

Studying the wetting of carbon-coated alumina substrates by different aluminum alloys

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The wetting of alumina ceramics was studied as a function of time and temperature by the sessile drop method using vacuum pressures of 10^{-5} Pa. Contact angles as small as 38 degrees were obtained for the combination carbon coated alumina and an Al-2.5 at.% La alloy, in a time of 4500s and at a temperature of 1300 °C. On the other hand, for the single system alumina-aluminum the final contact angle was of 82 degrees after 7200 seconds at the same temperature. Considering the free formation energy of different oxides and carbides that can be formed between elements used in the present experiments are favorable thermodynamically and considering the observation of undercutting of the substrate and ridge formation at the leading edge of the liquid aluminum in all samples it can be concluded that wetting is enhanced thanks to different chemical reactions that occur at the ceramic-metal interface.

Key words: Wetting, Carbon coated, Alumina substrates, Aluminum alloys.

Introduction

During recent years different studies about the wetting of Al_2O_3 by different metals have been made. In some of these studies it was established that the wetting of ceramics by liquid metals is due to different chemical reactions that occur at the ceramic-liquid interface between the liquid phase and the solid phase [1-9]. However, in spite of the research in this field collectively called reactive wetting the mechanisms that control the different wetting stages remain without agreement between scientific communities. As example, it was reported that the contact angle can vary from 130 degrees to 30 degrees depending of the study [10-11]. This can be attributed to the different factors that affect wetting during the experiments such as: the system components, impurities in the metals and substrates, roughness and defects in the substrate, oxygen partial pressure, etc. The Al- Al_2O_3 system is considered as a non-reactive system, because Al cannot wet Al_2O_3 by itself. However, if the Al_2O_3 is carbon coated or if the aluminum is alloyed with some reactive metal such as: Ti, Zr, Mg, etc, the system becomes reactive and different contact angles can be observed depending of the progress on chemical reactions at the ceramic-metal interface [10, 12].

The aim of this work is to analyze the possible mechanisms that affect the wetting of carbon-coated Al_2O_3 substrates by different aluminum alloys.

Experimental

Three different aluminum alloys were prepared by melting at 800 °C and 0.7×10^{-4} Pa in a vacuum induction furnace (Brew, 424-C, USA). The compositions of the samples were 100% Al (Alfa Aesar, USA, 99 wt% Al), Al-2.5 at.% La (Plasma Powders Co., LTD, China, 99.5 wt% La) and Al-3 at.% Y (Plasma Powders Co., LTD, China, 99.9 wt% Y). Samples codes are given in Table 1. The substrates were dense polycrystalline alumina (Coors Tek Substrate, USA, 99.6% Al_2O_3) and were diamond cut n square slices of 0.064 cm thickness by 1 cm side. Carbon films were deposited onto the substrates by a vapor deposition process using a sample coating device, each deposition event took 45 s. Small cubes weighing ~0.3 g were diamond cut from aluminum alloy bars. Sessile drop experiments were conducted using an induction heating generator (Lepel High Frequency Lab. Inc. model T-30-3-DF-TL, USA). The experiments were conducted in a stainless steel chamber under a controlled vacuum (10^{-4} to 10^{-5} Pa), at temperatures up to 1300 °C. The droplet profiles were recorded in-situ, as a function of time, using a video camera (Canon L2, Japan). The liquid metal

Table 1. Identification of the samples used in the sessile drop experiments

Sample	System	Carbon coated- Al_2O_3
Al	Al- Al_2O_3	None
AC3	Al- Al_2O_3	3 carbon deposition events
ACL	Al-2.5 at% La- Al_2O_3	3 carbon deposition events
ACY	Al-3 at% Y- Al_2O_3	3 carbon deposition events

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temperature was controlled with an infrared camera (Ircon, Inc. model MR-OR10-19C). Both the video camera and the infrared camera were sighted through quartz windows of the chamber. Before the wetting experiments the aluminum alloys were cleaning for 40 minutes in acetone by agitating in an ultrasonic bath. Contact angle measurements were made directly from images obtained from the video display using a digital camera (Olympus 350, Japan). The samples from the wetting experiments were analyzed by scanning electron microscope (Hitachi, S-4700, Japan) and energy dispersive spectroscopy (NORAN 457A-3SES, USA). The aluminum was removed from the substrates to reveal the interface, by etching in a distilled water-20% NaOH solution.

Results and Discussion

Effect of temperature

The contact angles (θ) obtained after 4500 s hold time are shown in Fig. 1 as a function of temperature for each of the samples. For each sample, it was found that the contact angle decreased with increasing temperature. However, wetting of substrates by aluminum only occurred at temperatures at or above 1200 °C, where a required contact angle of less than 90° (e.g., equation 1) was obtained. At 1300 °C, the lowest q values were found for the samples where the q values for samples A1, AC3, ACL and ACY are equal to 82, 80, 38 and 55 degrees respectively. These results indicate that the deposited carbon film on the substrate together with the alloying elements have an important effect in enhancing the wetting of the alumina substrate by the molten aluminum.

$$(\gamma_{l-v}) \cos \theta = \gamma_{s-v} - \gamma_{s-l} \quad (1)$$

Where: θ is the contact angle, γ_{s-v} , γ_{s-l} and γ_{l-v} surface energies at the solid-vapor, solid-liquid and liquid-vapor interfaces respectively.

Effect of time

Figure 2 shows the contact angle variation as a function of holding time of samples at 1300 °C. For all

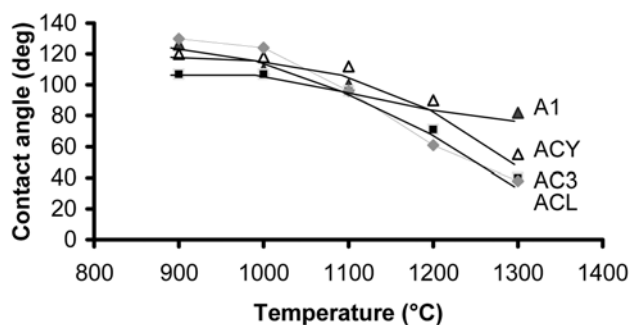


Fig. 1. Effect of temperature on the contact angle. Holding time 4500 s.

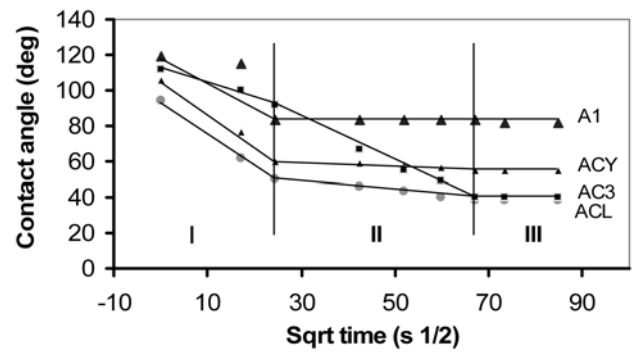
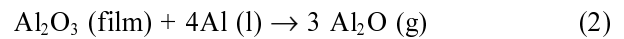


Fig. 2. Contact angle variation as a function of holding time of samples at 1300 °C.

samples the initial contact angle varies between 100 and 120°. However, in approximately 600 s the value of θ diminishes quickly. Laurent et al. [13] indicated that the large θ values measured here are typical of oxide metallic drops, therefore the reduction of θ is controlled by the drop deoxidizing that is achieved principally by the Al_2O_3 film making with the liquid aluminum with the formation of gaseous Al_2O in accordance with:



The contact angle measured in sample A1 after holding for 600 s at the indicated temperature agrees very well with results reported in the literature where the experiments with ceramic-metal systems avoided the influence of the oxide film on the wetting [3, 13].

For samples AC3, ACY and ACL Figure 2 shows the existent of three wetting stages, in this way the wetting kinetics can be characterized by the contact angle value at the beginning of each stage.

In the first stage, the contact angle diminishes from $\theta_0=112^\circ$ to $\theta_1=92^\circ$, from $\theta_0=113^\circ$ to $\theta_1=80^\circ$ and from $\theta_0=100^\circ$ a $\theta_1=78^\circ$ for samples AC3, ACY and ACL respectively. This first stage in these samples is very similar to the kinetics of sample A1. Consequently, it can be assumed that the control mechanisms during the first stage in samples AC3, ACY and ACL is the deoxidizing of the metallic drop given by equation 2. At 600 s there is a change in the slope of the curves, significantly slowing the kinetics of wetting of the substrate. Stage 2 occurs between 600 and 4500 s, the wetting during this stage is characterized by an almost lineal kinetic dependency with time. Finally, during stage 3, which occurs for times longer than 4500 s, the contact angle remain without major changes.

In agreement with different authors [1, 7, 8] the formation of carbides or oxides at the ceramic-metal interface between different elements of the systems help to diminish the interfacial energy in the system, in this way the work of adhesion between the liquid metal and the ceramic substrate will be substantial, consequently the wetting in the system is enhanced. In

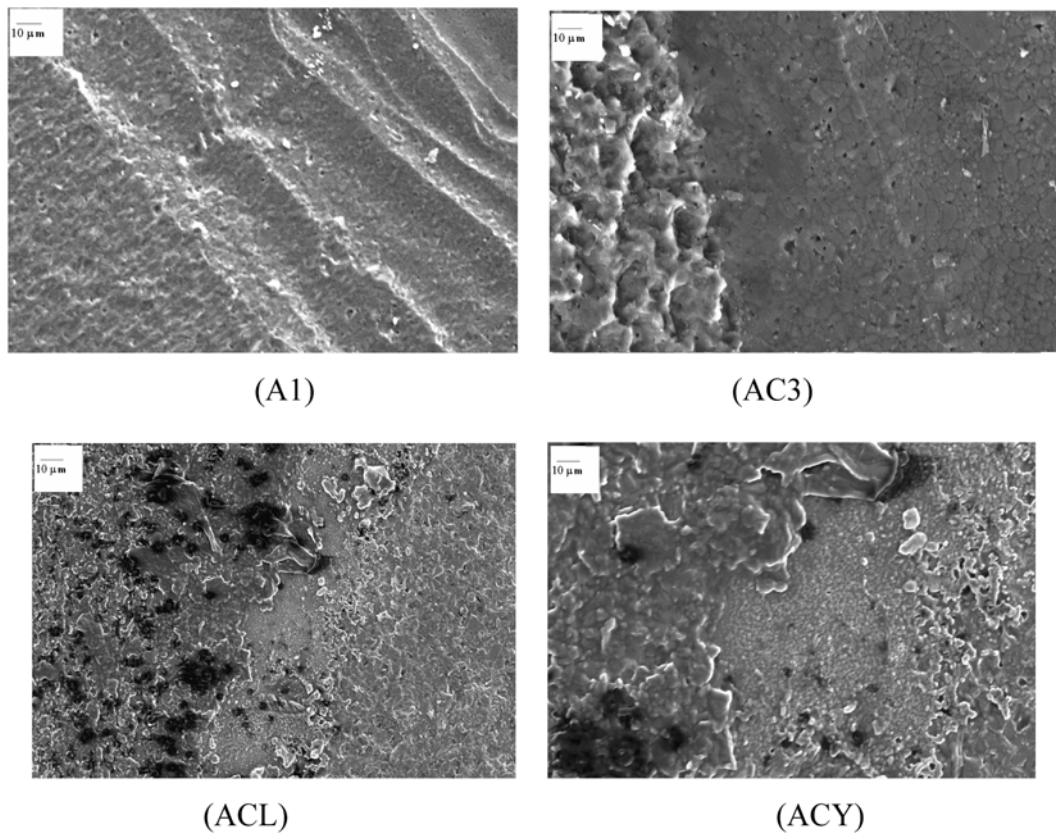
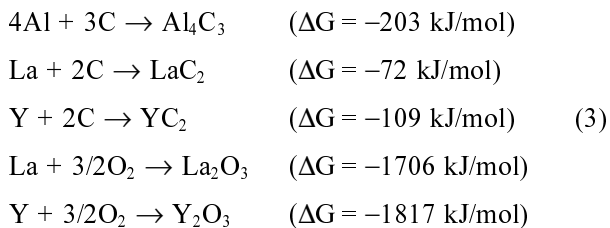


Figure 3. SEM images of the ceramic-metal interface. Holding time 4500 s at 1300 °C.

accord with the free energies of formation of different carbides and oxides one of the next reactions can take place in the present experiments during the second stage.



Therefore, it can be commented that the presence of carbon together with the alloying elements such as; yttrium (Y) and lanthanum (La) promotes the wetting of the substrate by the metal. However, it is important to comment that no chemical analysis was carried out in order to verify the presence of any of the products suggested by the set of equations (3).

Ceramic-metal interface

After samples were cooled; the aluminum drop was removed from the substrate by etching in a sodium hydroxide solution. Then the surfaces of the different substrates were observed with a scanning electron microscope (SEM). The observations in Fig. 3 of

undercutting of the substrate and ridge formation at the leading edge of the liquid aluminum in all samples shows that wetting is enhanced thanks to the different chemical reactions that occur at the ceramic-metal interface. The final contact angle for sample A1 was 82 degrees for experimental conditions of 4500 s at 1300 °C, whereas, the final contact angles for samples ACY, AC3 and ACL were 55, 40 and 38 degrees respectively. Detailed analysis at the ceramic-metal interface by energy dispersive spectroscopy (EDS) did not reveal any reaction product. However, zones rich in Y and La were detected in the corresponding systems. As was observed in Figures 1 and 2 the single combination of Al-Al₂O₃ means a non-reactive system with no significant changes in the wetting of the substrate. However, the addition of Y or La as alloying elements to aluminum together with the use of carbon-coated Al₂O₃ substrates reduced in a significant way the final contact angle of the metal on the ceramic. This contact angle reduction results from at least one of the surface energies given in equation 1. Therefore, it can be assumed that the segregation of alloying elements to the edge of the liquid aluminum with the substrate reduces the interfacial Gibbs energies due to an excess of the alloying element or to a chemical reaction at the interface and this is thus associated with the enhancement of the contact angle.

Conclusions

- At temperatures $> 1200\text{ }^{\circ}\text{C}$ there is wetting in the systems studied here.
- Holding time increments at $1300\text{ }^{\circ}\text{C}$ reduce the contact angle.
- The use of carbon-coated Al_2O_3 substrates together with alloying with Y or La of aluminum improves the wetting of the ceramic by the metal.
- Due to the observations of undercutting of the substrate and ridge formation at the leading edge of the liquid aluminum in all samples it is established that wetting is enhanced thanks to different chemical reactions that occur at the ceramic-metal interface.

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