Effect of Argon Ion Bombardment on Strength of Diffusion Bonded Joints of Cu and Ti[†]

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This study has been carried out to determine the changes in the surface of a metal which has been exposed to ion bombardment using different accelerating voltages.

The work shows that joint strength depends on the accelerating voltage used during ion bombardment. When ion bombardment is carried out at accelerating voltages of up to 2 kV, there is an increase in joint strength and this depends on the removal of contaminants at the bonding surface. Argon ion bombardment at the accelerating voltages from 2 up to 5 kV results in the formation of a hard surface layer. This layer is removed by the annealing treatment of samples after argon ion bombardment. With softening of the bonding surface, the joint strength increases. The reason for the decrease in bond strength at high accelerating voltages is due to the formation of this hard surface layer. The surface annealing treatments used after ion bombardment is effective in removing any surface damage.

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I. Introduction

In solid state bonding, the surface film has an influence on the properties of the joint. Therefore, many studies have been carried out to investigate the effects of various surface treatments during bonding and the behaviour of surface films at the bonding interface on the bond strength.

Surface treatments such as heating in a vacuum⁽¹⁾, sliding between the bonding surfaces⁽²⁾, and ion bombardment⁽³⁾⁽⁴⁾ have been previously studied and applied to the bonding process, and an improvement has been reported. Argon ion bombardment is found to be effective in removing all surface contaminants and oxides and allows the bonding of some metals at a room temperature⁽³⁾⁽⁴⁾. However, it is not clear what changes occur at the surface.

This investigation was carried out to determine the surface changes of a metal when exposed to ion bombardment at various accelerating voltages. To observe the surface changes, the composition, surface morphology, hardness and fracture surface were investigated by means of Auger spectroscopy, surface profile measurements, and hardness and scanning electron microscopy techniques.

II. Experimental Procedure

Oxygen-free high conductivity copper and commercial pure titanium were used and bonded in the form of rods

with a diameter of 12 mm.

These materials were heat treated in a vacuum of 10^{-4} Pa for 5 d at 400°C for copper and 700°C for titanium respectively, in order to remove any residual hydrogen gas.

Figure 1 shows the dimensions of specimens for joining. A flat specimen was brought into contact with a spherical specimen. As a fracture area of the joint is round, it is easy to measure the bonded area.

The surface preparation involved polishing with a 2 μ m diamond suspension and degreasing using acetone in an ultrasonic bath. The samples were further chemically polished using a solution of ammonium hydroxide of 25 cm³ and hydrogen peroxide of 2 cm³ in water of 50 cm³ to remove polishing materials at the surface.

Figure 2 shows the schematic diagram of an ultra-high vacuum joining equipment used. This system consisted of two main chambers for joining and surface analysis. These chambers were connected by traverse chambers and maintained at a vacuum of 10^{-8} Pa. Auger spectroscopy analysis was carried out in the surface analysis chamber. An argon ion bombardment source is also located within this chamber. The joining chamber also involved devices for applying uniaxial load and radio frequency heating.

Fig. 1 Dimensions of specimens for joining. (A) flat specimen (B) spherical specimen

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Fig. 2 Schematic diagram of ultra-high vacuum joining equipment.

The incidence angle for argon ion striking the copper surface was set to 45 degrees. The ion current was measured with a Faraday cage. Table 1 shows the relationship between the accelerating voltage and ion current. The ion current was in the range of 0.35 to 0.57 μA , corresponding to an ion density in the range of 5.5×10^{15} to $8.5 \times 10^{15}/\text{cm}^2 \cdot \text{h}$.

III. Results

1. Selection of materials

The surface contamination due to the residual gas in the system was investigated. Mass spectroscopy showed that the residual gas inside the bonding chamber consisted of significant amount of H_2 and slight amount of CO and CO_2 . When the device for moving a specimen in the chamber was operated, the pressure increased slightly. The released gases were mainly H_2 and slightly H_2O .

Table 2 shows the surface composition of copper and titanium after ion bombardment. In the case of titanium, it was difficult to remove oxide and carbon completely by ion bombardment for 2 h. Traversing the specimens of titanium in the system, the amounts of carbon and oxygen increased. The bonding surface was contaminated by the residual gas.

On the other hand, the surface composition of copper that was cleaned by ion bombardment, did not change after traversing the specimen in the ultra-high vacuum equipment. Vacuum pressures at the bonding and analyzing chambers were in the range of 2 to 7×10^{-8} Pa; at the traversing chamber it was in the range of 3 to 10×10^{-8} Pa.

Copper is difficult to contaminate with the residual gas present in the chamber and therefore, easy to produce a clean surface with argon ion bombardment. Based on these results copper was selected for further experiments.

2. Bonding results

All specimens prepared for bonding were argon ion bombarded for 2 h and the effect of accelerating voltage

Table 1 Argon ion current at various accelerating voltages.

V_a/kV	$I_a/\mu\text{A}$
5	0.35
4	0.40
3	0.48
2	0.54
1	0.57
0.5	0.47

Table 2 Surface composition of copper and titanium after argon ion bombardment (at%). Auger analysis carried out immediately after ion bombardment was compared with analysis of specimens which were traversed to bonding chamber and returned to analyzing chamber.

Cu					
	Ar	C	N	O	Cu
Before Moving	1	0	0	0	99
After Moving	1	0	0	0	99

Ti					
	Ar	C	N	O	Ti
Before Moving	1	7	—	1	91
After Moving	1	25	—	14	60

during argon ion bombardment on joint strength was investigated.

Figure 3 shows the relationship between tensile strength and accelerating voltage. Joints could not be made at room temperature but at 300°C using copper samples treated by argon ion bombardment for 2 h. The joint strength depended on the accelerating voltage during ion bombardment. It was best to bond the samples treated at an accelerating voltage of 2 kV. The joint strength decreased as the accelerating voltage was increased from 2 to 5 kV.

Figure 4 shows the fracture surface of joints formed at

Fig. 3 Effect of argon accelerating voltage on tensile strength of copper joint bonded at temperatures of 20°C and 300°C after ion bombardment.

300°C after argon ion bombardment. The fractograph (Fig. 4A) of the joint not treated by ion bombardment showed poor bonding area. On the other hand, the fractographs (Fig. 4B and C) of the joints treated by ion bombardment using accelerating voltages of 1 and 2 kV showed that the failure mode was ductile. However, the joints treated by an accelerating voltage of 5 kV showed flat fracture surfaces and bonded area was unclear at the fracture surface as shown in Fig. 4D.

IV. Discussion

It is clear from the experimental results that ion bombardment is effective for improving the strength of joints when an accelerating voltage of 2 kV is used, but the joint strength decreases with increasing accelerating voltage above 2 kV. The reasons for this are discussed below.

1. Effect of surface composition

Auger electron spectroscopy was carried out in order to confirm the effect of argon ion bombardment on changes in the surface composition. Table 3 shows the surface composition of copper treated by ion bombardment. There was no difference between compositions of flat specimens and spherical specimens. Carbon and oxygen, which forms surface films, decreases with an increase in accelerating voltage. The ion bombardment

produces a clean bonding surface.

The increase in tensile strength of the joints with a rise in accelerating voltage up to 2 kV is attributed to the removal of surface contaminants.

Figure 5 shows the Auger spectra at the bonding surface and the fracture surface of joints bonded at 300°C after argon ion bombardment at 5 kV. There was no difference between the surfaces. There is also little difference between Auger spectra taken from surfaces treated at the accelerating voltages of 2 and 5 kV. The decrease in joint strength at higher accelerating voltages depends on other factors rather than the purity of the bonding surfaces.

Table 3 Surface composition of copper treated by ion bombardment at various accelerating voltages (at%).

V_a /kV	S	Cl	Ar	C	Ca	N	O	Cu
0	3	2	0	16	1	2	4	72
0.5	0	0	1	1	0	0	2	96
1	0	0	1	1	0	0	1	97
2	0	0	1	0	0	0	0	99
3	0	0	1	0	0	0	0	99
4	0	0	1	0	0	0	0	99
5	0	0	1	0	0	0	0	99

Fig. 4 Fracture surfaces of joints bonded at 300°C after argon ion bombardment for: (A) Argon accelerating voltage 0 kV, (B) 1 kV, (C) 3 kV and (D) 5 kV.

Fig. 5 Auger spectra of bonding surface and fracture surface of joints bonded at 300°C after argon ion bombardment at 5 kV.

2. Effect of surface roughness

It is evident that when energetic ions strike the surface of a material there will be some surface damage as ions penetrate the surface and displace atoms.

Figure 6 shows the effect of accelerating voltage during ion bombardment on the surface roughness. There is an increase in surface roughness with an increase in accelerating voltage. However, it is suggested that the decrease in joint strength was not attributed to the change in surface roughness, because the change in the bonded area was insignificant according to the estimation using a bonding model developed by Hill and Wallach⁽⁵⁾.

3. Effect of hardness

Hardness was measured under various applied loads, using the specimens treated by ion bombardment at the accelerating voltages of 2 and 5 kV.

Figure 7 shows the effect of applied load on the Vickers hardness. In the case of greater applied loads, there is little difference in hardness, but at lower applied loads, the hardness increases and the difference in hardness is larger for the specimens treated at 5 kV.

The depth of indentation is 3.9 μm under an applied load of 100 μN . The increase in hardness toward the uppermost surface shows that a hard layer is formed at the bonding surface. The harder surface was produced for the surface treated by ion bombardment using the higher accelerating voltages.

Figure 8 shows the effect of annealing temperature on Vickers hardness for the specimens treated at the accelerating voltage of 2 and 5 kV. The hardness of the specimens at the accelerating voltage of 5 kV decreased with a rise in annealing temperature, the hardness after annealing at 450°C approached the value prior to ion bombardment.

In order to investigate the effect of annealing temperature on joint strength, the joints were made, after the specimens were ion bombarded by the accelerating voltage of 5 kV. Figure 9 shows the relationship between annealing temperature after ion bombardment and tensile strength of joint. The tensile strength of the joint increased with a rise in annealing temperature. It is found

Fig. 6 Effect of argon accelerating voltage on surface roughness.

Fig. 7 Effect of applied load at hardness testing on Vickers hardness.

Fig. 8 Relationship between Vickers hardness and annealing temperature of the specimens treated at various accelerating voltages.

that the joint strength increases with softening of the bonding surface and the decrease in bondability is attributed to the formation of the hard surface layer.

Figure 10 shows Auger spectra of specimens heated at

300 and 500°C. Although sulfur was detected at 500°C, it could not be measured at the fracture surface of the joints. It is thought that sulfur does not have an effect on the bond strength, because the sulfur diffuses into the base metal from the bond region.

Figure 11 shows the effect of accelerating voltage dur-

ing argon ion bombardment on the surface layer. At the accelerating voltages up to 2 kV, the increase in joint strength depends on the rise in purity of bonding surface. Argon ion bombardment at accelerating voltages from 2 up to 5 kV produces a clean surface but also forms a hard and rough surface layer. The decrease in bondability depends on the formation of this hard surface layer.

V. Conclusions

To study the effect of argon ion bombardment on diffusion bonded joints, the surface composition, surface morphology, hardness and the fracture surfaces were investigated. The following conclusions are drawn from the results obtained.

(1) Joints could be produced not at room temperature but at 300°C using copper samples treated by argon ion bombardment. The joint strength depended on the accelerating voltage used during ion bombardment. It was best to bond the samples treated at the accelerating voltage of 2 kV. The joint strength decreased with a rise in accelerating voltage up to 2 kV.

(2) When ion bombardment was carried out at the accelerating voltages up to 2 kV, the increase in joint strength depended on the removal of contaminants at the bonding surface.

(3) Argon ion bombardment at the higher accelerating voltages from 2 up to 5 kV formed a hard layer. This layer was removed by annealing the samples after argon ion bombardment. With softening of the bonding surface, the joint strength increased. The decrease in bondability was attributed to the formation of a surface layer.

(4) Surface roughness increased with the accelerating voltage. The decrease in joint strength was not attributed to the change in surface roughness, as there was little change in the bonding area.

(5) The use of a surface annealing after ion bombardment was found to be effective in removing any surface damages.

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Fig. 9 Relationship between annealing temperature after ion bombardment treatment and tensile strength of joint.

Fig. 10 Auger spectra of specimens heated at 300 and 500°C after ion bombardment.

Fig. 11 Effect of accelerating voltage at argon ion bombardment on the surface layer.