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Effects of coating thickness on thermal conductivities of alumina coatings and alumina/aluminum hybrid materials prepared using plasma electrolytic oxidation

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ABSTRACT

The aim of this paper was to investigate the effects of coating thickness on the thermal conductivities of alumina coatings and alumina-coated aluminum substrates as hybrid materials prepared using plasma electrolytic oxidation (PEO). The coating thermal conductivities were measured with the comparative guarded heat flow method (also called cut-bar method). The guarded insulation system adopted in this method can effectively reduce the radial heat exchange and the axial shunting exchange, thus improving the precise of the measurement. The experimental results show that with the coating thickness increasing from 10 μ m to 100 μ m, the thermal conductivity has a slight rise from 1.9 W/m·K to 2.5 W/m·K. The gradient structure of the coating in grain size was believed to be one of the possible reasons to cause this slight variation. For the alumina-coated substrates (i.e., alumina/aluminum hybrid materials) with a determined substrate thickness, the increase in the coating thickness has more influence on the equivalent thermal conductivity of the hybrid materials than that in the effective thermal conductivity of the coating themselves.

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1. Introduction

Alumina is frequently used as a coating material due to its good tribological, chemical, dielectric and optical, and biomedical properties [1]. The deposition of alumina on metal materials, such as aluminum and its alloys, can largely improve their resistances to frictional wear, corrosion, oxidation and high temperature attack, etc. In recent years, the coating deposition technique of plasma electrolytic oxidation (PEO) has attracted lots of researchers due to its superior coating performance. It is regarded as one potentially ideal technique to fabricate alumina coatings on relatively low-melting-point aluminum alloys due to low bulk material temperatures (below 100 °C) in the deposition process [2]. Also, the technique can produce thick, high hardness and largely crystalline oxide coatings and cause no environment hazard from electrolytic solutions.

Currently, the research on PEO oxide coatings is mostly focused on beneficial properties in friction, wear, corrosion and oxidation [2–5]. However, for the property related to thermal conductivity of oxide coatings, only a few papers are published in the open literature. Curran and Clyne [6] and Tan et al. [7] measured the thermal conductivity of thick alumina coatings (100 μ m thick) by the steady-state heat flow method and derived the value of 1.6 W/m·K, which was about an order of magnitude lower than typical values reported for single crystal Al₂O₃ (32–34 W/ m·K) [8]. The researchers [9] primarily contributed this low value to the presence of a high proportion of amorphous material together with a fine grain size. It was concluded that porosity plays no significant role in determining the thermal conductivity. Further, Curran et al. [10] used silicate-rich electrolytes to grow mullite-rich oxide coatings on aluminum alloys with an even lower value (0.5 W/m·K).

In this study, alumina coatings with different coating thicknesses were prepared and their conductivities were measured. The motivation to study effects of different thicknesses results from the fact that the increase in coating thickness will vary microstructures (e.g. the variations of porosity and gain size) which would influence thermal conductivity.

Since the PEO coatings generally have excellent adhesion to the substrates, it is almost impossible to get undamaged freestanding alumina coatings by physical detachment methods. Although wet chemical etching can do it, yet the etching process of removing substrates may to some extent alter the thermal conductivity property of such coatings. Therefore, in contrast with the commonly used techniques for thin film measurement such as 3ω method [11], laser flash method [12], time-domain thermoreflectance [13] and frequencydomain-based methods (photothermal reflectance method, thermal emission method, photothermal displacement, photothermal deflection (mirage method) and photoacoustic method [14–18]), the comparative guarded heat flow method (also called cut-bar method) is more suited for such porous non-freestanding samples, although it is a time consuming technique. Moreover, in this method, the longitudinal guard shell filled with insulation material can effectively minimize the radial heat exchange and the axial shunting exchange in the test stack [19], thus offering a higher measurement precision than the unguarded heat flow method in [6,7]. So far little research has been

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reported on this method being used to measure the thermal conductivities of coatings.

Thus, the main objectives of the present study are to develop the experimental setup for coating measurement and investigate how coating thickness influences the effective thermal conductivities of alumina coatings and the equivalent thermal conductivities of hybrid alumina/aluminum materials. In this article, the experimental details involving setup and measurement strategy were first introduced. Then, the microstructures of the thin and thick coatings with different thickness were measured and the equivalent thermal conductivities of the hybrid materials were derived.

2. Experimental details

2.1. Test sample and PEO process

In this study, right-circular cylinders of A356 aluminum alloy with height of 10 mm and diameter of 38 mm were employed as the substrates. All the uncoated samples were prepared from the same cast ingot so as to minimize the differences in composition and microstructure. The polished samples were as the electrodes emerged in an alkaline electrolyte containing dissolved sodium aluminate (7 g/l NaAlO₂) and potassium hydroxide (1 g/l KOH). A high voltage was utilized to induce micro-arc locally at the sample surfaces. During the PEO process, the current density was maintained at 0.1 A/cm² in the pulsed unipolar mode. The voltage was gradually increased with process time as the coating grew. The coating thickness was thereby primarily determined by the processing time.

After the PEO process, each coated sample was manipulated by removing the coating on one end face, and the surface roughness of the resulting free end face was then made to be the same as that of the coating remaining on the opposite end face. The surface roughness was measured with a profilometer (SJ-201P, Mitutoyo Corp., Kawasaki City, Kanagawa, Japan). The coating thickness was determined using an eddy current coating thickness gage (PosiTector 6000, DeFelsko Corp., Ogdensburg, New York, USA) with accuracy of $(\pm 0.5\,\mu m + 1\%$ of reading).

2.2. Experimental setup

The comparative guarded heat flow method was employed for the thermal conductivity measurement. As shown in Fig. 1, a test sample (an alumina coating with an aluminum alloy A356 substrate) was sandwiched between a pair of similar meter bars with the known thermal conductivity. The meter bar material (reference material) was selected to be stainless steel 310 (SS310) whose thermal conductivity provided by NPL (the National Physical Laboratory, UK). The meter bars and the sample had the same diameter. They were surrounded by an annulus of the insulation material of diatomaceous earth powder which was encased in a longitudinal aluminum guard shell. The guard shell was again surrounded with an alumina fiber blanket (Saffil Ltd. Cheshire. UK) to decrease the heat loss by natural convection. The purpose of this design was to minimize the radial heat exchange and the axial shunting exchange in the test stack (the two meter bars and the sample), thus establishing approximately the same temperature gradient in the shell and the test stack [19]. In addition, a hot plate (Scholar 170, Corning Inc., Corning, NY, USA) as the heat provider and a liquidcooled container as the heat sink with a mixture of water and ice were placed at the bottom and the top of the test stack, respectively.

In order to achieve excellent interfacial contact and minimize air gaps between the meter bars and the test sample, a dead weight (about 360 N) was indirectly applied on the stack across the heat sink. To ensure most of the load being imposed on the stack and the heat sink contacting the guard end face, a gap was left between the heat sink and the guard, and inserted with an annulus of a soft gap filler (Tflex 640, Laird Technologies, Chesterfield, MO, USA). This gap filling material was a boron nitride filled silicone elastomer with a good thermally conductive performance. Moreover, the mating faces of the meter bars were polished up to 0.03 µm finish and introduced with a good conductive silicone-based thermal grease (Tgrease 880, Laird Technologies, Chesterfield, MO, USA).



Fig. 1. Schematic of the test stack and guard system.

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