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Effect 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, θ, of a sessile drop on SiC substrate decreased with the addition of Pb, Mg and Ca to pure aluminium. The reduction in θ of the Al-Pb alloy was proportional to the reduction in surface tension, γ_{lv}, of aluminium, whereas in Al-Mg and Al-Ca alloys the reduction in θ was greater than the reduction in γ_{lv} of Al. This was attributed to reactions that took place at the Al-alloy/SiC interface. Scanning electron microcopy (SEM) analysis showed that severe interactions had taken place at the interfaces of Al-Mg alloy/SiC and Al-Ca alloy/SiC.

Key Words: Wetting, 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 θ 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 γ_{sv} is the surface tension of the solid, γ_{sl} is the solid/liquid interfacial energy and γ_{lv} is the surface tension of the liquid. A liquid is said to wet a solid when θ is smaller than 90°, while in non-wetting systems θ 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 θ of Al-alloy/SiC systems. In particular, no data have yet been published on θ 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 $1/4\mu$m diamond solutions. Alloy pieces approximately 2 g 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 $\sim$ 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 $1/4\mu$m 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 by with Al-8.6Mg and Al-13.9Mg alloys respectively, in which the 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.

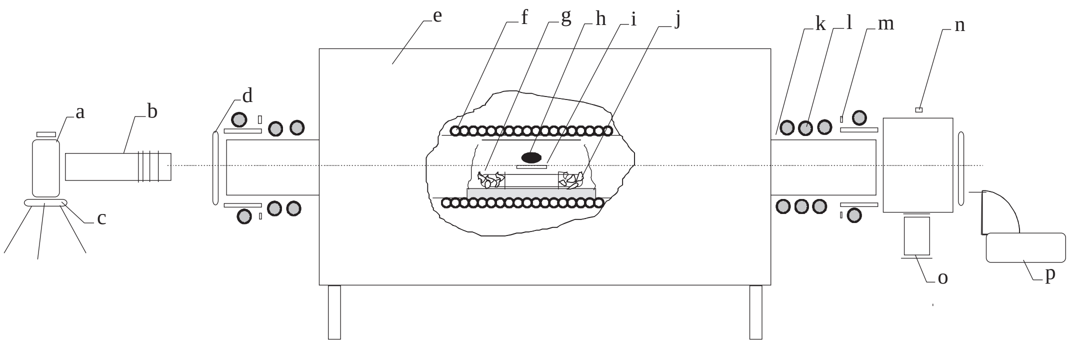

Figure 1. Schematic of sessile drop apparatus.
Key: a. Camera, **b**. Photo lens, **c**. Tripod, **d**. Observation window, **e**. Furnace body, **f**. Nichrome wire heating element, **g**. Alumina boat, **h**. Sessile drop, **i**. Ceramic substrate, **j**. Getter, **k**. Mullite tube, **l**. Cooling pipe, **m**. Clamp, **n**. Air admittance valve, **o**. To diffusion pump, **p**. Fibre optic illuminator.

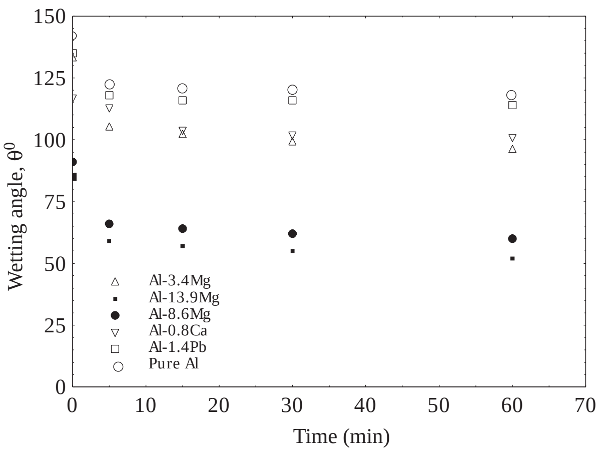

Figure 2. Contact angle, θ, versus time for Al-alloy/SiC systems in vacuum at 750°C.

Figure 3. SEM micrograph showing the etched interface between pure Al sessile drop and sintered SiC (Etched in 2M NaOH).

Increased contact time in the present study reduced θ 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 θ 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, γ_{lv}, and the strength of the solid-liquid interaction at the interface, which leads to a reduced interface tension γ_{ls}. Previous work (Candan et al., 1997a; 1997b) showed that the formation of Al_4C_3 had occurred in pure aluminium and aluminium alloy infiltrated SiC powder compacts due to interfacial reactions. Iseki et al. (1984) also reported that Al_4C_3 forms at the interface of the Al/SiC system.

Figure 4. SEM micrograph showing the etched interface between Al-13.9Mg alloy sessile drop and sintered SiC (Etched in 2M NaOH).

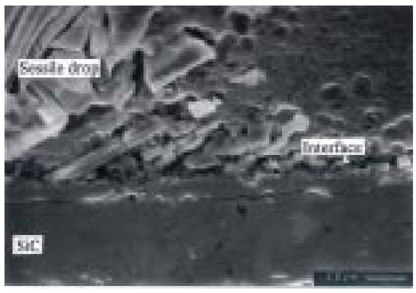

Figure 5. SEM micrograph showing the etched interface between Al-0.8Ca alloy sessile drop and sintered SiC (Etched in 2M NaOH).

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).

Author	Contact angle θ, (degrees)	Conditions
Kohler (1975)	157	99.99% Al, SCSiC,10^{-6}torr, Zr-gettered
Laurent et al. (1987)	90	99.9999% Al, SCSiC, 10^{-6} - 10^{-7} torr,
Han (1993)	82	99.99% Al, RBSiC,10^{-4} -10^{-5} torr, Ti-gettered
Han (1993)	127	99.99% Al, SSiC, 10^{-4} -10^{-5} torr, Ti-gettered
Present work	123	99.95% Al, SSiC, 10^{-4} -10^{-5} torr, Ti-gettered

The progressive reduction in θ by increasing Mg addition to pure aluminium is attributed to both reduction in γ_{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 γ_{lv} of Al and modification of the oxide film. Derived values of γ_{lv} of Al from the works of Korol'kov (1963) and Lang (1974) and the θ values of the present work, together with percentage reductions in γ_{lv} and θ by alloying additions are presented in Table 2. However, the addition of 3.4Mg and 1.4Pb had a similar effect in reducing γ_{lv} of Al (27 and 26% respectively); the degree of reduction in θ is larger in the 3.4Mg added alloy (see Table 2). The 7% reduction in θ by the addition of 1.4Pb was attributed to the reduction in γ_{lv} of Al, while 11% reduction was attributed to both the reduction in γ_{lv} of Al and increased interfacial reactions promoted by Mg additions. Evidently, the wetting condition (i.e. $\theta < 90°$) was achieved by the addition of 8.6 and 13.9Mg, as shown in Figure 2. The rate of reduction in γ_{lv} by additions of 8.6 and 13.9Mg are 35 and 40% respectively, whereas the reduction rates in θ are much higher (48 and 54% respectively). This was attributed to the severe interaction that occurred between the Al-13.9Mg alloy and SiC, as shown in Figure 4. Previous research (Candan et al., 1997a; 1997b) showed that a Mg_2Si intermetallic phase had occurred due to the dissolution of SiC at the interface. Naidich (1981) argued that intermetallic phases wet ceramic surfaces better than metals. Gribkow (1995) reported that an enrichment of Mg at the Al/SiC interfaces had occurred. Wetting angles of θ for the Al-Ca alloy/SiC system do not appear to have been reported previously. The rate of reduction in γ_{lv} of Al by the addition of 0.8Ca to Al is only 8%, while its effect in reducing the rate of θ is much higher (17%). The reduction in θ by the addition of Ca to pure Al is also attributed to both a reduction in γ_{lv} and a strong reaction between Al-Ca alloy and SiC. Figure 5 shows that a severe reaction had taken place at the interface leading to the formation of clustered phases. The clustered phase was identified as an Al_2Si_2Ca phase by XRD analysis (Figure 6). The formation of an Al_2Si_2Ca phase has also been reported in SiC particle compacts infiltrated by Al-Ca alloy (Candan et al. 1997a).

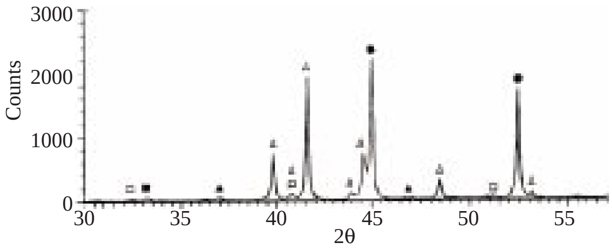

Figure 6. X-ray diffraction patterns for Al-0.8Ca/SiC system. (Key: ● = α Al, Δ = SiC, □ = Al_2Si_2Ca, ▲ = Al_4C_3, ■ = Si)

Conclusions

The wetting angle, θ, 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 γ_{lv} and γ_{sl}. The wetting angle, θ, 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

Table 2. Derived values of surface tension, γ_{lv}, of Al alloys from the works of Korol'kov (1963) and Lang (1974) and wetting angles, θ, of present work together with percentage reductions in γ_{lv} and θ by alloying additions to pure Al for 5 minutes contact at 750°C.

Alloy	Surface tension γ_{lv} (dyn/cm)	Contact angle $\theta°$	% reduction in γ_{lv} (dyn/cm)	% reduction in angle $\theta°$
Pure Al	862 (Korol'kov 1963)	127	0	0
Al-1.4Pb	640 (Korol'kov 1963)	118	26	7
Al-3.4Mg	630 (Korol'kov 1963)	113	27	11
Al-8.6Mg	560 (Korol'kov 1963)	66	35	48
Al-13.9Mg	520 (Korol'kov 1963)	59	40	54
Al-0.8Ca	791 (Lang 1974)	105	8	17

at the interface. The results of the present work indicated that the effect of the reaction in reducing interface tension γ_{ls} dominates any corresponding reduction in γ_{lv} for a reduction in θ.

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