



# Wettability of zirconium-coated alumina by molten aluminum



Qian-Gang Xu <sup>\*</sup>, Li-Wei Guo, Li Zhang, Hong Liu

Department of Materials science and Engineering, Shenyang Aerospace University, Shenyang 110136, China

## ARTICLE INFO

### Article history:

Received 25 January 2016

Revised 3 May 2016

Accepted in revised form 23 May 2016

Available online 27 May 2016

### Keywords:

Wettability

Alumina

Zirconium

Interfacial interaction

Precursor film

Thin film

## ABSTRACT

The wetting behavior and interfacial phenomena of alumina, Zr-coated alumina, and bulk Zr with liquid aluminum at 700 °C was investigated by the sessile drop technique. Zirconium film deposited on alumina improved the wettability with aluminum and promoted the formation of the precursor film. With the increase of zirconium film thickness from 160 nm to 670 nm, the terminal contact angle decreases gradually and spreading rate increased and the precursor film became wider and thicker. Marangoni flow resulting from the surface tension gradient due to the interfacial interaction is a major factor resulting in the formation of the precursor film.

© 2016 Elsevier B.V. All rights reserved.

## 1. Introduction

Wettability of solid by liquid metal is important for the fabrication of metal-matrix composites by liquid routes and the joining of metals and particularly of ceramics by brazing, because wettability at high temperature has a significant influence on the properties of the final product. Coating of ceramic is one of the successful techniques adopted to enhance the interfacial bonds and improve the wettability between ceramic and liquid metals. Various coating techniques such as chemical vapour deposition (CVD), physical vapour deposition (PVD), thermal spraying, sol-gel process, the formation of in-situ coatings, electrolytic and electroless plating methods are applied to deposit coatings (such as Ni, Cu, Ti, Mo, TiC etc.) on the surface of ceramic to promote wettability [1–5]. The interaction and the variation of wettability between molten metals and ceramic depend on the properties of the phases in contact, the structure and the thickness of the deposited metallic coatings. Investigations on the wettability and interfacial interaction between molten metals and metal-coated ceramic form a basis for the design of interlayer coatings and development of methods of deposition of metals on ceramic materials.

The amount of published articles on wetting of metal-coated substrate by liquid metals is far less than the wide range of practical application of metal-coated nonmetallic materials in the fabrication of metal-matrix composites by liquid routes and the brazing of ceramics [3,6–12]. However, wetting behaviors become more complex when a coating is used. Wettability studies of liquid metals on coated substrates are in short supply compared to demand because of the many possible

variables on wettability (e.g., substrate, coating, and molten-metal compositions, which can range from simple [pure elements] to complex alloys; surface roughnesses; temperature dependence; atmospheric dependence [e.g., O<sub>2</sub> partial pressure]; path to molten state, as slower heating can allow more time for reaction with the substrate and/or coating, possibly producing different initial or boundary conditions [compositions] or the complete alloying of diffusion of what would have been the molten metal; effect of gravity, etc.). In general, the limitations come up when specific combinations of compositions and/or temperatures are desired or required.

The addition of Zirconium to the metal melt may promote wetting of ceramics. However, wettability study of Zr-coated ceramics by molten metal is scarce. In this study, sputter deposited zirconium thin films were used to improve the wettability of alumina by molten aluminum. The influence of film thickness on the wetting, interfacial interaction and the precursor film in the Al/Zr-coated  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> system was investigated.

## 2. Experimental procedure

Wetting studies were carried out on alumina, Zr-coated alumina, and Zr. Cubic pieces of 120 ± 10 mg of high purity aluminum (99.99 wt.%) were used. Pure Zr sheets (99.5 wt.%) of 1.5 mm in thickness were used as the wetting substrate and the sputtering target. Zr films were deposited on high purity polycrystalline  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (99.9 wt.%) substrates by RF magnetron sputtering of a 50-mm-diameter Zr target at room temperature. The substrates rotated at a speed of 10 rpm. The target to substrate distance was kept constant at 65 mm. Base pressure in the process chamber was 5 × 10<sup>-4</sup> Pa and the working gas was pure argon (purity 99.99%) at 0.5 Pa in a continuous flow

<sup>\*</sup> Corresponding author.

E-mail address: [xuqiangang@126.com](mailto:xuqiangang@126.com) (Q.-G. Xu).

(15 sccm [Standard-state Cubic Centimeter per Minute]). Zr thin films in various thicknesses (about 70 nm, 160 nm, 350 nm, 550 nm and 670 nm) were deposited on the alumina substrate. Fig. 1 shows the surface micrograph of Zr thin film deposited on alumina by RF magnetron sputtering. It can be seen that the Zr film showed nanocrystalline morphology with few voids. Each substrate was polished using 1.0  $\mu\text{m}$  diamond paste before sputtering or wetting experiment. Before use, substrates and pieces of Al were cleaned ultrasonically in anhydrous alcohol and acetone.

Wetting examinations were performed using a sessile drop technique at 700 °C under high vacuum of  $1.6 \times 10^{-3}$  Pa. The apparatus essentially consisted of a molybdenum resistance furnace fitted with two windows, enabling the illumination of the sessile drop on the substrate and the projection of its image on a screen. The contact angles were measured directly from the image of the drop section. After the experiments, the solidified samples were analyzed using scanning electron microscope equipped with energy dispersive spectrometers (EDS).

### 3. Results

Fig. 2 shows the variation in apparent contact angle with time during the isothermal wetting of molten aluminum on alumina, Zr-coated alumina, and Zr at 700 °C. Non-wetting behavior is evident in Al/Al<sub>2</sub>O<sub>3</sub> system. The terminal contact angle after 1 h holding was about 137°. The contact angle of aluminum on alumina is sensitive to O<sub>2</sub> partial pressure and surface rough, hence much of the variability in the reported literature (ranges from about 90 to 167° at 700 °C [13]). This indicated that the surface of molten aluminum was oxidized to a certain extent in this experiment. Compared with alumina, bulk zirconium showed excellent wettability with molten aluminum. The contact angle decreases rapidly in the initial several minutes and then quickly approaches a very low steady value (about 8°). Zirconium thin films applied to the alumina plates improved wettability with molten aluminum. The wettability of molten aluminum on the 70 nm thick Zr film is still poor and the terminal contact angle is 118°. A sharp transition from non-wetting to wetting appears in the film thickness from 70 nm to 160 nm. With further increase in the thickness of a zirconium film on alumina, the incubation time before spreading, the time needed to reach the steady value and the terminal contact angle decrease gradually. The terminal contact angles of aluminum on zirconium film with thickness of 160 nm, 350 nm, 550 nm and 670 nm are 35, 31, 29 and 22°, respectively, which progressively approaches the terminal contact angle of molten aluminum on bulk zirconium.

Fig. 3 shows the top-view macrographs of aluminum on Zr-coated alumina and bulk zirconium substrate after wetting at 700 °C for 1 h. Clearly, a precursor film, which looks like a halo with the naked eye, detaches from the bulk drop on the surface of Zr-coated alumina and bulk zirconium substrate. With the increase of the thickness of zirconium film, the precursor film significantly widened and a bright white narrow

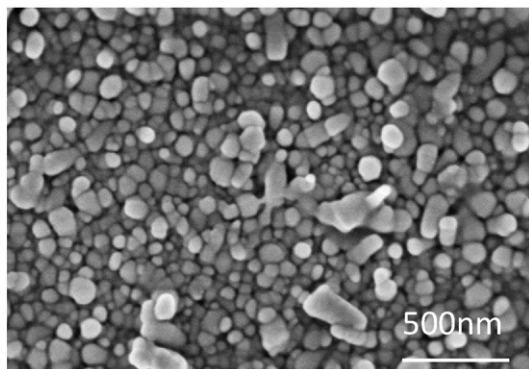


Fig. 1. Surface micrograph of Zr thin film deposited on alumina by RF magnetron sputtering.

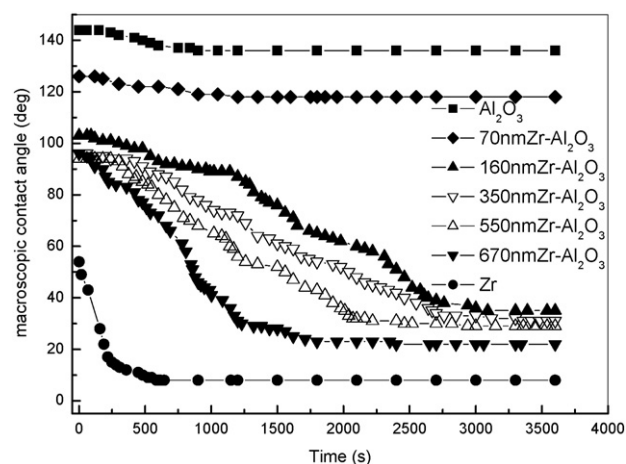


Fig. 2. Variation in macroscopic contact angle with time during the isothermal wetting of molten aluminum on zirconium, plain and zirconium coated alumina substrates at 700 °C.

ring forms in the halo frontier (Fig. 3(b) and (c)). Although the spreading area of aluminum on bulk zirconium is larger than that on Zr-coated alumina, only a narrow precursor film forms in the aluminum-bulk zirconium system. Pronounced cracking occurs at the center of solidified droplet on bulk zirconium.

Fig. 4 shows the surface micrographs of the solidified precursor film on Zr-coated alumina with Zr film thickness of 550 nm at 700 °C for 1 h. A concentric ring forms in the solidified precursor film, one wide inner ring in front of the droplet and one narrow outer ring in the front of the precursor film (Fig. 4(a)). In the wide inner ring, there are some fine Zr-rich phases dispersed in aluminum and the concentration of Zr-rich phase occurrence increases with the distance from the droplet. There are a small amount of Zr-rich phases with larger size uncovered by aluminum in the narrow outer ring (Fig. 4(c)). The outer edge of the precursor film is not uniform but spreads preferentially along the boundaries of Zr-rich phases (Fig. 4(d)). EDS analysis results of the surface of solidified wetting sample is shown in Fig. 5 and Table 1. The zirconium concentration at the surface of droplet is very limited and only about 0.03 at.%, which is smaller than the solubility of zirconium in molten aluminum at 700 °C (about 0.08 at.%) [14]. Zirconium concentration increases gradually from the droplet frontier (0.42 at.%) to the outer edge of the inner ring (about 8.27 at.% at position 5 in Fig. 5) and then has a very slight decrease in the outer ring (about 7.93 at.% at position 6 in Fig. 5) of the precursor film. Owing to the very limited solubility of zirconium in aluminum at 700 °C, the high zirconium content indicates that zirconium at the surface of precursor film exists in two forms, Al-Zr solution and Al-Zr compound. EDS analysis indicates that the composition of Zr-rich phase (about 77 at.% Al, 23 at.% Zr) at the surface of precursor film is close to ZrAl<sub>3</sub>. Clearly, there is a positive concentration gradient of zirconium in the sample surface from the droplet to the precursor film, and correspondingly the content of ZrAl<sub>3</sub> phases at the surface of precursor film increases gradually. Sample surface contains a certain amount of oxygen, which indicated that the surface is oxidized to some extent, especially in the areas with high zirconium content.

The cross-sectional microstructures and the corresponding EDS analysis for the map of elements of the wetting sample with zirconium film thickness of 550 nm are shown in Fig. 6. Obviously, a discontinuous reaction layer appears at the interface from the droplet adjacent to the triple line to the front edge of the precursor film and the reaction product increases progressively along the spreading direction. But there is no discernible reaction product at the interface far behind the triple junction (Fig. 6(c)). Fig. 6(d) and Fig. 6(e) are cross-sectional images of the precursor film at the spreading frontier and adjacent to the droplet, respectively. The precursor film is thicker than the interfacial reaction layer and reduces in thickness toward the spreading frontier (Fig. 6(d)

Download English Version:

<https://daneshyari.com/en/article/8025198>

Download Persian Version:

<https://daneshyari.com/article/8025198>

[Daneshyari.com](https://daneshyari.com)