Effect of Argon Ion Bombardment on Diffusion Bonding of SUS304L Stainless Steel and Pure Iron

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1. Introduction

Diffusion bonding is one of important solid state joining processes in which permit the production of high quality joints with little or no need for post-bonded machining. Joints produced by diffusion bonding can meet the requirements for most critical structures in terms of strength, toughness, resistance to heat and corrosion. It has been widely applied to bond metals, electronic materials and dissimilar materials.

In the diffusion bonding process, the deformation and grain growth of the bonded joints are main problem because the bonding process is carried out at a high temperature. It has been desired that the bonding temperature is lowered during the diffusion bonding process. Ion bombardment as a surface treatment of the bonding surface is considered to be an effective method. It has been reported that it is possible to make micro-bonding of similar and dissimilar materials, such as Si/Si, Al/Al, and Al/Si3N4 at a room temperature after the argon ion bombardment treatment. However, it is not clear about the effect of ion bombardment treatment for the diffusion bonding of various metals yet.

In the present study, to investigate the effect of argon ion bombardment for bonding surface, SUS304L stainless steel and pure iron specimens were treated with argon ion bombardment and then diffusion bonding was carried out. The effect of argon ion bombardment treatment on the properties of diffusion bonding joint was investigated by the tensile tests of joints and microstructure analyses of the fractured surfaces. The results showed that the argon ion bombardment treatment before bonding was effective to clean the bonding surface and reduce the inclusions at the bonded interface, so that the tensile strength of the bonded joints was improved and bonding temperature was lowered. The joint properties of pure iron were more excellent than those of SUS304L stainless steel. The amount of the inclusions at the joints with argon ion bombardment treatment depended on affinity of oxygen to metal.

KEY WORDS: argon ion bombardment; diffusion bonding; SUS304L stainless steel; pure iron; microstructure; inclusion; oxide film.

2. Experimental Methods

The cylindrical specimens of SUS304L stainless steel and pure iron with 12 mm in diameter and 30 mm in length were used in this study. The chemical composition of the materials is presented in Table 1. Faying surface to be bonded was prepared by a lapping method. The surface profiles were obtained using a surface roughness tester (Taylor Hobson From Talysurf Series 2). Figure 1 shows the typical surface-asperities of the specimen by lapping method. The roughness (Ry) is 100 nm for both of SUS304L stainless steel and pure iron specimens at 1 mm in measuring length. Both bonding surfaces have large waviness.

The surfaces of SUS304L stainless steel and pure iron specimens prepared by a lapping method were treated with argon ion bombardment, and then diffusion bonding was carried out. The effect of argon ion bombardment treatment on the properties of diffusion bonding joint was investigated by the tensile tests of joints and microstructure analyses of the fractured surfaces. The results showed that the argon ion bombardment treatment before bonding was effective to clean the bonding surface and reduce the inclusions at the bonded interface, so that the tensile strength of the bonded joints was improved and bonding temperature was lowered. The joint properties of pure iron were more excellent than those of SUS304L stainless steel. The amount of the inclusions at the joints with argon ion bombardment treatment depended on affinity of oxygen to metal.

KEY WORDS: argon ion bombardment; diffusion bonding; SUS304L stainless steel; pure iron; microstructure; inclusion; oxide film.

Table 1. Chemical composition of SUS304 stainless steel and pure iron specimens.

| SUS304L stainless steel (mass%) | Fe  | Cr  | Ni  | S   | P   | Mn  | Si  | C   |
|-------------------------------|-----|-----|-----|-----|-----|-----|-----|-----|
| Bals                          | 18.41 | 10.30 | 0.002 | 0.029 | 1.76 | 0.51 | 0.015 |
| Pure Iron (ppm)               |     |     |     |     |     |     |     |     |
| H                             | 2   | 22  | 2   | 1   | 2   | 1   | 1   | 6   |
| Cu                            | 6   | 6   | 6   | 6   | 6   | 6   | 6   | 6   |

Fig. 1. A typical surface-asperities of the specimen by lapping method.
Figure 2 shows a schematic illustration of the argon ion bombardment treatment and the diffusion bonding apparatus. The specimens were ultrasonically rinsed in acetone. Then, the two specimens were held in the bonding chamber, subsequently bonding chamber was evacuated for 12 h using a molecular turbo pump. The pressure was about 0.5–1.2×10⁻¹⁰ Pa. The surfaces of the specimens were bombarded by argon ion beam with 2.1 kV accelerating voltage and 20 mA ion current. The incident angle of argon ion beam was 10°. The time of argon ion bombardment treatment was 20 min. Then specimens were brought into contact together within two minutes after ion bombardment.

Bonding pressures applied to the specimens were 2.0 kN for SUS304L stainless steel and 600 N for pure iron, respectively. The specimens were heated using a high frequency induction heating method. Holding time at bonding temperature was 20 min.

The mechanical properties of the bonded joints were evaluated by the tensile tests at a room temperature using AG-250KNG autograph tensile tester. The tensile velocity of 1 mm/min was used. The microstructure observations of the fractured surfaces and the analyses of the inclusions were carried out using JSM-6400 scanning electron microscope (SEM) equipped with JED-2110 energy dispersive X-ray spectroscopy microanalyzer (EDS).

3. Results and Discussion

The effect of ion bombardment was investigated by evaluating tensile properties of joints for SUS304L stainless steel. Figure 3 shows tensile strength of the SUS304L stainless steel joints for two different surfaces with and without argon ion bombardment. The tensile strength of joints increased with an increasing in bonding temperature for both surface treatments. The specimens using argon ion bombardment treatment could be bonded at bonding temperature of 600°C, and tensile strength came up to 680 MPa at bonding temperature of 900°C, which was equal to that of parent metal SUS304L stainless steel. However, the joints could not be formed until 700°C for specimens without bombardment. Figure 4 shows relationship between bonding temperature and reduction in area of joints made with two surface treatments. The reduction in area of joints with argon ion bombardment treatment was larger than without bombardment in total bonding temperature range. The reduction in area was about 58% for the joints by argon ion bombardment treatment and was about 28% for the joints without bombardment treatment at bonding temperature of 1 000°C. These results indicated that argon ion bombardment improved tensile strength and reduction in area of joints, and lowered bonding temperature.

In order to investigate the improvement by argon ion bombardment, observations at fractured surface were carried out after the tensile testing of joints. Figure 5 shows a fractured surface of joints at bonding temperature of 800°C for SUS304L stainless steel specimen. White area shows the fractured zone at real bonded area. It could be seen that the bonded area was only in the center region. From the photograph of fractured surface, the real contact area was measured. Tensile strength per real bonded area was calculated using the measured real bonded area. Figure 6 shows the relationship between tensile strength at real bonded area and bonding temperature for the joints of SUS304L stainless steel. The tensile strength at real bonded area increased with an increasing in bonding temperature. The tensile strength of joints with argon ion bombardment treatment was larger than without bombardment treatment in the experimental temperature range. Based on these experimental
results, it was clear that tensile strength at real bonded area was improved with argon ion bombardment treatment.

In order to understand the cause of the difference of tensile strength of joints for both surface treatments, microstructure of the fractured surfaces of the bonded joints was observed by SEM. Figure 7 shows the micrographs of the fractured surfaces of the SUS304L stainless steel joints bonded at bonding temperature of 1000°C. Figure 7(a) gives the result of the joint bonded with argon ion bombardment treatment. It could be observed that the fractured surface was ductile and there were very small amount of the inclusions at the fractured surface. Figure 7(b) shows the joint bonded without bombardment treatment. Many small dimples at the fractured surface and the numbers of the inclusions in the dimples (as shown by arrows in Fig. 7) were seen. The EDS analyses indicated that the inclusions consisted of the elements of oxygen (O), chromium (Cr), manganese (Mn), and so on. It was reported that the inclusions was formed from oxide film at bonding surface.15)

In order to investigate the effect of argon ion bombardment treatment on the amount of the inclusions at the fractured interface, inclusions ratio, which is defined as the fraction of the area occupied by inclusions to the total measured regions, was obtained using SEM micrographs (more than 8 different regions) taken from one fractured joint. Figure 8 shows the relationship between inclusions ratio at the fractured surface and bonding temperature for SUS304L stainless steel joints. The amount of inclusions at fractured surface decreased with argon ion bombardment treatment, and it also decreased with an increasing in bonding temperature. As the amount of inclusions depended on the thickness of surface oxide film, and it could be suggested that the thickness of oxide film on specimen bombarded by argon ion was very thin. These results indicated that ion bombardment treatment was effective to clean the bonding surface and reduced the inclusions. Cleaning of the bonding surface was useful to improve the bonding strength and lowered the bonding temperature.

Next pure iron specimens with and without argon ion bombardment treatment were bonded, and the tensile tests and the fractured surface microstructure observations of joints were carried out. Figure 9 shows the results of the
tensile strength of pure iron specimens with and without argon ion bombardment treatment. The results showed that the tensile strength was improved and bonding temperature was lowered by argon ion bombardment treatment in the same manner of SUS304L stainless steel joints. Furthermore, SEM observations of fractured surface showed that amount of inclusions at fractured surface of pure iron joints were less than those of SUS304L stainless steel.

In the present study, the time of argon ion bombardment treatment was 20 min. Based on the measurement results from mass change before and after argon ion bombardment treatment, the surface layer with 12 nm was removed for SUS304L stainless steel specimens. Then under this ion bombardment condition, the oxide layer on bonding surface was completely removed, and the clean surface was formed. In this study, the time span after argon ion bombardment treatment to diffusion bonding was about 2 min. The pressure in the bonding chamber was about $1 \times 10^{-4}$ Pa. The possibility of specimen surface re-contamination by residual gas in bonding chamber after argon ion bombardment treatment existed. Moreover, the affinity of oxygen to metal increases in Fe, Cr, Mn in turn. EDS analyses showed that there were Cr, Mn elements in the fractured surface in the SUS304L stainless steel joints. As SUS304L stainless steel had high affinity of oxygen, the bonding specimens surfaces were easily contaminated again after argon ion sputter-cleaning, and the inclusions could be formed at the bonded interface. It is known that the inclusions are formed owing to spheroidizing of oxide film at the bonded interface. The amount of inclusions decreases with a decreasing in the thickness of oxide film. On the other hand, the affinity of oxygen to Fe was lower than those of Cr and Mn. The surface re-contamination of pure iron specimens was more difficult than that of SUS304L stainless steel specimens. Therefore, the amount of inclusions in the SUS304L stainless steel joints was more than that in pure iron joints.

Then, it was tried to compare bonding properties of SUS304L stainless steel and pure iron specimens. Figure 10 shows the relationship between joint efficiency (ratio of tensile strength at real bonded area to tensile strength of parent materials) and equivalent temperature (ratio of bonding temperature to melting point of parent material) of SUS304L stainless steel and pure iron specimens.

One cause of the difference in bonding properties between the metals was considered to depend on amount of inclusions and bonded area. The amount of inclusions in the pure iron joints was less than that in SUS304L stainless steel joints, and the diffusion bonding properties of pure iron specimens was more excellent than those of SUS304L stainless steel specimens. Moreover, the effect of bonded area should also be considered. The bonded area was depended on plastic deformation, creep deformation, interface diffusion and volume diffusion. The effect of bonded area is discussed in the next paper.

4. Conclusions

SUS304L stainless steel and pure iron specimens were treated by argon ion bombardment. Effect of ion bombardment treatment on tensile strength and microstructure of diffusion bonding joints was investigated. The results obtained are summarized as follows.

(1) Argon ion bombardment treatment is effective to clean the bonding surface, reduce the inclusions, and results in improving the bonding strength of joints and lowering of bonding temperature.

(2) Diffusion bonding properties of pure iron specimens are more excellent than those of SUS304L stainless steel.

(3) The amount of the inclusions at the joints by argon ion bombardment treatment depends on affinity of oxygen to metal.

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![Fig. 9. Relationship between tensile strength of pure iron joints by argon ion bombardment and un-bombardment treatment and bonding temperature.](image)

![Fig. 10. Relationship between joint efficiency (tensile strength at bonded area to tensile strength of parent material) and equivalent temperature (bonding temperature to melting point of parent material) of SUS304L stainless steel and pure iron specimens.](image)
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