Elucidation of wetting mechanism of aluminum brazing on silicon nitride by high-temperature wettability tests

Takumi Kusumoto^{1*1}, Mitsuhiro Ootaki¹, Shoichi Hirosawa¹

¹ Department of Mechanical Engineering and Materials Science, Yokohama National University, 79-5 Tokiwadai, Hodogaya-ku, Yokohama 240-8501, Kanagawa, Japan.

Wetting mechanism of molten 2N(99 %)-, 5N(99.999 %)-purity aluminum and Al-12.6 % Si alloy on silicon nitride (Si₃N₄) substrate was investigated by conducting high-temperature wettability tests with a sessile drop method and extrusion method. From the results of the sessile drop experiment, it was found that oxide films on the surface of aluminum (alloy) droplet disappear at 1300-1400 °C, and then their wettability was improved with wetting angles of 37-44 °. The formation of aluminum nitride (AlN) reaction layers between aluminum droplet and Si₃N₄ substrate and the dissolution of Si from Si₃N₄ substrate into the droplet were also responsible for the improved wettability because wetting angles of molten aluminum are much smaller on AlN than on Si₃N₄, and surface tension between molten aluminum and the surrounding atmosphere is decreased with increasing the amount of dissolved Si, respectively. From the results of the extrusion experiment, on the other hand, oxide film formed after extrusion of droplet was found to move upwards by convection flow, resulting in the improved wettability even at lower temperatures of 1100-1300 °C. Therefore, the formation and removal of oxide film could be controlled by optimizing temperatures and oxygen pressure during aluminum brazing.

Keywords: Wettability, Joining, Aluminum alloy, Silicon nitride ceramics, dissimilar material bonding

1. Introduction

Bonding of aluminum heatsink and ceramic substrates with aluminum brazing is an important technology for electrical components such as power semiconductors because heat generated at electric circuits has to be quickly removed from the substrates. However, thin aluminum oxide film formed on the surface of molten aluminum lowers its wettability, and thus degrades the resultant joining strength between aluminum brazing and ceramic substrate. Although under lower oxygen partial pressures, such an oxide film turns into a gas of Al₂O above a specific temperature¹⁾, aluminum brazing is generally conducted below the temperature, so the extent to which the oxide film inhibits wetting and spreading of molten aluminum becomes important. In this study, wetting mechanism between molten 2N(99%)-, 5N(99.999%)- purity aluminum or Al-12.6 %Si alloy and silicon nitride (Si₃N₄) substrate was investigated by high-temperature wettability tests with a sessile drop method and extrusion method. Not only the effect of oxide film but also the formation of reaction layers between aluminum droplet and Si₃N₄ substrate as well as the mass transfer of dissolved Si from Si₃N₄ substrate to molten aluminum were considered.

2. Experimental procedures

In this study, small pieces of 2N-, 5N-purity aluminum and Al-12.6%Si alloy, and Si₃N₄ discs with 10 mm^{ϕ} x 0.63 mm^t were utilized. High-temperature wettability tests were performed in an Ar inert gas atmosphere with a sessile drop method and extrusion method. In the sessile drop experiment, each solid sample on a Si₃N₄ substrate was heated in steps from 1100 to 1450 °C, and the temperature dependence of wetting angles was measured from the shapes of aluminum droplet. In the extrusion experiment, on the other hand, molten sample within a heated specimen holder was extruded by a piston through a hole of 0.5 mm^{ϕ} in the center of a Si₃N₄ substrate, and then held on the Si_3N_4 substrate at isothermal temperatures of 900, 1100 and 1300 °C for clarifying the formation behavior of oxide film from clean surface of molten aluminum. The solidified microstructures after wettability tests were observed by optical microscopy and scanning electron microscopy (SEM) with an energy-dispersive X-ray spectroscopy (EDS).

3. Results and discussion

From the sessile drop experiment, it was found that, irrespective of compositions of aluminum samples, oxide film disappears at 1300-1400 °C, and thus their wettability can be improved with wetting angles of 37-44°. SEM/EDS analysis around interfaces between solidified aluminum and Si_3N_4 substrate revealed that aluminum nitride (AlN) reaction layers are formed in the Si₃N₄ side of the interface with a thickness of $\sim 40 \,\mu m$. Another interesting feature was also found from the results of optical microscopy observation; i.e. the dissolution of Si from Si₃N₄ substrate into aluminum droplet. These findings suggest that the improved wettability is also attributed to the two microstructural features because wetting angles of molten aluminum are much smaller on AlN than on Si₃N₄, and surface tension between molten aluminum and the surrounding atmosphere is decreased with increasing the amount of dissolved Si, respectively.

In the extrusion experiment, on the other hand, no oxide film was observed just after extrusion, but oxide film was formed in a few to a few tens of seconds after exposure to residual gas within the evacuated chamber. At 900 °C, the sample was oxidized rapidly after extrusion, with the entire surface of aluminum droplet covered with oxide film, resulting in an inferior wettability. At 1100 and 1300 °C, however, the sample was slowly oxidized from triple points of aluminum droplet, Si₃N₄ substrate and the surrounding atmosphere, and then thin ribbon of oxide film formed at the bottom of the droplet has moved upwards by convection flow. This result indicates that the wettability of molten aluminum can be improved even at lower temperatures of 1100-1300 $^\circ C$ by optimizing temperatures and oxygen pressure during aluminum brazing.

References

1) S.-J. Park, H. Fujii and H. Nakae: J. Japan Inst. Metals 58 (1994) 208–214.