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Penetrating behavior of eutectic liquid during Al/Cu contact reactive brazing①

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[Abstract] The behavior of eutectic liquid penetrating into the Al base during Al/Cu contact reactive brazing process was studied. Analysis results show that the eutectic liquid prefers to expand along the grain boundary in the depth direction. Meanwhile, dissolution of solid Al and Cu into the eutectic liquid promotes the eutectic reaction and the continuously formed eutectic liquid leads to the reactive penetrating.

[Key words] Al/Cu; contact reactive brazing; eutectic reaction; penetrating; expanding
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1 INTRODUCTION

In the traditional soldering technology, the "big and small enveloping" phenomenon will occur during the flowing of liquid filler metal in the fillet so that the compactness of the seam could not be assured[1]. At beginning of 1970s, the method of contact reactive brazing (CRB) was developed[2~5]. Such method uses the liquid phase formed by eutectic reaction between metals to realize joining. The macro filling process of solder alloy was avoided and no flux is needed, therefore the compactness of the seam is rather high. Meanwhile, the soldering temperature in CRB method is lower and only little pressure is needed so that it is suitable to the soldering assembly where higher geometric precision is required. Up to now, the CRB method has been applied in the assemblies of Al alloys[6,7], high-temperature alloys[8] and Ti alloys[9], the interface joining of composites[4,5] and chip bonding in integrated circuits (IC)[10~12].

Although there are more and more applications of CRB method, the micro-mechanism of contacting reaction is lack of deep investigation. The concept of reactive spreading was proposed in Ref. [6] and the spreading process of liquid Al-Cu eutectic on the Al surface was described. It is pointed out that Al/Cu eutectic reactive spreading is a continuous micro-process that Cu atoms prefer to diffuse along the interface between Al and its oxide layer and then take part in the eutectic reaction continuously. Distinguishing from the traditional soldering technology, CRB method can not rely on the formation of intermetallic compounds to achieve reliable joining. During the contacting reaction, the prefered spreading of eutectic liquid on the surface of base metal is helpful for large area joining[13], but on the other hand, the penetraed spreading of eutectic liquid along the depth direction in the base metal is essential to assure the joining strength. Therefore, the penetrating behaviour of eutectic liquid into Al base metal during Al/Cu CRB process is studied in this work, and the results will be valuable for reliable large-area joining of assemblies.

2 EXPERIMENTAL

The schematic diagram for Al/Cu contact reactive brazing is illustrated in Fig. 1. The 99.99% pure Cu piece with dimension of 2 mm × 2 mm × 0.2 mm was put on the L2 pure Al plate with dimension of 20 mm × 10 mm × 3 mm. The elastic jig is made of stainless steel and high-temperature alloy spring. It

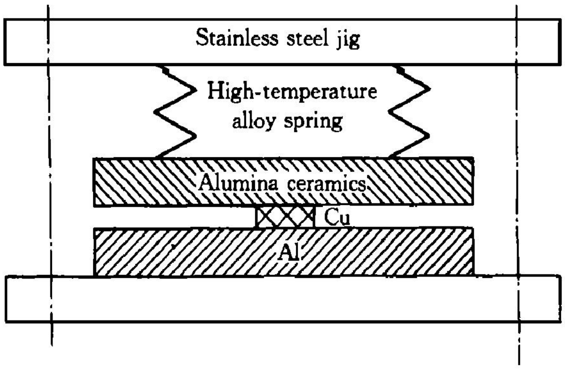

Fig. 1 Schematic diagram for Al/Cu contact reactive brazing

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brings flexible pressure on the soldering assembly through alumina ceramic piece and then keeps the tight contacting of Al/Cu interface all the time during the soldering process. The test was carried out in Ar atmosphere and Mg pieces were put surrounding the assembly in order to further reduce the partial pressure of oxygen. Soldering temperature was 580 ± 10 ℃ and holding time was 10 min.

3 PENETRATING BEHAVIOUR

The penetrating of Al-Cu eutectic liquid into Al base metal at the initial stage of contact reaction is shown in Fig. 2. It is obvious that the eutectic liquid prefers to spread along the grain boundary in the depth direction. Fig. 3 shows the SEM morphology of cross-section of the sample after Al-Cu contact reaction. The compositions of arbitrary four points in the white area which distributes as network are analysed and the results are 59.5Al+40.5Cu, 70.1Al+29.9Cu, 73.9Al+26.1Cu and 59.7Al+40.3Cu (mass fraction, %), respectively. Since the eutectic point of Al-Cu is 66.8Al+33.2Cu (mass fraction, %), the penetrating of eutectic or near eutectic liquid into the Al base metal is presented as network. The penetrating front prefers to spread along the grain boundary. In the middle area, the grain boundary seems to be wider due to the eutectic reaction. In the initial area, the eutectic reaction is extended into the grains, for example, in the depth range of tens of micrometers under the surface of Al base metal, the metallurgical microstructure is made of Al-Cu eutectic and pure Al phases.

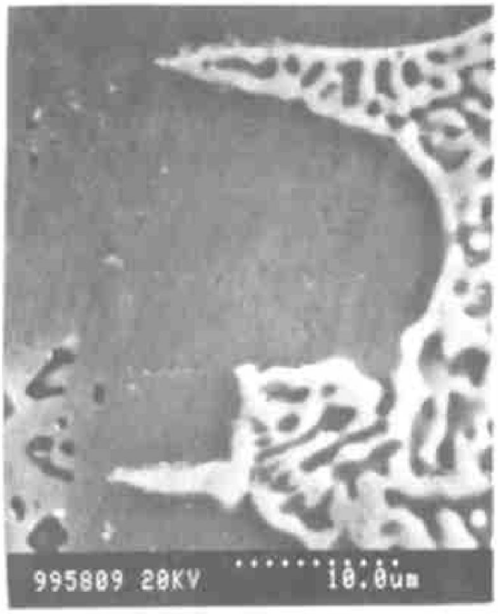

Fig. 2 SEM morphology of eutectic liquid penetrating at initial stage of Al/Cu contact reaction

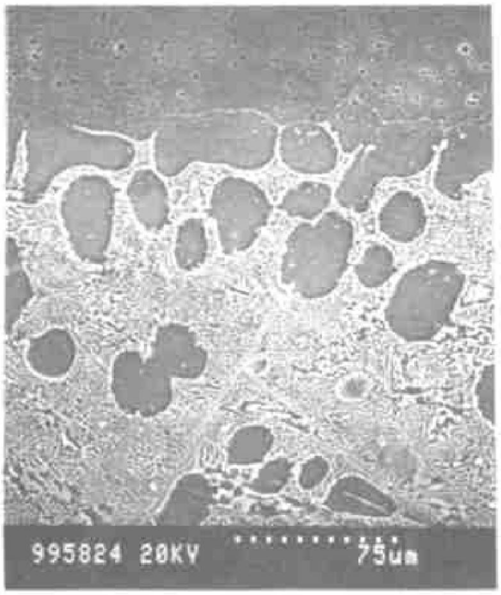

Fig. 3 SEM morphology of cross-section of sample after Al/Cu contact reaction

4 DRIVING FORCE FOR REACTIVE EXPANDING

From the view of thermodynamics, the Al atoms at the grain boundary have higher chemical potential. Meanwhile, the system free energy will decrease if the eutectic reactions take place between Cu atoms and Al atoms at the grain boundary. The above chemical potential gradient and energy tendency are the driving force for eutectic liquid to expand along grain boundary. From the view of energy, the interface energy to be overcome for the matter in liquid phase to diffuse through liquid/solid interface into solid phase is much more than the matter in solid phase to diffuse through solid/liquid interface into liquid phase[14]. Fig. 4 shows the expanding front of eutectic liquid. The EDS analysis results of these four

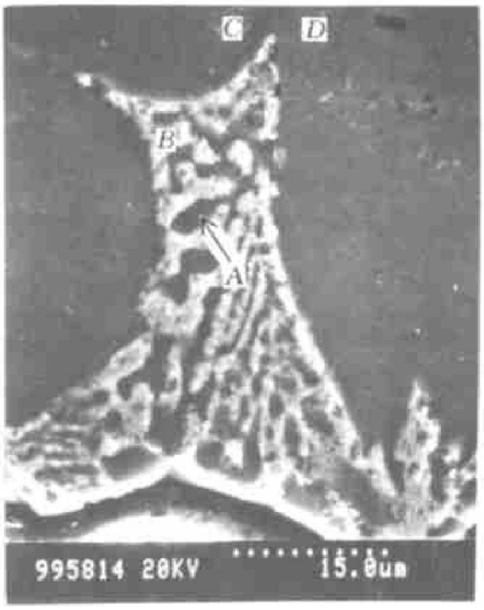

Fig. 4 SEM morphology of expanding front of eutectic liquid

(*A* —dark region among white eutectic phases; *B* —white region with eutectic phases; *C* and *D* —minute regions near sharp angle of expanding front of eutectic liquid) regions are listed in Table 1. It can be seen that the Cu content in *A*, *C* and *D* regions is very little, that is, it is difficult for Cu atoms in eutectic liquid to diffuse through liquid/solid interface into Al base metal. Therefore, after the liquid forms at the contacting points, the driving force for eutectic reaction to proceed continuously should be the diffusion and dissolution of Al and Cu atoms into eutectic liquid across the solid/liquid interface.

Table 1 Compositions of different regions in expanding front of eutectic liquid

Region	$w(\mathrm{Al})/\%$	$w(\mathrm{Cu})/\%$
A	99.16	0.84
B	78.83	21.17
C	99.08	0.92
D	99.01	0.99

5 CONCLUSIONS

The penetrating of eutectic liquid into Al base metal during Al/Cu CRB prefers to expand along the grain boundary. With the soldering time increasing, the eutectic reactions gradually expand from the grain boundary into the grains. The dissolution of solid Al and Cu atoms into eutectic liquid makes the eutectic reaction and the liquid formation proceed continuously. Consequently, the eutectic liquid penetrates and expands into the Al base metal.

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