Wettability of Al_2O_3 by Aluminum and Al-Mg Alloys

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Abstract

The wettablity at a liquid of pure-Al or Al-Mg alloys/ Al_2O_3 interface was evaluated by the sessile drop method at 700°C and with a vacuum pressure of 10^{-4}Pa. The value of contact angle varies (decreases) with time of contact with Al_2O_3 substrate. The reduction in θ of the Al-Mg alloy was proportional to the reduction in surface tension γ_{lv} of Al, and to reactions that took place at the Al-alloy/Al_2O_3 interface.

Keywords: Wettability, Contact angle, Sessile drop method, Surface tension, Aluminum alloys.

التبللية للالومينا من قبل منصهر الالمنيوم وسبيكة Al-Mg

الخلاصة

تم قياس التبللية بين منصهر الالمنيوم وسبيكه الالمنيوم - مغنيسيوم مع سطح الالومينا بطريقة القطرة السائلة الموضوعة على سطح أملس ومستوي عند درجة حرارة 700°C وضغط تفريغ 10^{-4}Pa. قيمة زاوية الترطيب تتغير (تنخفض) مع زمن التماس بين قطرة المعدن المنصهر وسطح الالومينا, وكذلك تنخفض مع أنخفاض قيمة الشد السطحي للمعدن المنصهر, و تنخفض مع حدوث التفاعل عند السطح البيني الفاصل.

Introduction

The Al/α-Al_2O_3 interface is of great scientific and technological significance due to the important role which it plays in such diverse applications as metal-ceramic composites, metal-ceramic joints, casting and smelting processes, microelectronics, corrosion/wear protection, and coating technology. As a consequence, this system has attracted a substantial amount of interest from experimentalists who have largely focused on the measurement and analysis of wetting liquid Al on an α-Al_2O_3 surface via the sessile drop method [1-4].

The study of low temperature wetting of ceramics by liquid aluminum gives rise to a number of difficulties related to the appearance of an oxide layer covering the aluminum and inhibiting the development of the Al/ceramic interface [5-9].

In the fabrication of MMCs the wettability is the most important variable determined by the wetting angle θ of a sessile drop resting on a substrate given by Young-Dupre equation as follows[7];

$$\mathbf{Cos\theta} = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}} \quad \ldots\ldots(1)$$

where γ_{sv} is the surface tension of the solid, γ_{sl} is the solid/liquid interfacial energy and γ_{lv} the surface tension of the liquid.

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A liquid is said to wet a solid when θ is smaller than 90^o, while in non-wetting systems θ is larger than 90^o. The aim of this investagation is to study the influence of addition of an alloying element to aluminum to improve the wettability of Al/Al_2O_3 systems.

Experimental work

Pure Al (99.99%), Al-2.5%Mg, Al-5%Mg alloys were used for sessile drop experiment. Al-alloys were prepared by melting in an atmosphere controlled furnace and cast into steel permanent mold. The sintered Al_2O_3 used in our experiments was supplied by Wacker chemicals company.

The sessile drop apparatus employed, which consists essentially of a tube electrical resistance furnace enabling the sessile drop on the substrate to be illuminated and projected on a camera.

The sample was freshly cut on all of its faces a few minutes before introducing it into the furnace since a very short exposure to air limits the thickness of the oxide coating to the polished substrate. A schematic of the wetting equipment is given in figure(1) and figure(2); basically it consists of: Outer Mulite tube was located between the heat shield cover and the wire resistance element, the tube has outer diameter of 50mm and internal diameter of 30mm, 270mm in length.

Internal mulite tube was located inside the loop of wire resistance element, and contained Al_2O_3 substrate. The tube has outer diameter of 25 mm and internal diameter of 20mm, 275mm in length.

The Al_2O_3 material was slit to 12×12×5mm slices for wettability studies. One flat face was grinded by 240, 400,500, 800, and 1200 grade abrasive papers followed by polishing sequentially with 3, 1 and 1/4μm diamond paste.

An alloy piece approximately 0.7g in weight was cut from ingot materials; each sample was ground to a cubic shape and immediately immersed into dry methanol to minimize the formation of oxide film on the surface and then ultrasonically cleaned.

The experiments were carried out under vacuum of 1×10^{-4} Pa with pure nitrogen gas, evacuation was carried out by a rotary pump and diffusion pump.

A camera unit was set up in the front internal tube to record the image of the metallic drop. Photographs were taken at 0, 15, 30, 45 minute intervals after the temperature of the sessile drop reached 700^oC.

Results and Discussion

The wetting experiments show that contact angle between liquid metal of Al and Al-Mg with Al_2O_3 substrate decreases with time, due to the dissociation vaporization and decrease thickness of an aluminum oxide layer under vacuum with time. Table (1), fig(3) and fig(4) show that the wetting angle(θ_o) for pure Al is (150^o) at the beginning and decreases with time until it becomes (120^o) after (45) minutes (θ_{45}).Magnesium is an element that can reduce the aluminum oxide film, which will allow the melt to come into contact with alumina and give better wetting [10].

$$Al_2O_3(s) + 3Mg(l) \longrightarrow 3MgO(s) + 2Al(l) - 32.6\ kCal\ at\ 1000K$$

The negative sign of ΔH_f^{Φ} means that spontaneous formation is thermodynamically possible [10]. The surface of the alumina therefore can be reduced by magnesium at 1000K to form MgO and Al(l).

The driving force for wetting is affected by only two factors; surface tension of liquid γ_{lv} , and the chemical reaction between solid and liquid at the interface, which leads to lower interface tension γ_{ls} .

Surface tension of liquid γ_{lv} of Mg is (0.560 Nm^{-1}) as compared with that of pure-Al (0.866 Nm^{-1}). Addition of 3% of Mg to Al melt reduces its surface tension to (0.62 Nm^{-1}) at 993K^o, a reduction of about 30% from its original value [5]. Magnesium have high vapor pressures and are relatively volatile. The vapor pressure for Mg is approximately 1atm at 1100^{o}C the effect of the high vapor pressure of Mg in the spontaneous infiltration in Al-Al_2O_3 system is that it can be spread over the ceramic particles in MMCs, thus suppressing thick oxide scale formation and promoting wetting [5]. It is well known that Al_2O_3 readily reacts with many divalent transition metal oxides to form aluminates which are isostructural with spinel of composition $MgAl_2O_4$ mineral. Spinel or similar oxides promote interfacial bonding since they form strong bonds metals and ceramic. A large thermodynamic driving force exists for spinel formation ($MgOAl_2O_3$) which increases with increasing Mg content [2].

The reason for the spread of data can be found in the different thicknesses of the oxide layers, formed at different partial pressures of oxygen, as shown in table(2) and fig(5). However no sessile drop can formed at the presence of air (without vacuum)

Conclusions

The most effective addition will be elements which react with the substrate added in quantities sufficient to form an atomic monolayer of reaction product. Wetting angles decreased exponentially with time for Al-Alloy/Al_2O_3 systems.

References

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Table (1) Values of contact angle with time and composition.

Time (min)	*θ(deg) Pure Al*	*θ(deg) Al-2.5%Mg*	*θ(deg) Al-5%Mg*
0	**150**	**127**	**120**
15	**125**	**104**	**98**
30	**121.5**	**102**	**96**
45	**120**	**100**	**94.5**

Table (2) the contact angle with different temperature and pressure obtained by many researchers.

Researcher	Pressure (pa)	θ (deg) at 700°C	θ(deg) at 1000°C
Walf [2]	**1.33×10^{-2}**	**167**	**98**
Pask [2]	**3.8×10^{-3}**	**120**	**88**
Kohler [2]	**3×10^{-5}**	**164**	**72**
Hausner [2]	**4×10^{-5}**	**123**	**88**
V-Laureant [2]	**4×10^{-5}**	**101**	**86**
This work	**1×10^{-4}**	**125**	**-**

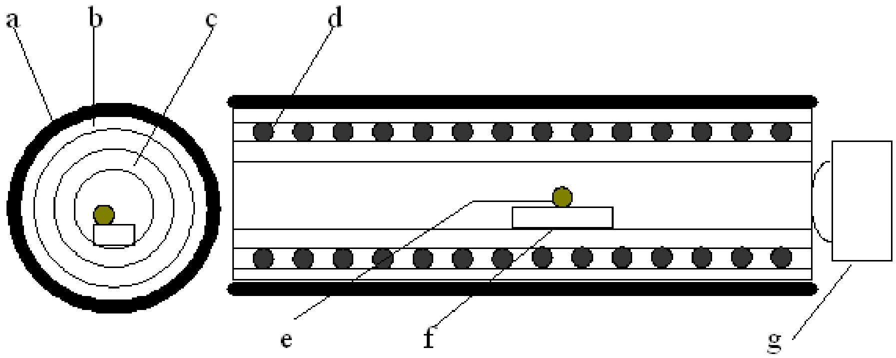

Figure (1) Schematic of sessile drop apparatus.
a-heat shield cover, b-outer mulite tube, c-internal mulite tube, d-Nichrome wire heating element, e-sessile drop, f-Alumina substrata, and g-Camera.

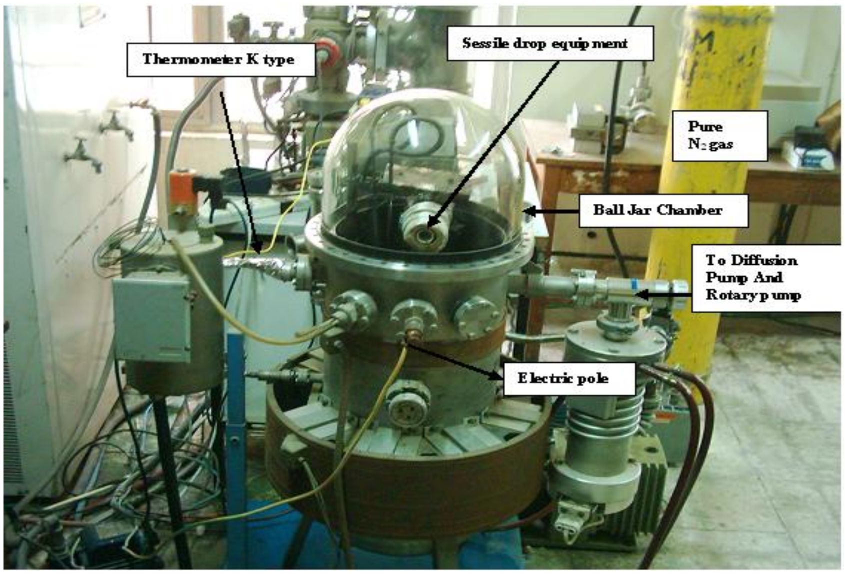

Figure (2) Equipment used for measurement contact angle by sessile drop method, Department of applied Sciences laboratory.

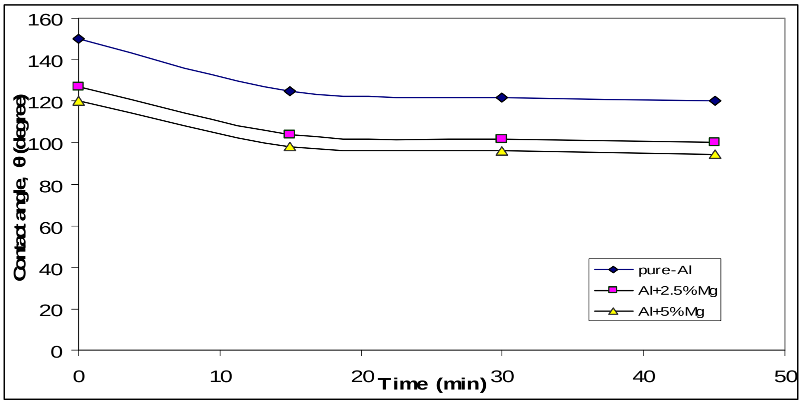

Figure (3) Contact angle versus time for Al-alloy/Al_2O_3 system at 1×10^{-4} Pa and 700°C.

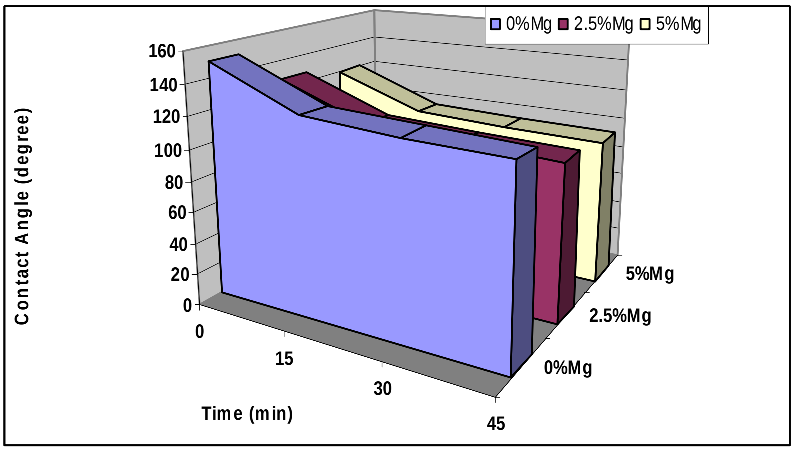

Figure (4) contact angle versus time and Mg% For Al-alloy/ Al_2O_3 system at 1×10^{-4} Pa and 700°C

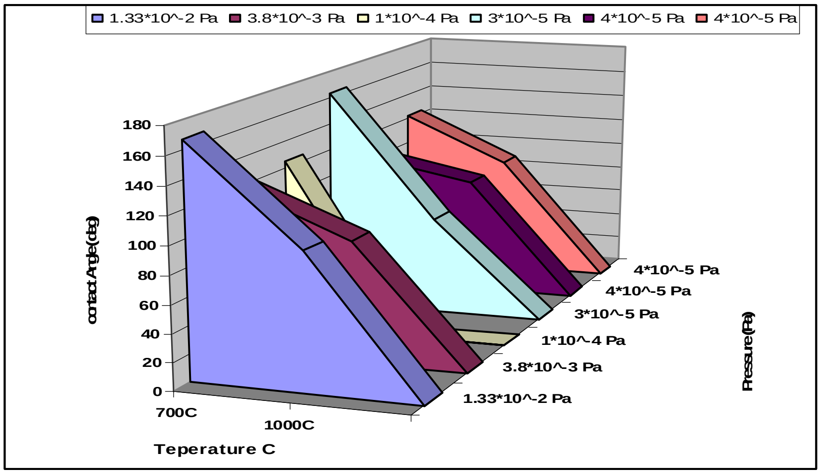

Figure (5) The contact angle for different research profile with temperatures and pressures.