Study on contact angles of Au, Ag, Cu, Sn, Al and Al alloys to SiC

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Contact angles of Au, Ag, Cu, Sn, Al and Al alloys to SiC were measured by use of sessile drops heated by a high frequency induction coil designed to be convex against the SiC plate. Three crystal configurations of α-SiC, polycrystalline plane of sintered SiC, SiC (1 1 1) plane, and SiC (1 0 0 0) plane were used as base plates. Au, Sn, Al and Al alloys showed a large contact angle of about 150° at each melting temperature, however those of Ag and Cu were in the range of 105–121°. Every contact angle of Al and its alloys decreased to under 90° when held at 1350 °C. SiC (1 0 0 0) plane gave a lower contact angle than the other two SiC planes for Cu, Al and Al–Si alloys.

1. Introduction

As is generally known, silicon carbide is very hard and strong, and has a very high thermal conductivity and a high melting temperature [1]. Recently fibre and whisker types of silicon carbide have been industrially produced and applied to composite materials. For further development of new composite materials, it is very important to know the wettability of the interface between metal and silicon carbide.

The interaction of the interface between molten copper and silicon carbide was studied by Gnyeshin and Naydich [2] and those of other metals were reported by Naidich and Nyevodnik [3]. It is known that there are both α-SiC and β-SiC in the crystal system of silicon carbide, and α-SiC has many crystal forms which differ in the arrangement of the atom layers. Therefore, the interfacial interaction should vary with crystal plane.

This is a study of the contact angle between the metals Au, Ag, Cu, Sn, Al and Al alloys and three kinds of α-SiC crystal planes.

2. Experimental procedure

2.1. Samples

2.1.1. Silicon carbide

α-SiC used as a base plate was about $14 \times 14\ mm^2$ in area and 2 mm in thickness. Table I shows three kinds of α-SiC used for these measurements. SiC (sintered), symbolized by △, refers to the polycrystalline plane of sintered silicon carbide, SiC (1 1 1), symbolized by ●, refers to the (1 1 1) plane of 15R type, and SiC (1 0 0 0), symbolized by ○, refers to the (1 0 0 0) plane of 6H type.

The crystal structures of these SiC planes were identified by X-ray analysis. Each plane was polished with woollen cloth, then washed with acetone under ultrasonic vibration. Surface roughness of the plane was measured by a profilometer equipped with a scanning needle probe of 5 μm tip diameter. Table II shows the surface roughness of planes.

2.1.2. Metal and alloy for liquid drop

Granules of about 1–2 mm in diameter were thoroughly cleaned and used for the liquid drops. Table III shows purity and chemical composition of the metals and alloys for the drop.

2.2. Measuring equipment

The metal drop was mounted on the silicon carbide plate in the constant temperature zone of a belljar-type vacuum furnace. The drop was observed through the glass window in the lid of the vacuum vessel, then photographed to measure the contact angle. The drop and its surroundings are illustrated in Fig. 1. To check the accuracy of the contact angle obtained, a model of a drop having known contact angle was used for comparison. The error for this measuring system was within $\pm 2°$. The optical axis of a camera was located to coincide with the horizontal line of the silicon carbide plate, and then the picture of the drop was taken on 35 mm film and enlarged. The photographed silhouette of a drop is shown in Fig. 2. We assumed the angle θ between line BC and the tangent line at B of the silhouette made the contact angle, and by drawing a circle through B and C with centre O and with the line AB perpendicular to the line OB, then the angle θ was measured using a protractor.

The sample system was heated by a high frequency induction coil rated at 10 kW and 400 kHz. The induction coil was designed to be convex against the silicon carbide plate with a drop in order to keep their

0022–2461

TABLE I Crystal structures of α-SiC planes used for the measurement of wettability

Type and symbol	Crystal structure
SiC (sintered) -△-	Polycrystalline plane of sintered silicon carbide
SiC (1 1 1) -●-	(1 1 1) plane of 15R type
SiC (1 0 0 0) -○-	(1 0 0 0) plane of 6H type

TABLE II Roughness of SiC plates

Type	Maximum depth (μm)	Average depth (μm)
SiC (sintered)	2.9	2.0
SiC (1 1 1)	0.78	0.35
SiC (1 0 0 0)	unmeasurable owing to smooth surface	

TABLE III Purity and chemical composition of metals and alloys used for making liquid drops (wt %). Purity: Au (99.99%), Ag (99.999%), Cu (99.99%), Sn (99.99%), Al (99.99%)

Al alloys	Chemical composition				
	Si	Fe	Cu	Mn	Mg
1[a]	1.12	0.015	0.005	0.001	not detected
2[a]	5.95	0.053	0.005	0.002	not detected
3[a]	14.68	3.20	0.005	0.026	0.002
4[b]	0.46	0.010	0.002	0.001	0.88

[a]Al–Si system alloy.
[b]Corrosion resistant alloy, 63S.

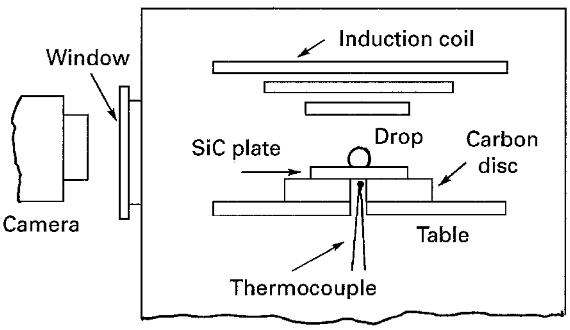

Figure 1 Schematic illustration of the sample and its peripheral situation. The sample was positioned just under the induction coil through rotation of the table operated from outside of the vessel without breaking vacuum.

temperature uniform as shown in Fig. 1. The metal–silicon carbide system was placed on the carbon disc so that the induced heat of the carbon disc could be used to initially heat the sample system. After reaching a certain temperature, the sample system was directly heated by the induction coil and kept at the desired temperature by controlling the current. The temperature was measured and controlled by a Pt–PtRh thermocouple attached just under the SiC plate and an optical pyrometer. Contact angles were measured at both the melting point temperature of the metals or the alloys and 1350 °C under a vacuum of 10^{-5} mmHg.

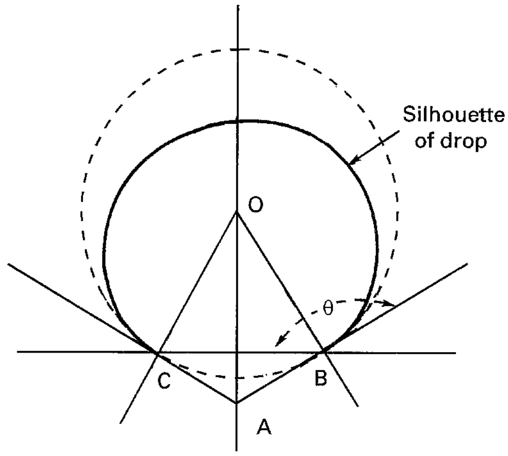

Figure 2 Geometrical process to obtain the contact angle.

3. Experimental results and explanation
3.1. Contact angle of Au, Ag, and Cu to SiC plate
Fig. 3 shows the time dependence of contact angles for Au, Ag, Cu on SiC whilst being held at their melting temperature. The following results were obtained.

3.1.1. Au
There was little difference in the contact angles for Au on both the SiC (sintered) and SiC (1 0 0 0). Moreover, no trace due to interfacial reactions could be found at the Au–SiC interface after cooling from the melting temperature.

3.1.2. Ag
Each point shown in Fig. 3 for this system was the average of four measurements, using a new Ag granule each time. The scatter in the measured values was small, less than $\pm 1°$. The contact angle between Ag and SiC (sintered) was larger than those for SiC (1 1 1) and SiC (1 0 0 0), and the maximum difference was 10°. These contact angles changed little in the temperature range from the melting point of Ag to 1350 °C, but these are not shown in Fig. 3. An Ag drop could be easily separated from the SiC plate after cooling and no trace due to interfacial reactions was observed on their surfaces.

3.1.3. Cu
Each point in Fig. 3 was the average of two measurements, using a new Cu granule each time. The contact angles of Cu to the three kinds of SiC were very different as shown in Fig. 3. A black layer of about 0.5 mm thickness was observed at the interface between Cu and SiC (sintered) and silicon was detected in the copper side by X-ray microanalysis. Similar phenomena were observed in both SiC (1 1 1) and SiC

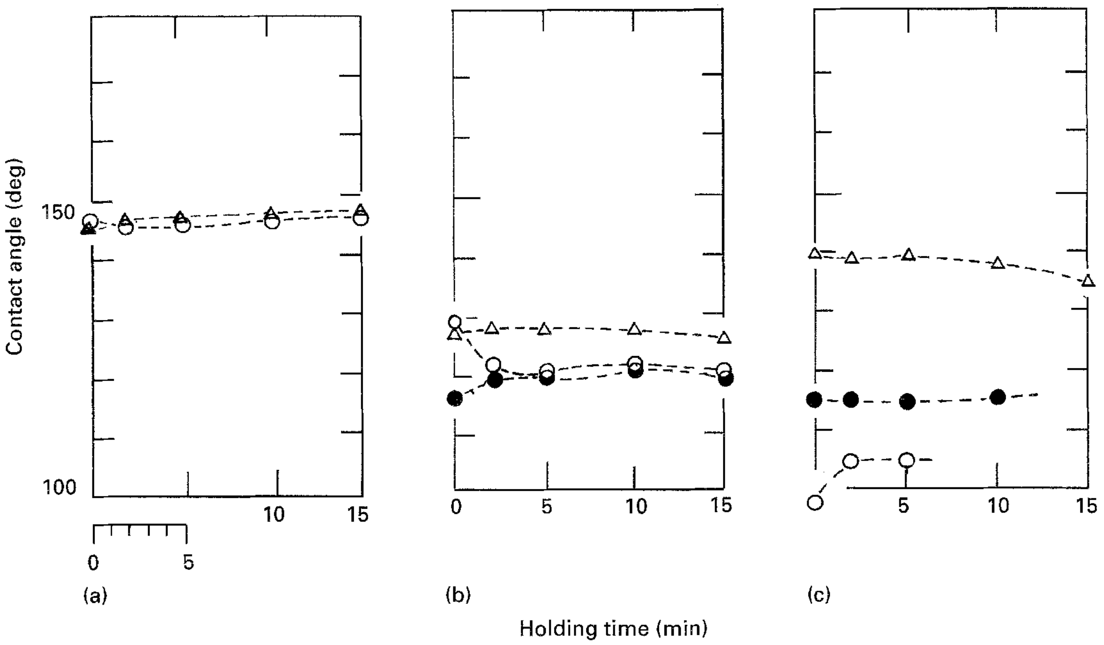

Figure 3 The contact angles of (a) Au, (b) Ag, and (c) Cu to SiC whilst holding at their melting temperature. △ SiC (sintered); ● SiC (1 1 1); ○ SiC (1 0 0 0).

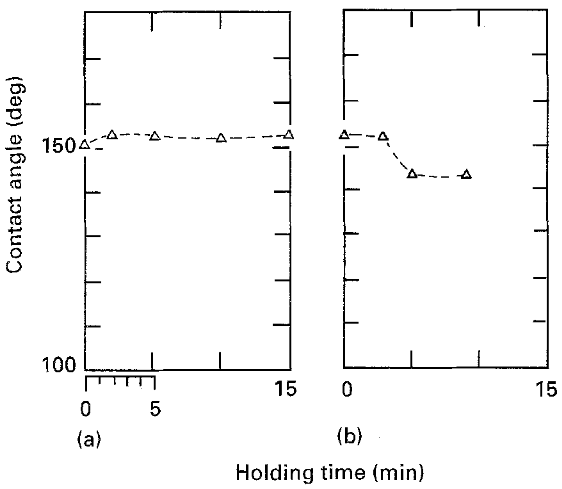

Figure 4 The contact angles of Sn to SiC (sintered) whilst holding at (a) melting temperature (231.9 °C) and (b) 1350 °C.

(1 0 0 0) plates; however adhesion such as reported by Gnyeshin [2] was not observed.

3.2. Contact angle of Sn to SiC

The contact angle of Sn was measured only to a SiC (sintered) plate and the results are shown in Fig. 4 at the melting point of Sn and 1350 °C. The contact angle was above 150° at the melting point and did not change when held at temperature, but it decreased by about 10° after being held for 5 min at 1350 °C.

3.3. Contact angle of Al and Al alloys to SiC

Fig. 5 shows the relation between contact angles of Al, Al–1% Si, Al–6% Si, Al–15% Si–3% Fe to the three kinds of SiC and holding time at their melting temperatures. Each point in Fig. 5 is the average of

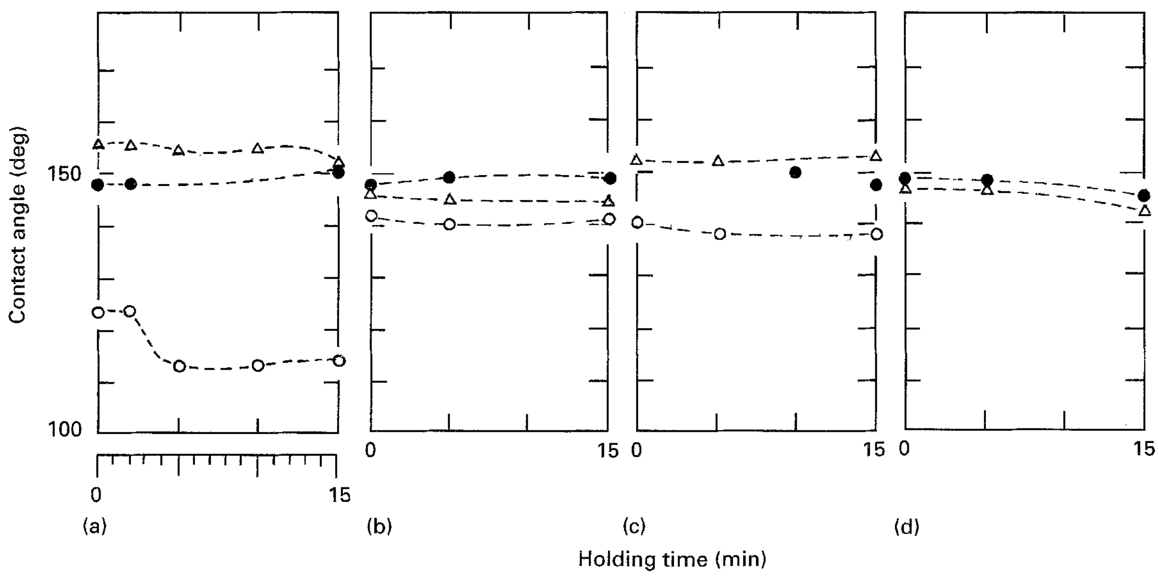

Figure 5 The contact angles of (a) Al, (b) Al–1%Si, (c) Al–6%Si, and (d) Al–15%Si–3%Fe to SiC whilst holding at their melting temperatures. △ SiC (sintered); ● SiC (1 1 1); ○ SiC (1 0 0 0).

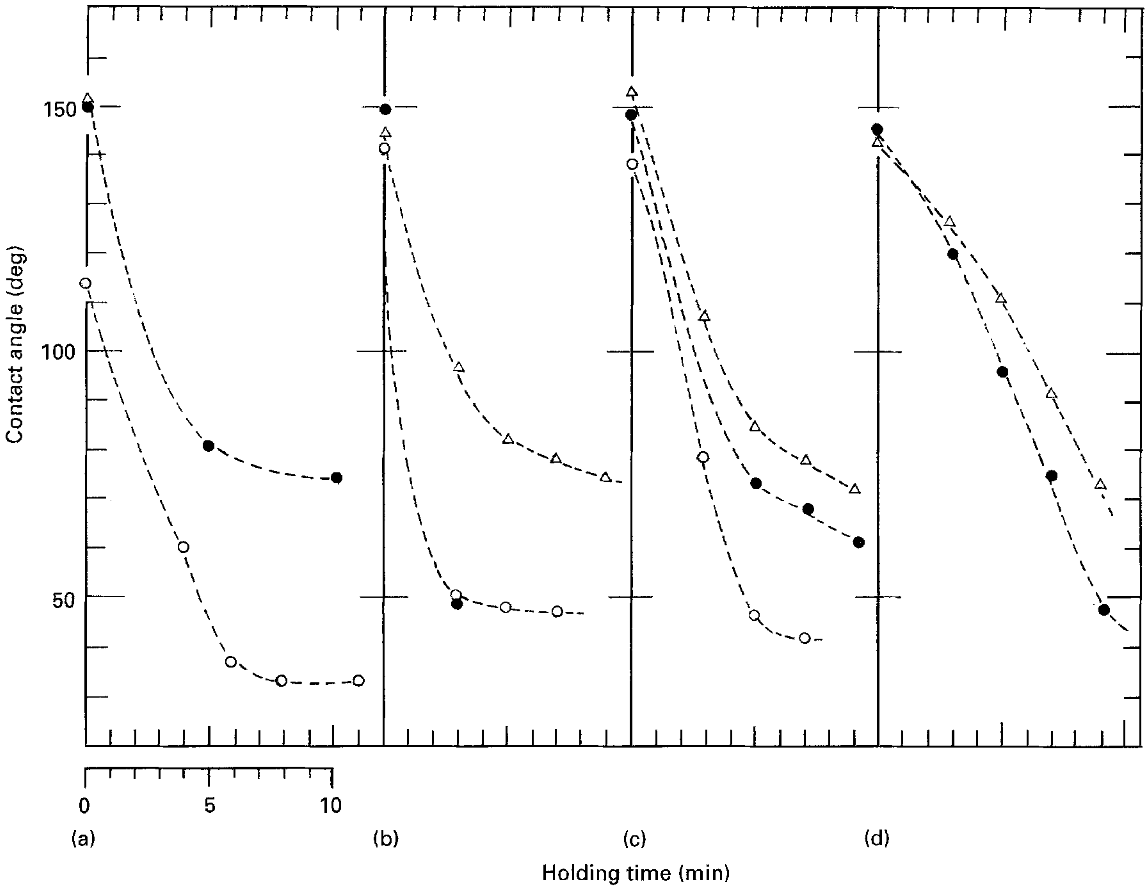

Figure 6 The contact angles of (a) Al, (b) Al–1%Si, (c) Al–6%Si, and (d) Al–15%Si–3%Fe to SiC whilst holding at 1350 °C. △ SiC (sintered); ● SiC (1 1 1); ○ SiC (1 0 0 0).

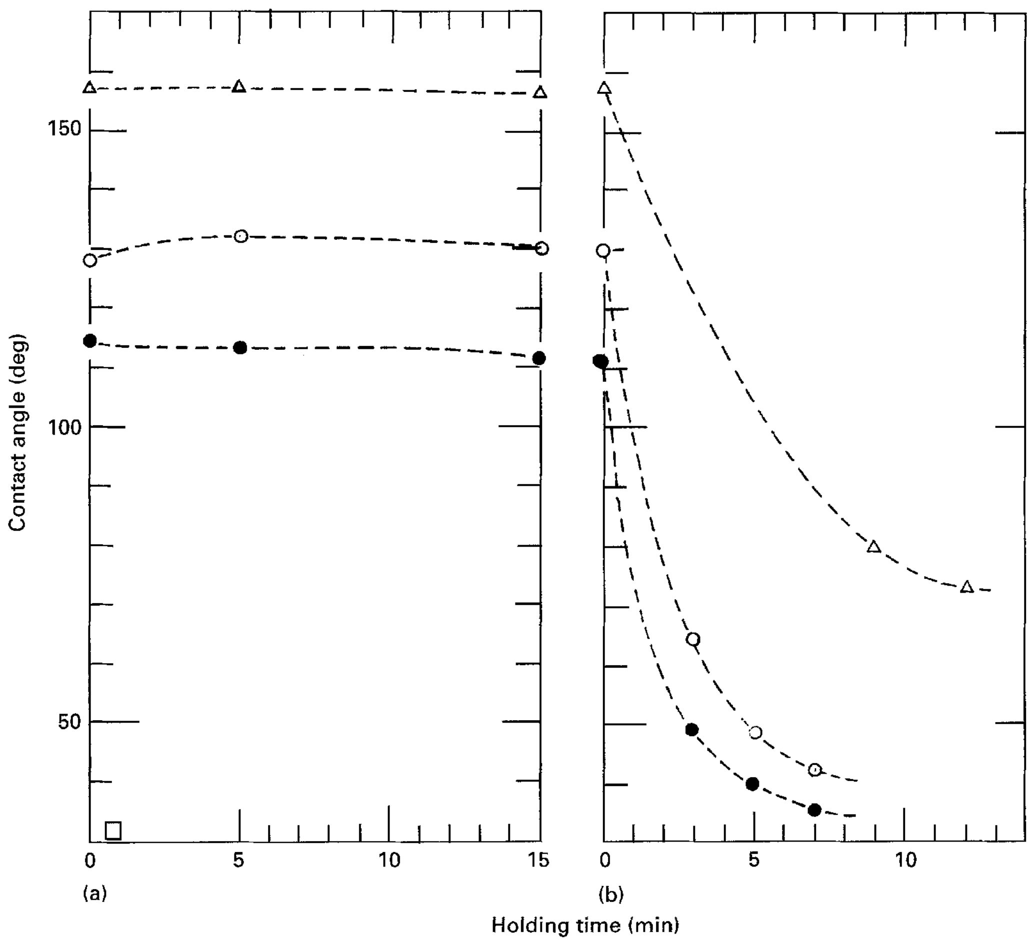

Figure 7 The contact angles of Al–0.46%Si–0.88%Mg alloy to SiC whilst holding at (a) melting temperature and (b) 1350 °C. △ SiC (sintered); ● SiC (1 1 1); ○ SiC (1 0 0 0).

TABLE IV Measured contact angles of various metals and alloys to silicon carbide at the melting points and the reference data [2, 3, 6, 7]

	Au	Ag	Cu	Sn	Al	Al–1%Si	Al–5%Si	Al–20%Si	Al–1.35%Mg_2Si	Au	Ag	Cu	Sn
m.p. (°C) [5]	1063	960.5	1083	231.9	660.1								
SiC (sintered)	147°50′	127°10′	138°	152°40′	154°50′	144°30′	152°	143°	157°	138° to single SiC at 1150 °C [3], about 118° to single α-SiC at 1100 °C [6]	128° at 1100°C [3]	140° to single α-SiC at 1430 °C [2], 165° to hot pressed SiC at 1135 °C [7]	135° at 1050 °C [3], about 140° to single α—SiC at 1024 °C [6]
SiC (1 1 1)		119°50′	114°40′		150°	148°50′	149°20′	145°50′	112°40′				
SiC (1 0 0 0)	146°30′	120°30′	105°		112°20′	140°50′	138°		131°10′				

two measurements, using a new Al granule each time. The contact angle of Al and Al–Si alloys to SiC (1 0 0 0) showed a large difference from those obtained for the two other SiC plates. Al and Al alloys were kept at their melting temperatures for 15 min, then heated to 1350 °C and held. The change in contact angle with holding time at 1350 °C is shown in Fig. 6. Every contact angle decreased remarkably with holding time at 1350 °C, and the contact angle of Al to SiC (1 0 0 0) was smaller than that for Al to SiC (1 1 1), then it tended to a constant value with holding time. This tendency was also observed in Al–1% Si and Al–6% Si alloys but not in Al–15% Si–3% Fe. The SiC with Al and Al alloys could not be mechanically separated at room temperature after cooling from 1350 °C. They seemed to have been strongly connected due to interfacial reaction at the high temperature of 1350 °C.

The time dependence of the contact angle between Al–Mg–Si alloy and SiC whilst held at their melting point and 1350 °C is shown in Fig. 7. The contact angle to SiC (1 0 0 0) was larger than that to SiC (1 1 1), and each of them decreased remarkably with holding time at 1350 °C, then became constant. It can be considered that such a remarkable decrease was related to the fact that carbon, which is a component of silicon carbide, has a little solubility of about 0.11 at % in molten aluminium in the temperature range of 1300–1500 °C. The solubility of carbon in molten metals, except aluminium, is so small that it can be neglected at 1350 °C [4]. In this case, 1350 °C is the temperature corresponding to the liquidus curve at near 90 at % Si in the binary alloys of Si and each of the elements Au, Ag, Cu, Sn and Al [5].

Table IV shows the contact angles of the above described metals and alloys to the three kinds of α-SiC after 10 min holding at their melting temperature and the reference data.

4. Conclusions

1. The contact angles of Au, Ag, Cu, Sn, Al, and Al alloys to the three crystal configurations of α-SiC were measured, respectively, at their melting temperature and 1350 °C under high vacuum.

2. The measured contact angles of Au and Sn to α-SiC were large, about 150°, at their melting points.

3. Most of the metals and alloys used in this study made large angles with SiC (sintered) compared with the other types of SiC.

4. The contact angles of Ag, Cu, and Al to SiC (1 0 0 0) were in the range of 105° – 121° and lower than those obtained for Au and Al alloys.

5. The contact angles of Al and Al alloys to each kind of α-SiC rapidly decreased to under 90° whilst holding at 1350 °C.

6. The contact angle of Al–Mg–Si alloy to SiC (1 1 1) was lower than those obtained for the other two SiC plates at both the melting point and 1350 °C.

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