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within H2020



Proceedings of the International Symposium on  
Thermal Effects in Gas flows In Microscale  
October 24-25, 2019 – Ettlingen, Germany

**ISTEGIM 2019 - 283409**

## **MEASUREMENT OF HEAT TRANSFER IN HIGH KNUDSEN NUMBER FLOW FROM ANODIC OXIDE ALUMINUM FILMS**

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### **KEY WORDS**

Gas-Surface Interaction, Thermal Accommodation Coefficient, Conductive Heat Transfer, Vacuum

### **ABSTRACT**

*The conductive heat transfers thorough a rarefied gas were studied on anodic oxide aluminum films with different anodizing times. The heat flux between two surfaces with different temperatures was measured from the free-molecular to near free-molecular flow regimes and analyzed to obtain the energy accommodation coefficient on anodic oxide aluminum film. The energy accommodation coefficients were qualitatively compared between a non-treated surface and the anodizing time of 30 min. The effect of anodizing time was studied by preparing a 90-min sample surface, where the film would be partially dissolved by long time exposure. SEM images were obtained for the three sample surfaces. Unfortunately the images were in low magnification, and it was difficult to observe the hexagonal cylindrical cell structures. Even though, the 30-min sample surface was shown to be rough, and it was relatively smooth and powdery for the 90-min sample surface. The formation of the anodic oxide aluminum film was checked by an electrical resistance. By using these sample surfaces with anodic oxide aluminum films, the heat fluxes were measured, and they were well fitted by the fitting curve. The obtained energy accommodation coefficients suggest that the value increases by anodizing an aluminum sample surface, while it decreases with increasing the anodizing time up to 90 min.*

### **1. INTRODUCTION**

The heat transfer from a hot to a cold surface in vacuum is a basic problem. In vacuum, the Knudsen number is large due to a large mean free path of gas molecules. In a high Knudsen number flow, the number of collisions between gas molecules and a solid surface cannot be neglected compared with that between gas molecules. Then, the gas-surface interaction plays an important role in a thermal-fluid field.

The gas-surface interaction, which is scattering processes of gas molecules from a solid surface, is known to be a complicated process depending on many parameters of gas molecules and a solid surface [1]. To understand and represent the gas-surface interaction practically, it is enough and convenient to obtain statistically averaged characteristics. The accommodation coefficient [2] is often employed for such purpose in the gas-surface interaction. For a heat transfer problem, energy transfer is related; thus, the energy

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accommodation coefficient (EAC) or the thermal accommodation coefficient, which are equivalent for a static equilibrium gas, is employed. This EAC  $\alpha$  is defined as [1 - 3],

$$\alpha = \frac{E_i - E_r}{E_i - E_s}, \quad (1)$$

where  $E_i$ ,  $E_r$  and  $E_s$  are the mean incident and reflected energy fluxes, and the energy flux of gas molecules fully accommodated to the surface, respectively. This EAC represents an averaged probability, fraction or efficiency of the energy transfer process between gas molecules and a solid surface.

The thermal accommodation coefficients, equivalent to EAC, for various pairs of gas species and surface materials have been measured [2]. It has been considered that EAC becomes large, *i.e.* gas molecules accommodate well to a surface, when the surface is rough. Therefore, the engineering surfaces have been considered to have EAC around unity. Recent measurement [4] studied the effect of the surface roughness on EAC by comparing the results on the machined, polished and deposited surfaces with or without plasma treatment. The rms roughness of these surfaces were reported as  $\sim 2 \mu\text{m}$  for machined surfaces and  $\sim 0.02 \mu\text{m}$  for polished surfaces. In the work, it was reported that the surface roughness was appeared to have only a minor effect.

In this study, the effect of surface roughness on the energy accommodation coefficient is studied by employing an anodic oxide aluminum film. An anodic oxide aluminum film is known to the hexagonal cylindrical cell structure with several to several hundreds nm diameter [5, 6]. It is also important that the anodization process is a wet process, and the surface anodization would roughen all the area of sample surfaces even though there are distortions, large adsorbates or dimples on sample surfaces.

## 2. MEASUREMENTS

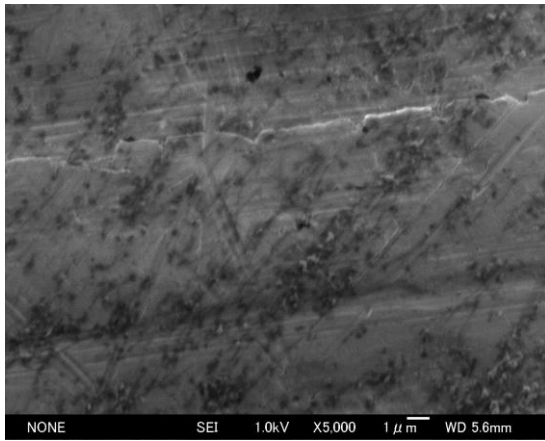
The heat flux from a sample surface of an anodic oxide aluminum film in vacuum was measured to extract EAC.

### 2.1 Sample surface

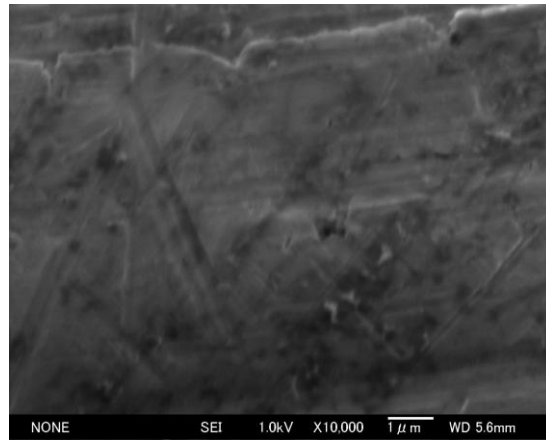
The sample surfaces were prepared by anodizing a 0.3-mm-thick aluminum plate (A1050P, AS-ONE), which is a general-purpose product. A strip of a plate was cut from the plate. Then, it was wiped by acetone, dipped in a solution of NaOH, and rinsed in distilled water. The strip was then immersed in a diluted solution of  $\text{H}_2\text{SO}_4$  (1 mol/L), and the electric current was applied with the formation voltage of 20 V and the current density of  $12.5 \text{ mA/cm}^2$ . To keep the temperature of the solution, chilled water was circulated through a coil placed in a solution to keep temperature constant at around  $5^\circ\text{C}$ .

The anodizing time is usually set about 10 - 30 min [5, 6]. It is well known that a hexagonal cylindrical cell structure of an anodized film is chemically dissolved by long time exposure to the solution. Thus, we selected 30 min for an anodized sample surface and 90 min for a long time exposure sample surface. These sample surfaces were compared with a non-treated sample surface, which is hereafter called as a 0-min sample surface.

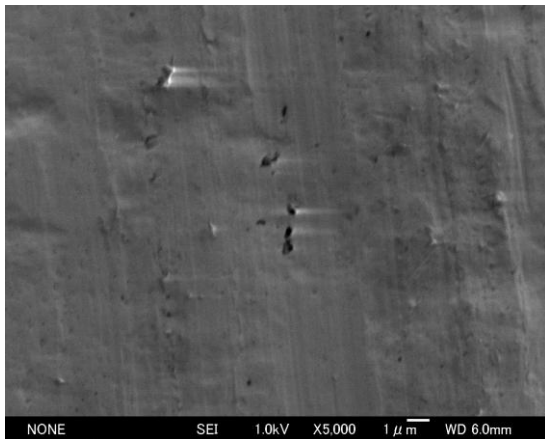
The sample surfaces were measured by SEM (JSM-7000F, JEOL). The obtained SEM images are shown in Fig. 1. We tried to obtain high magnification images; however, it was difficult to observe nanostructures of an anodic oxide film, because of the nature of the film as an insulator. This nature was easily checked by measuring an electrical resistance of the sample surface with a tester. From the images, it is easily observe that the sample surfaces are quite rough. There are many scratches and roughness already in the 0-min sample surface. Several quite large bumps or dents dimple are appeared on the 30-min sample surface. Meanwhile, the sample surface becomes relatively smooth but powdery for the 90-min sample surface. This might be the result of the dissolution of the film by long time exposure [5, 6].



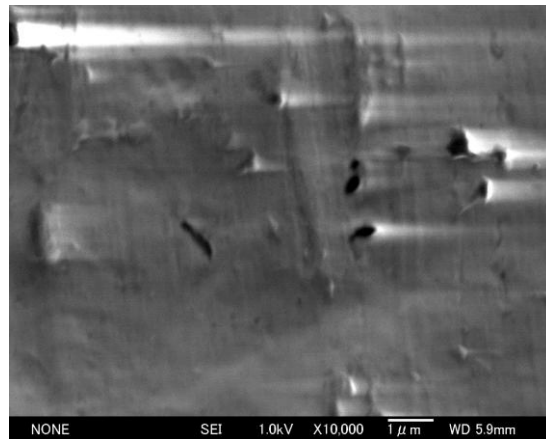
(a-1) 0-min,  $\times 5,000$



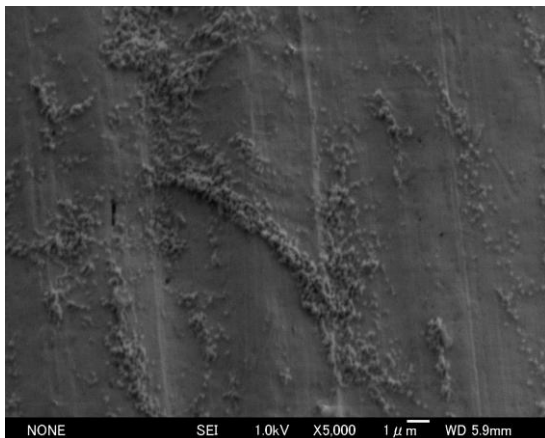
(a-2) 0-min,  $\times 10,000$



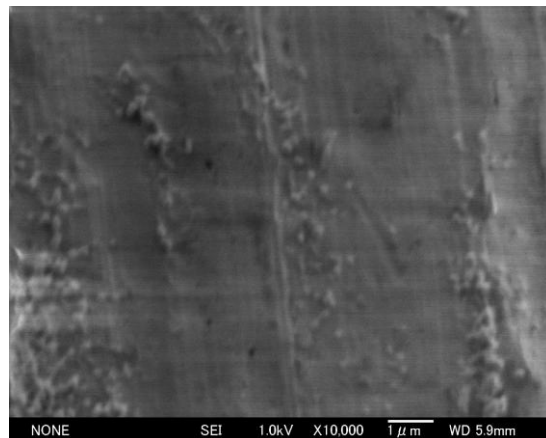
(b-1) 30-min,  $\times 5,000$



(b-2) 30-min,  $\times 10,000$



(c-1) 30-min,  $\times 5,000$



(c-2) 90-min,  $\times 10,000$

**Figure 1:** SEM images of the anodic oxide aluminum sample surfaces: (a) 0-min, (b) 30-min and (c) 90-min with the magnification of (1)  $\times 5,000$  and (2)  $\times 10,000$ .



## 2.2 Method

The heat flux between two surfaces with different temperatures in the free-molecular flow regime is explained by the energy transfer by molecules. The heat flux in the free-molecular flow regime  $q_{FM}$  is theoretically expressed as,

$$q_{FM} = \frac{1}{8} \alpha \frac{\gamma+1}{\gamma-1} \frac{\bar{v}}{T} p \Delta T, \quad \bar{v} = \sqrt{\frac{8kT}{\pi m}}, \quad (2)$$

where  $\gamma$ ,  $\bar{v}$ ,  $T$ ,  $p$ ,  $\Delta T$ ,  $k$  and  $m$  is the specific heat ratio, the mean molecular speed, temperature, pressure, the temperature difference of two surfaces, the Boltzmann constant, and the molecular mass of the gas, respectively. The heat flux is proportional to pressure and EAC; thus, by measuring the heat flux as a function of pressure, EAC can be derived.

However, it was not easy to accurately measure only in the free-molecular flow regime, *i.e.* at a high vacuum condition, in a simplified low-cost apparatus as explained in our previous study [7 - 9]. Therefore, we used pressure conditions slightly higher than the upper limit of the free-molecular flow regime. A general model expression to describe the heat flux from the free-molecular flow regime up to the continuum flow regime was employed [10, 11], which is expressed as,

$$\frac{1}{q} = \frac{1}{q_{FM}} + \frac{1}{q_C}, \quad (3)$$

where  $q_C$  is the heat flux in the continuum flow regime. The heat flux as a function of pressure would be slightly curved by this expression. The obtained heat flux was fitted by this expression to obtain EAC in Eq. (2).

## 2.3 Setup

The experimental setup is explained in detail elsewhere [7 - 9].

A spherical vacuum chamber made by Pyrex was employed. The inner radius of the chamber  $R_C$  was 49.5 mm. The chamber was immersed in a water bath to keep the temperature of the chamber  $T_C$  at a room temperature. The measured temperature on the chamber was about 290 K. The test gas was supplied from commercially available gas cylinders of pure helium and argon. The pressure in the chamber was set by measuring with a temperature-controlled capacitance manometer (Baratron<sup>®</sup> 627B, MKS). The pressure conditions were limited below 1.4 Pa to be in the near free-molecular flow regime, so that the effect of the general model expression of Eq. (3) was minimized.

The sample surfaces of the anodic oxide aluminum films were placed on the surfaces of a tiny flat-shaped heater with the size of  $11.8 \times 12.0 \times 0.38 \text{ mm}^3$  (Toyo Precision Parts MFG). The heater was placed at the center of the spherical vacuum chamber. Since the heater was very small compared with the spherical vacuum chamber, it was appeared that the system was able to be approximated as a concentric spherical shells system [7 - 9]. Under such approximation, the sample surfaces were treated as a sphere with radius  $R_H$  having the same surface area with two sample surfaces, and the heat flux in the continuum flow regime can be expressed as,

$$q_C = \kappa(T_C) \frac{\mathcal{T}^{\omega+1}-1}{(\omega+1)(\mathcal{T}-1)} \Delta T \frac{R_C R_H}{R_C - R_H} \frac{1}{R_H^2} \quad (4)$$

where  $\kappa(T)$ ,  $\omega$ ,  $\mathcal{T}$  is the thermal conductivity of the gas at temperature  $T$ , the thermal conductivity index and the temperature ratio of two surfaces, respectively. The thermal conductivity was assumed to be proportional to  $T^\omega$  following the model with the inverse power law potential for a monatomic gas.

An analog electrical bridge circuit was employed to maintain the temperature of the heater. The heat transfer from the sample surfaces was measured by an electrical consumption to keep the sample temperature constant. The energy consumption consisted of the heat conduction through gas, which we wanted to measure, the radiation and the heat loss through the electrical leads of the heater. Since only the first term depends on pressure, this term could be extracted by evaluating the latter two terms by the heat flux in the vacuum limit. The convection was negligible due to the low pressure condition.

The temperature of the sample surfaces were estimated from the electrical resistance of the heater. The calibration curve between temperature and the electrical resistance of the heater with sample surfaces was measured beforehand. The sample surface temperature  $T_H$  was set about 360 K.

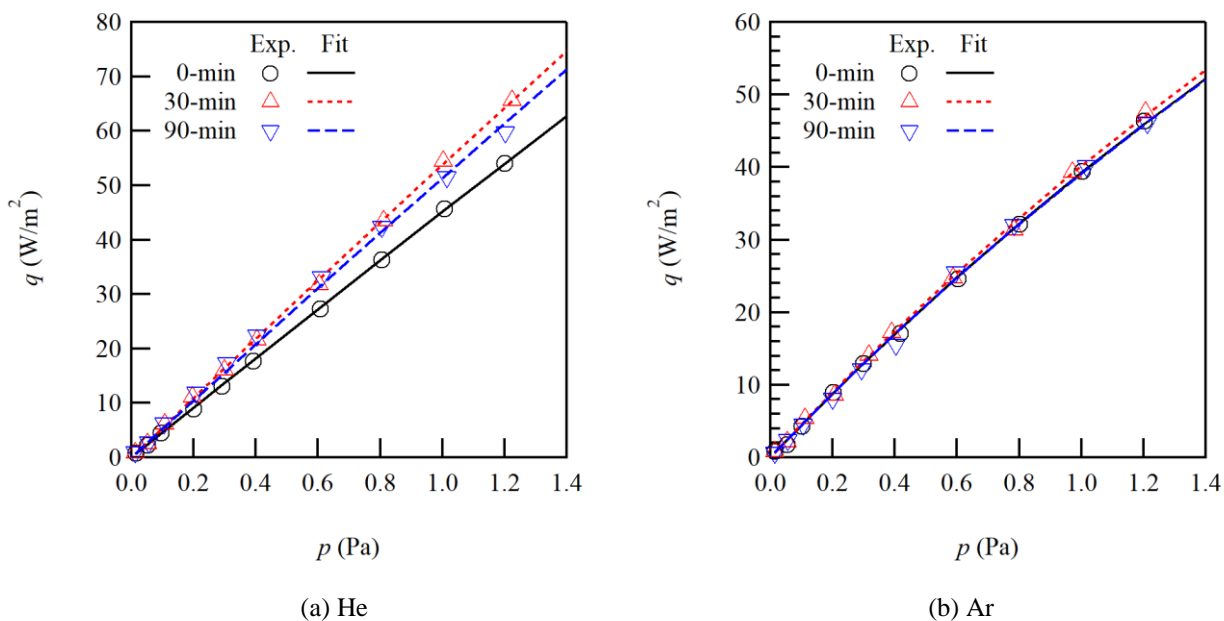
The uncertainty of the measurement was quite difficult to evaluate; however, the error of the measurement was known to be less than 5 % [7 - 9]. Therefore, it could be possible to compare the results qualitatively between the conditions.

### 3. RESULTS AND DISCUSSIONS

#### 3.1 Heat flux

The heat flux was measured as a function of pressure four times for each conditions to check the repeatability. Typical examples of the measured heat fluxes and the fitted curves by Eq. (3) for the 0-min, 30-min and 90-min sample surfaces are plotted in Fig. 2 for He and Ar. From the figure, the experimentally measured data are well explained by the fitting curves.

For the three samples, the size of the heat flux is clearly different for He; while it is almost similar for Ar. It is well known that He is quite sensitive to the surface due to its light molecular mass and small size. From the figure for He, the heat flux increases by the anodization. However, it slightly decreases when the anodizing time increases up to 90 min.



**Figure 2:** Typical results of the measured heat fluxes and the fitted curves as a function of pressure for 0-min, 30-min and 90-min sample surfaces for (a) He and (b) Ar.

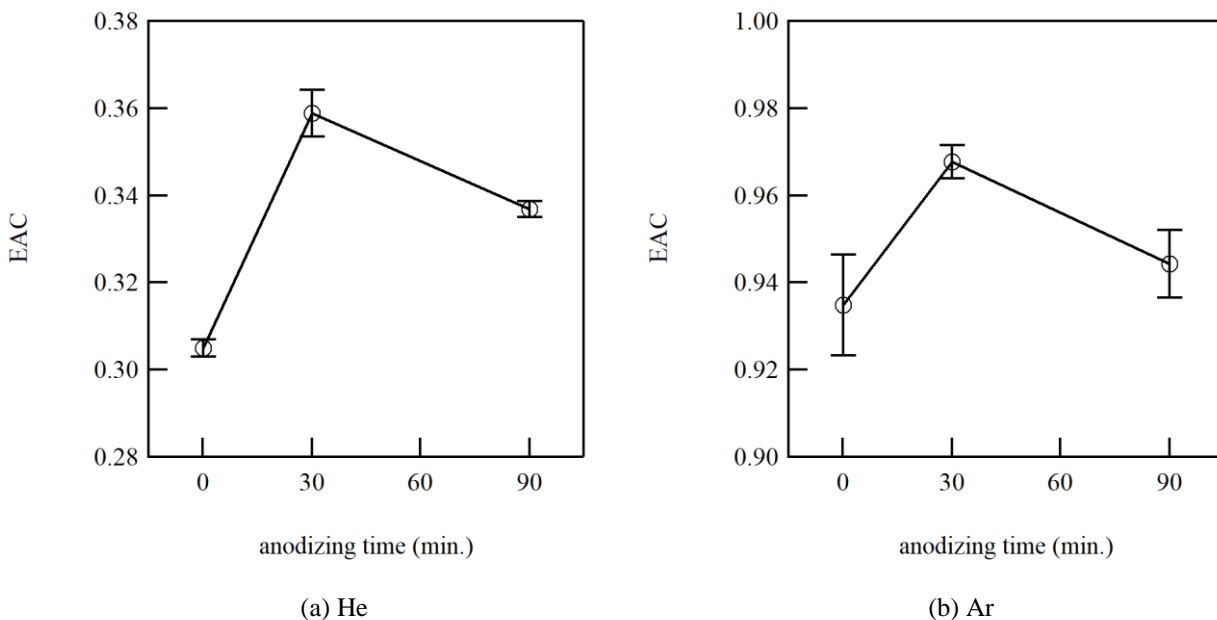
### 3.2 Energy accommodation coefficient

By fitting the measured heat flux with the curve, EAC in Eq. (2) was obtained. EAC was obtained for four measurements on each condition. To evaluate the repeatability, the standard error of the data was calculated. The size of the standard error could be an idea for the uncertainty of the measurement. The averaged EAC are tabulated with the standard error in Table 1 and plotted with the error bar representing the standard error in Fig. 3 for He and Ar.

From the figure, EAC increases at the anodizing time of 30 min; while it decreases at 90 min. Compared with Fig. 2, it is easy to imagine this characteristics for He, the trend appears even for Ar, where the discrepancy between the conditions are small. The error bars for Ar looks much larger than that for He. However, the absolute values of EAC are quite different for these two gases; EAC for Ar is about three times of that for He. Therefore, if we focus on the relative error, they are almost in the same range; less than 1.5 %. This result indicates that the measurement accuracy was independent of gas species, even though the size of the error bar looks different.

	0-min	30-min	90-min
He	$0.3050 \pm 0.0020$	$0.3589 \pm 0.0053$	$0.3369 \pm 0.0019$
Ar	$0.9349 \pm 0.0115$	$0.9677 \pm 0.0038$	$0.9443 \pm 0.0077$

**Table 1:** A table of the measured EAC with the standard error on 0-min, 30-min and 90-min sample surfaces for He and Ar.



**Figure 3:** The measured EAC as a function of the anodizing time on 0-min, 30-min and 90-min sample surfaces for (a) He and (b) Ar. Error bars show the standard error of 4 measurements for each condition.



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Comparing the results of 0-min and 30-min sample surfaces, EAC is clearly shown to increase by anodizing an aluminum surface. Even though it was difficult to observe the nano-scale hexagonal cylindrical cell structure in our SEM images in Fig. 1, the anodic oxide aluminum film was formed on the sample surfaces, which was validated by an electrical resistance of the surface. Thus, the surface should have many nano-scale pores and be roughened, indicating the coincidence with the well-known characteristics of EAC.

In Ref. 4, it is stated that the effect of macroscopic surface roughness plays only a minor role in EAC by comparing the results on the machined and the polished surfaces for argon, nitrogen and helium. Meanwhile, there is an apparent change by the anodization as a roughen surface in this study. If you carefully check the results in the reference, EAC for He on a machined 304 Stainless steel surface was 0.46, and that on a polished surface was 0.42. Though the authors suggested that the effect was minor, EAC decreased by reducing the roughness. The difference in the relative size of the effect from this reference to our study could be coming from the approach to roughen a surface. In the reference, the machining process was employed for changing the roughness. This means that the surface morphology was modified only for the accessible area from outside. Whereas, the anodization process, which is a wet process, was employed in this study, and it could modify the surface morphology to the whole surface area where gas molecules can approach. Therefore, the variation between sample surfaces became large in this study. Therefore, it is reasonable to show larger EAC on the 30-min sample surface than that on the 0-min sample surface.

For 90-min samples, EACs decreased to nearly the same value with those on the 0-min sample surfaces. This could be coming from the dissolution of the anodic oxide aluminum film on the surface by long time exposure [5, 6], and the relatively smooth surface is realized as shown in Fig. 1 (c). The powdery parts would increase EAC a little, but the size of the powder is quite large compared with the nano-scale pores, resulting in only a little increase in EAC from the 0-min sample surface.

#### 4. CONCLUSIONS

The heat flux from an anodic oxide aluminum film was measured in the free-molecular to near free-molecular flow regime. The sample surfaces were prepared for three conditions; without anodization (0-min), the anodizing time of 30 min and 90 min. For our anodization conditions, 90 min was too long, and a part of the film would be dissolved. SEM images were taken for the three sample surfaces; 0-min, 30-min and 90-min. The surface seemed roughened by the anodization, but it was relatively smoothed and became powdery for 90-min sample surfaces. We failed to capture the detailed hexagonal cylindrical cell structure; however, the formation of the film was validated by an electrical resistance.

The heat flux from the sample surface was measured, and the obtained heat flux was fitted by the curve to extract the energy accommodation coefficient. The obtained energy accommodation coefficient was larger for the 30-min sample surface than that for the 0-min sample surface, indicating the increase in the surface roughness by the anodization process. Meanwhile, the value decreased by increasing the anodizing time up to 90 min. This result coincided with the relatively smooth surface observed in the SEM image.

#### ACKNOWLEDGEMENTS

This work was partially supported by JSPS KAKAENHI Grant No. 18K03946.

This work was partially supported by Nanotechnology Platform Program (Nanofabrication Platform Consortium) of the MEXT, Japan. The SEM images were acquired in Nagoya University.



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