Effects of Alloying Elements to Aluminium on the Wettability of AL/SiC System

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Abstract

The wettability at a liquid Al-alloy/SiC interface was evaluated by the sessile drop method at 750°C. The wetting angle, \( \theta \), of a sessile drop on SiC substrate decreased with the addition of Pb, Mg and Ca to pure aluminium. The reduction in \( \theta \) of the Al-Pb alloy was proportional to the reduction in surface tension, \( \gamma_{lv} \), of aluminium, whereas in Al-Mg and Al-Ca alloys the reduction in \( \theta \) was greater than the reduction in \( \gamma_{lv} \) of Al. This was attributed to reactions that took place at the Al-alloy/SiC interface. Scanning electron microscopy (SEM) analysis showed that severe interactions had taken place at the interfaces of Al-Mg alloy/SiC and Al-Ca alloy/SiC.

Key Words: Wettin, Sessile drop method, Surface tension, Aluminium alloys

Introduction

The wettability of ceramic surfaces by Al or its alloys has been the subject of a number of studies (Kohler, 1975; Delannay et al., 1987; Laurent et al., 1987; Laurent et al., 1988; Nakae et al., 1992; Han et al., 1993; Asthana, 1994; Asthana, 1995; Laurent et al., 1996; Fujii and Nakae, 1996; Drevet, 1996). In the fabrication of metal matrix composites (MMCs) and the joining of ceramics to aluminium and its alloys, wettability is the most important variable in processing industries where a molten aluminium is used. The wettability is determined by the wetting angle \( \theta \) of a sessile drop resting on a substrate given by Young-Dupré equation as follows:

\[
\cos \theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}}
\]

where \( \gamma_{sv} \) is the surface tension of the solid, \( \gamma_{sl} \) is the solid/liquid interfacial energy and \( \gamma_{lv} \) is the surface tension of the liquid. A liquid is said to wet a solid when \( \theta \) is smaller than 90°, while in non-wetting systems \( \theta \) is larger than 90°. Recent investigations (Laurent, 1987; Laurent, 1988; Asthana, 1994; Drevet, 1996) showed that Al exhibits a non-wetting nature below 1123K on most ceramics. Laurent et al. (1987) studied the wettability of SiC by Al-Si alloys. Han et al. (1993) investigated the effect of free Si in the carbide and of Mg, Si and Cu alloy additions to Al in a SiC/Al-alloy system.

Limited data is found in the literature about the effect of Al-alloy/SiC systems. In particular, no data have yet been published on \( \theta \) changes on SiC induced by Ca and Pb additions to pure Al as alloying additions. Thus our aim was to study the influence of alloying elements to aluminium on the wettability of Al/SiC system.

Experimental work

Pure Al (99.95%), Al-1.4Pb, Al-0.8Ca, Al-3.4, 8.6 and 13.9Mg alloys were used for sessile drop experiments. Al-alloys were prepared by melting in an atmosphere controlled furnace and cast into a 20mm diam. chill cast ingot. The sintered SiC used in our experiments was supplied by Wacker Chemicals Ltd, UK., with a nominal composition of 98.5 wt% SiC, 1.0 wt% free carbon and 0.3 wt% Al as sintering aids and trace amounts of oxygen and nitrogen.
A schematic of the wetting equipment is given in Figure 1. Basically, it consists of a mullite work tube in a resistance furnace and a vacuum unit. The mullite tube has an internal diameter of 25mm and is 675mm in length, sealed at both ends by flanges and “O” ringed glass windows. The SiC material was slit with a diamond saw to 12x12x5mm for wettability studies. One flat face of the samples was ground by 400, 800 and 1000 grade SiC papers followed by polishing sequentially down to 6, 3, 1 and 1/4μm diamond solutions. Alloy pieces approximately 2g in weight were cut from ingot materials. Each sample was ground to a cubic shape, then immediately immersed into dry methanol to minimise the formation of oxide film on the surface and then ultrasonically cleaned. The SiC was placed on an alumina boat with the polished surface upwards, the rest of the alumina boat being filled with titanium sponge to act as a getter for residual oxygen in the vacuum environment. The experiments were carried out under vacuum (10^{-4}/10^{-5} torr). A camera unit was set up in front of the observation window to record the image of the metallic drop. Photographs of sessile drops at rest on SiC were taken at 0, 5, 15, 30 and 60 minute intervals after the temperature of the sessile drop reached 750°C. Immediately after the photographs were taken, the furnace was switched off. As the temperature dropped to ~50°C, the system was let up to atmosphere and the sample was removed. The dimensions of the sessile drop were derived from the printed photographs. Details of calculations of θ from sessile drops have been explained elsewhere (Candan, 1998). In order to examine the interface, a vertical slice was taken from the middle of the sessile drop and ceramic substrate. The sample was then cold mounted, sequentially ground by 400, 800 and 1000 grade SiC papers and polished to 6, 3, 1 and 1/4μm diamond solutions followed by etching in 2M NaOH for 1 min.

Results

Figure 2 shows that θ of aluminium alloys on SiC was sequentially reduced by the addition of 1.4Pb, 3.4Mg, 0.8Ca, 8.6Mg and 13.9Mg to pure aluminium. The most significant reduction in θ was observed with Al-8.6Mg and Al-13.9Mg alloys respectively, in which the wetting condition was achieved. In all cases, θ decreased with increasing contact time at 750°C, with the largest decrease in the first five minutes, after which it decreased more slowly. Figures 3 to 5 show SEM micrographs of sessile drops of pure Al, Al-13.9Mg and Al-0.8Ca alloys on SiC substrates respectively indicating that interaction had taken place between the Al-alloy and the SiC substrate interface. The most severe interaction had taken place in the Al-13.9Mg alloy/SiC and Al-0.8Ca alloy/SiC system, as shown in Figures 4 and 5 respectively.

Discussion

The wetting angle, θ, of pure Al on SiC was 123° after thirty minutes contact at 750°C. Table 1 shows that this is in good accord with the results of Han et al. (1993) but is different from the results of Laurent et al. (1987) and Kohler (1975). The differences in wetting angle may arise from combinations of the purity of Al selected, the type of SiC substrate used (reaction bonded RBSiC, sintered SSiC, single crystal SCSiC etc.) and the level of vacuum employed. Han et al. (1993), for example, reported that θ on RBSiC was 82° while on SSiC it was 127° for otherwise identical conditions.
Increased contact time in the present study reduced \( \theta \) in agreement with many published works (Laurent et al., 1987; Nakae et al., 1992; Han et al., 1993; Asthana, 1995; Fujii and Nakae, 1996; Drevet, 1996). This was attributed to the increased extent of reaction, which led to a decreased \( \theta \) as supported by Figure 3. From the literature (Delannay et al., 1987; Asthana, 1994), the driving force for wetting is affected by only two factors: surface tension of the liquid, \( \gamma_{lv} \), and the strength of the solid-liquid interaction at the interface, which leads to a reduced interface tension \( \gamma_{ls} \). Previous work (Candan et al., 1997a; 1997b) showed that the formation of \( \text{Al}_4\text{C}_3 \) had occurred in pure aluminium and aluminium alloy infiltrated SiC powder compacts due to interfacial reactions. Iseki et al. (1984) also reported that \( \text{Al}_4\text{C}_3 \) forms at the interface of the Al/SiC system.

### Table 1. Contact angles for the Al/SiC system presented by different researchers for 30 minutes contact at 750°C (SCSiC = Single crystal SiC, RBSiC = Reaction bonded SiC, SSiC = Sintered SiC).

<table>
<thead>
<tr>
<th>Author</th>
<th>Contact angle ( \theta ), (degrees)</th>
<th>Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kohler (1975)</td>
<td>157</td>
<td>99.99% Al, SCSiC, 10^{-6} torr, Zr-gettered</td>
</tr>
<tr>
<td>Laurent et al. (1987)</td>
<td>90</td>
<td>99.999% Al, SCSiC, 10^{-6} - 10^{-5} torr,</td>
</tr>
<tr>
<td>Han (1993)</td>
<td>82</td>
<td>99.99% Al, RBSiC, 10^{-4} - 10^{-5} torr, Ti-gettered</td>
</tr>
<tr>
<td>Han (1993)</td>
<td>127</td>
<td>99.99% Al, SSiC, 10^{-4} - 10^{-5} torr, Ti-gettered</td>
</tr>
<tr>
<td>Present work</td>
<td>123</td>
<td>99.95% Al, SSiC, 10^{-4} - 10^{-5} torr, Ti-gettered</td>
</tr>
</tbody>
</table>
The progressive reduction in $\theta$ by increasing Mg addition to pure aluminium is attributed to both reduction in $\gamma_{lv}$ of Al and increased interfacial reactions at the solid-liquid interface. Previously, Candan et al. (1997a; 1997b) reported that the threshold pressure for infiltration of molten Al into SiC particle compacts was proportionally reduced by Mg additions. This was attributed to a reduction in $\gamma_{lv}$ of Al and increased interfacial reactions at the solid-liquid interface. Previously, Candan et al. (1997a; 1997b) reported that the threshold pressure for infiltration of molten Al into SiC particle compacts was proportionally reduced by Mg additions. This was attributed to a reduction in $\gamma_{lv}$ of Al and increased interfacial reactions at the solid-liquid interface. Previously, Candan et al. (1997a; 1997b) reported that the threshold pressure for infiltration of molten Al into SiC particle compacts was proportionally reduced by Mg additions. This was attributed to a reduction in $\gamma_{lv}$ of Al and increased interfacial reactions at the solid-liquid interface. Previously, Candan et al. (1997a; 1997b) reported that the threshold pressure for infiltration of molten Al into SiC particle compacts was proportionally reduced by Mg additions. This was attributed to a reduction in $\gamma_{lv}$ of Al and increased interfacial reactions at the solid-liquid interface. Previously, Candan et al. (1997a; 1997b) reported that the threshold pressure for infiltration of molten Al into SiC particle compacts was proportionally reduced by Mg additions. This was attributed to a reduction in $\gamma_{lv}$ of Al and increased interfacial reactions at the solid-liquid interface. Table 2. Derived values of surface tension, $\gamma_{lv}$, of Al alloys from the works of Korol’kov (1963) and Lang (1974) and wetting angles, $\theta$, of present work together with percentage reductions in $\gamma_{lv}$ and $\theta$ by alloying additions to pure Al for 5 minutes contact at 750°C.

![Figure 6. X-ray diffraction patterns for Al-0.8Ca/SiC system. (Key: $\bullet = \alpha$ Al, $\Delta = $ SiC, $\Box = \text{Al}_2\text{Si}_2\text{Ca}$, $\blacktriangle = \text{Al}_4\text{C}_3$, $\blacksquare = \text{Si}$)](image)

**Conclusions**

The wetting angle, $\theta$, decreased in the Al-alloy/SiC system with the addition to pure Al of 0.8Ca, or 1.4Pb and 3.4, 8.6 and 13.9Mg. The wetting condition was achieved in 8.6 and 13.9Mg alloys. Reductions in the wetting angle were attributed to the relative effects of these additions on $\gamma_{lv}$ and $\gamma_{st}$. The wetting angle, $\theta$, with SiC decreased with time for pure Al, Al-0.8Ca, Al-1.4Pb and Al-3.4, 8.6 and alloys, attributed to continuing interaction 13.9Mg
at the interface. The results of the present work indicated that the effect of the reaction in reducing interface tension $\gamma_{ls}$ dominates any corresponding reduction in $\gamma_{lv}$ for a reduction in $\theta$.

References


