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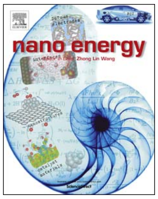

Nanoporous anodic alumina oxide layer and its sealing for the enhancement of radiative heat dissipation of aluminum alloy

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ABSTRACT

Various types of nanoporous anodic aluminum oxide layers and their sealings were studied to enhance the thermal emissivity and hence improve the heat dissipation of aluminum alloy for energy application. Dissipating heat fluxes from the anodized aluminum surfaces were measured using a modified steady-state method and investigated with respect to the various nanoporous morphologies obtained with different anodizing conditions and sealing methods. Results show that the anodized nanoporous oxide layers significantly enhance the thermal emissivity and heat dissipation of aluminum alloy, compared to bare aluminum alloy, and such enhancement is further improved with sealings. A thicker nanoporous oxide layer anodized in oxalic acid results in higher thermal emissivity and better heat dissipation than that in sulfuric acid, showing a darker color which is attributed to the more irregular and disordered pore size and pattern of the nanoporous oxide layer. The nanoporous oxide layer with cold NiF_2 or black sealing shows further enhancement in thermal emissivity and heat dissipation, demonstrating the highest enhancement in emissivity up to 0.906 in case of the nanoporous oxide layer anodized in oxalic acid with black sealing, which is seven times greater than that of bare aluminum. The nanoporous oxide layer with black sealing also results in the significant improvement of the cooling efficiency of a heat exchanger system of aluminum alloy by 36.4%, suggesting great energy saving for real energy application.

1. Introduction

Heat transfer by thermal radiation has been explored to overcome limitations of convection and conduction modes for managing heat-related problems in heating and cooling in vacuum environments and buildings [1–7]. Thermal radiation is traditionally explained by the Stefan-Boltzmann equation, where the radiative heat flux is dependent on the emissivity (ε) and temperature of the surface. In order to enhance the heat transfer by thermal radiation, both the higher emissivity close to that of an ideal black body (ε=1) and the higher temperature of the radiative surface, which are controllable by surface treatment, are required [8–10].

Heat dissipation by thermal radiation is related with a passive cooling state, which does not involve a fan or fluid circulation for cooling [11–13]. Since the passive cooling does not require additional energy to operate cooling units, it has been considered to be an economical and environmental-friendly technology [14]. This system has been applied to various electric devices, which do not require considerably active cooling [15,16]. In such a passive cooling state, thermal radiation of a heated surface is an effective way to transmit the heat, and thus the heat dissipation can be improved by the thermal radiation of cooling parts (e.g., heat sinks). Therefore, surface modification technologies to enhance the emissivity (ε) have a great potential in thermal management [17,18].

Ceramics [19–24] or carbon-based materials [25–27], which are known to have a high emissivity, can be coated on metallic materials having high thermal conductivity (e.g., Al and Cu alloys) to improve heat dissipation, because typical metallic materials have a low surface emissivity of ~0.1. In particular, anodizing or coatings with ceramic materials on aluminum alloys, which have been employed to various heat sinks, can be a promising technique to solve heat management problems [28–30]. Especially, an enhancement of the thermal emissivity and absorptivity of anodic aluminum oxide (AAO) with cobalt sulfide (CoS) sealing (i.e., black sealing) was reported for the application to solar absorbing films [31–33]. However, most of ceramic materials examined so far have relatively lower thermal conductivity

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than metallic materials [34,35]. Therefore, when only considering the conduction, the ceramic coating layer works as a thermal barrier, suppressing an effective heat transfer toward the coating surface with an increase in its thickness. In contrast, if the thickness of a coating layer is not enough to have a high emissivity, effective radiative heat dissipation could not be implemented [36]. Therefore, when considering the heat dissipation by thermal radiation, the optimized thickness of the coating layer is as critical as the coating material itself. Thus, it is critical to design the surface coating layer to have the optimized heat dissipation performance with the careful consideration of such complicated characteristics of the surface layer for efficient thermal management.

In this study, for a new surface coating layer, we employ anodizing and sealing techniques, which are scalable for mass production and readily available for industrial applications, to explore and maximize the combined effects of the synergistic increase in thermal emissivity and conductivity to enhance the overall heat dissipation performance of aluminum alloy. A nanoporous oxide layer directly grown on an aluminum substrate by the anodizing process enhances the surface's radiative emissivity. In addition, unlike the composite coatings using carbon-based materials or polymer resin that have relatively high interfacial thermal resistance with substrate metals [7,25–27], the anodic aluminum oxide has negligible interfacial thermal resistance with the aluminum substrate [37], which is beneficial for the overall heat dissipation performance. In order to find out the optimized surface conditions for the best heat dissipation performance of the anodized aluminum oxide (AAO) surface, the effects of anodizing conditions (i.e., anodizing time and temperature) and additional sealing treatments (e.g., hydrothermal, cold NiF_2, and black sealings) on the heat dissipation of aluminum alloy are explored and discussed with respect to the porous nanostructures of the AAO and thermal emissivity. For the effective measurement of such properties, we introduce a measurement method for the dissipating heat flux from a modified surface, such as anodized aluminum alloy, so that it can provide a reliable estimation for the heat dissipation performance of such a modified metallic surface. Furthermore, an enhancement of cooling performance of the treated aluminum alloy with improved dissipating heat flux is demonstrated for real applications.

2. Materials and methods

2.1. Sample preparation and evaluation methods

A plate of Al 1050 alloy (99.5%Al – 0.24% Fe – 0.15% Si, Alcoa Inc., USA) was used as a substrate and cut into 50×50×1.5 mm in size. The plates were cleaned ultrasonically in acetone before use. The substrates were electropolished in ethyl alcohol (99.9+%, SK Chemicals, Korea) and perchloric acid (Junsei Chemical Co., Ltd., Japan) mixture solution (3:1 in volumetric ratio) at 25 °C under constant voltage of 20 V for 2 min.

Two different types of electrolytes were employed for anodizing, including 0.3 M (2.7 wt%) oxalic acid (Junsei Chemical Co., Ltd., Japan) and 1.63 M (15 wt%) sulfuric (Junsei Chemical Co., Ltd., Japan). Aeration into the electrolytes was used for agitation, and the temperature of the electrolyte was regulated in a range from 0 to 30 °C. Constant current density of 50 mA/cm^2 was applied for anodizing. In order to control the thickness of anodic oxide layer, the anodizing time was also regulated in a range from 10 to 40 min. After the anodizing, hydrothermal, cold NiF_2, and black sealing methods were applied respectively for comparison. Detail conditions for the anodizing and sealing processes were summarized in Table 1.

Dissipated heat from the anodized aluminum surface was measured (see Section 2.2 for the details of the dissipated heat flux measurement setup). During the measurement of the dissipated heat, the temperature of ambient was maintained at 25 °C using an air conditioner. Pore structures of anodic oxide layer were observed using field emission scanning electron microscopy (FE-SEM, Hitachi S-4700, Japan). The thickness of anodic aluminum oxide was measured from the cross-sectional image obtained by the FE-SEM. In order to obtain clear images showing pore structure, the surface of anodized aluminum was mildly polished with 1 µm magnesia (MgO) and diamond paste using a nap cloth. Polished specimens were cleaned with 10 vol% H_3PO_4 for 30 s and washed in deionized water with ultrasonication. Captured optical and SEM images were analyzed using Image-Pro software to determine the averaged values and the standard deviations of a gray level of surface appearance, pore diameters, eccentricity of pore, inter-pore distance, and porosity.

Table 1
Conditions of anodizing and sealing processes.

Electrolyte	1.63 M (15.0 wt%) sulfuric acid and 0.3 M (2.7 wt%) oxalic acid	
Temperature	0, 15, 30 °C	
Current density	50 mA/cm^2	
Process time	10–40 min	
Sealing solution	Hydrothermal	98 °C, 30 min
	Cold NiF_2	2.5 g/l, 25 °C, 30 min
	Black	1st step: 250 g/l cobalt acetate ($Co(C_2H_3O_2)_2$), 45 °C, 20 min 2nd step: 20 g/l, ammonium sulfide ($(NH_4)_2S$), 25 °C, 30 min

Emissivity of the anodized aluminum surface was measured using Fourier transform infrared (FT-IR) spectroscopy (Nicolet Avatar 360 FT-IR spectroscopy, Thermo Scientific, USA) at 200 °C, where the measured range of the wavelength was 5–20 µm. Four samples fabricated in the same conditions were measured to estimate the average value and the standard deviation at the given condition. The improved heat dissipation performances were compared to those of the bare aluminum alloy by cooling hot water contained in a cylindrical aluminum tube with anodized oxide layers.

2.2. Dissipated heat flux measurement setup

Fig. 1 presents a schematic diagram of the designed radiation heat flux measurement setup modified from the thermal conductivity measurement setup using a flow-meter method [38,39]. Stainless steel 304 block (thermal conductivity=16.2 W/m K at 200 °C, POSCO, Korea) cut into 50×50×120 mm was used as a meter bar and thermally insulated to minimize heat loss. The fabricated aluminum alloy sample

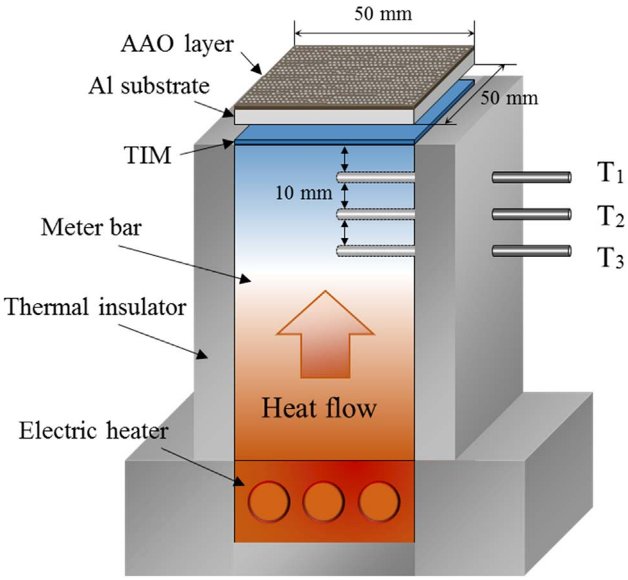

Fig. 1. Schematic diagram of dissipated heat measurement setup.

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