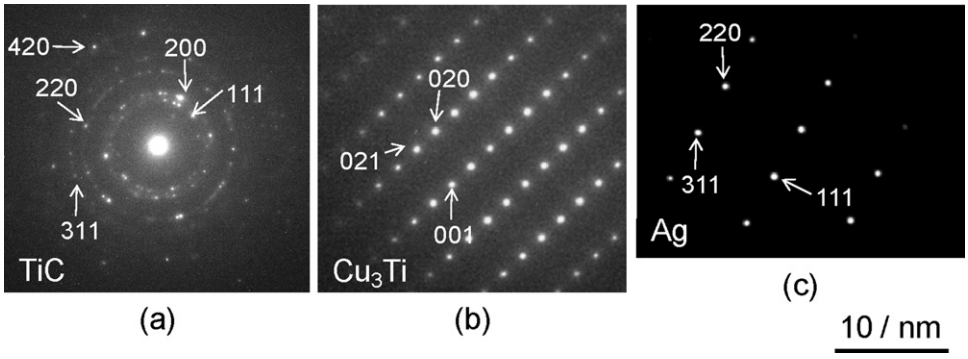

Fig. 6. Selected area diffraction patterns at (a) position 2, (b) position 3, and (c) position 4 in the bright field image in Fig. 5.

found to be the filler metal at position 4. These results indicated that the Ti-rich layer formed at the interface consisted of dual thin layers of TiC and Cu_3Ti. Ti in the filler metal has high affinity for the carbon of the isotropic graphite, and it would thus diffuse to the interface and form a thin (~300 nm) TiC layer. The formation of TiC is thermodynamically favorable, as the standard free energy for the reaction Ti + C → TiC is approximately −170 kJ/mol at 1102 K [26]. In addition, the main alloying element of the filler metal, Cu, also has high affinity for Ti, and thus, a Cu_3Ti compound was formed following the formation of TiC.

4. Conclusions

The effects of Ti as an activator in the Ag–Cu–Ti filler metal of a dissimilar laser-brazed joint of isotropic graphite to WC–Co alloy on the joint strength and the interface structure of the joint were investigated. The following conclusions were drawn about the shear strength and the interface structure of the joint:

(1) Ti serving as an activator in the Ag–Cu braze filler metal enabled the joining of isotropic graphite to WC–Co alloy via heating for a short time using laser brazing. The threshold Ti content required to join isotropic graphite to a WC–Co alloy was 0.4 mass%.
(2) The shear strength of the laser brazed joint increased with increasing Ti content in the range of 0.4–1.7 mass%, in proportion to the area fraction of the fractured surface in the isotropic graphite. A higher Ti content is likely to saturate the shear strength to a constant value of about 14 MPa.
(3) A Ti-rich layer was formed at the interface between isotropic graphite and the filler metal using laser brazing, and this consisted of TiC and Cu_3Ti compound layers. The thickness of the TiC layer using the 1.7 mass% Ti content filler metal was approximately 300 nm.

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