Fabrication of radiative cooling devices using Si$_2$N$_2$O nano-particles

Hidetoshi MIYAZAKI,*, Kazuya OKADA, Kenta JINNO and Toshitaka OTA

Interdisciplinary Graduate School of Science and Engineering, Shimane University, 1060 Nishikawatsu, Matsue 690–8504, Japan
*Ceramic Research Laboratory, Nagoya Institute of Technology, 10–6–29 Asahigaoka, Tajimi, Gifu 507–0071, Japan

In this study, Si$_2$N$_2$O particles intended for use as radiative cooling materials were synthesized by heat-treatment of mixtures of Si and SiO$_2$ powders in an N$_2$ atmosphere at 1450°C. The resulting Si$_2$N$_2$O particles were used as Si$_2$N$_2$O coatings on Al substrates as a radiative cooling device, and these coatings were generated using pulsed electrophoretic deposition. The average difference in temperature from the ambient temperature of the SSRM devices with the film thicknesses of 6.5, 13.2 and 35.2 μm were -0.82, -0.44 and +0.83°C, respectively. A radiative cooling effect was observed for the device with the film thicknesses of 6.5 and 13.2 μm.

Key-words : Si$_2$N$_2$O, Film, Radiative cooling, Electrophoretic deposition

©2016 The Ceramic Society of Japan. All rights reserved.

[Received July 4, 2016; Accepted September 10, 2016]

1. Introduction

The clear night sky acts as a passive heat sink and is able to lower the temperature of Earth’s atmosphere, the effect is known as the radiative cooling phenomenon. The sky allows heat to escape because infrared (IR) radiation in the wavelength range from 8 to 13 μm is not absorbed by atmospheric gases, and hence this wavelength region is known as the atmospheric window.\(^1\) Materials on Earth’s surface that can emit IR radiation in this window also have the ability to lose heat via radiative cooling. A rule that can assist in the selection and design of such materials is that compounds will emit IR radiation across approximately the same wavelength range over which they absorb heat.\(^2\) Thus, in order to design radiative cooling materials, we must select compounds with high absorption in the 8 to 13 μm wavelength range and low absorption elsewhere. These devices can cool buildings and vehicles at all hours with no electrical energy, therefore; the radiative cooling devices are useful for energy savings.

Silicon monoxide, silicon dioxide, silicon oxynitride and polyvinyl chloride are all well-known radiative cooling materials, with a significant capacity for cooling.\(^3\)\(^-\)\(^4\) In previous research, Granqvist et al. designed radiative cooling materials using SiO and SiO$_2$ films, based on their radiative cooling abilities.\(^5\) In our previous study, we synthesized Si$_2$N$_2$O particles by a solid state reaction using Si and SiO$_2$ powders, and confirmed that the resulting Si$_2$N$_2$O particles also showed broad absorption in the atmospheric window range (8 to 13 μm).\(^6\) Using the Si$_2$N$_2$O particles, new spectrum selective radiative material devices (SSRM devices) can be designed.

In previous reports, SSRM devices have been formed as coatings of radiative cooling material on metal substrates.\(^2\)\(^-\)\(^4\) In order to fabricate an SSRM device, we used the pulsed electrophoretic deposition (EPD) method. In this method, coatings can be formed using powders alone and there are few limitations on source materials. Furthermore, the pulsed EPD method enables us to form a dense film on metal substrates.\(^6\)\(^-\)\(^7\) A Si$_2$N$_2$O radiative cooling device can thus be formed easily using the above-mentioned Si$_2$N$_2$O particles with the pulsed EPD method.\(^6\)\(^-\)\(^7\)

In the present investigation, we synthesized Si$_2$N$_2$O particles using the previously reported method,\(^6\) and fabricated radiative cooling devices with Si$_2$N$_2$O coatings on metal Al plates using Si$_2$N$_2$O nano-particle with a pulsed EPD. Furthermore, we evaluated the radiative cooling ability of the obtained devices.

2. Experimental procedure

The Si$_2$N$_2$O powder was fabricated by heat-treatment of a mixture of Si powder (Kojundo Chemical Laboratory Co. Ltd.) and SiO$_2$ powder (Quartz, Kojundo Chemical Laboratory Co. Ltd.) at 1450°C for 1 h in an N$_2$ atmosphere, where the molar ratio of Si to SiO$_2$ in the powder mixture was 3 to 1. The suspension solution for EPD was prepared using ion-exchanged water and the obtained Si$_2$N$_2$O particles at the concentrations of 0.005 and 0.5 vol.%. The precursor suspension solution was dispersed for 10 min using an ultrasonic cleaner bath, and subsequently was magnetically stirred for 10 min. The substrates used as electrodes were planar Al metal plates, and the substrates were placed 1 cm apart in the pulsed EPD cell. The opposite side of the deposition side was masked using freon tapes to prevent Si$_2$N$_2$O deposition. Electrical bias was then applied to the substrates at 1 kHz (a square wave) with a bias voltage of 0 V or alternatively of −0.0032 to −0.5 V using a universal source (HP-3245A, Agilent Technologies). In the previous investigation, we fabricated Y$_2$O$_3$ film by a pulsed EPD method using Y$_2$O$_3$ nano-particles, and the pulse frequency of the deposition. Electrolysis of the solution was observed with low frequency deposition, and this electrolysis caused some damage (pores and voids) to the resulting film. On the other hand, we could obtain dense and crack-free films with higher than 1 kHz frequency deposition. Therefore; we employed the deposition condition of 1 kHz frequency in this investigation. After applying bias to the substrates, the resulting films were removed from the suspension solution and the masking tape was peeled off. The films were then dried at 200°C. The resulting substrates with Si$_2$N$_2$O coatings were cut to 2 × 2 cm, and the
samples were used for evaluation of the radiative cooling ability. We call this sample the “device” hereinafter.

The structures of the resulting powders were characterized using an X-ray diffractometer (XRD, Rigaku Miniflex, Rigaku) with Cu Kα radiation (30 kV, 15 mA). IR absorption spectra of these materials in KBr pellets were measured using a Fourier transform infrared spectrometer (Jasco FT-IR 660 Plus). The microstructure in the nano-particle of Si2N2O was characterized by transmission electron microscopy (TEM, EM-002B; Topcon Corp.). The microstructures and the film thickness of the resulting films were observed using a scanning electron microscope (SEM, JSM-6510, JEOL). The temperatures of the devices were evaluated using thermocouples (USB-TC01, National Instruments). Figure 1 shows a schematic drawing of the measuring temperatures.

3. Results and discussion

As noted above, Si2N2O powders were fabricated at 1450°C under a N2 atmosphere for 1 h, from a mixture of Si and SiO2 powders combined at a Si/SiO2 molar ratio of approximately 3:1. Figure 2 shows the XRD pattern (a), the IR spectrum (b) and the TEM image (a) of the Si2N2O particles prepared along with the previous report. The XRD pattern indicated that the obtained particle was Si2N2O as the main phase and Si3N4 as the second phase. The particle showed a broad absorption in the atmospheric window region from the IR spectrum. The TEM observations of the resulting Si2N2O particles indicated that they were a sub-rounded form, and their grain size was about 20 to 80 nm. This nitridation reaction of Si and SiO2 powders takes place in two steps. In the first, silicon and silicon dioxide react to produce gaseous silicon suboxide which, in the second step, reacts with nitrogen gas to form Si2N2O.5) Thus, nano-order size Si2N2O particles were assumed to be obtained.

Using the Si2N2O particles, we used the pulsed EPD method to generate Si2N2O coatings on Al substrates with three different film thicknesses. The deposition conditions of the films are shown in Table 1. Figure 3 presents surface and cross-sectional SEM images of the resulting films. All the films showed a flat surface and dense without pores. We evaluated the film thickness using these positions and two other positions, and 5 points of thickness were calculated for each position. The film thicknesses were determined using an average of 15 of the above points, and the thicknesses of the resulting films were 6.5, 13.2 and 35.2 μm. Where thick film (greater than 20 μm) could not be obtained under a low bias voltage, a high bias voltage of 0.5 V was applied to the substrate in order to form a film with a thickness of about 30 μm. When the film thickness was large, the film resistance was high because the Si2N2O had high resistivity. The effective electrical field of the film surface decreased with increasing film thickness because of the increased film resistance. Thus, the above film thickness was assumed to be close to the limit of Si2N2O film formed by the pulsed EPD method. The resulting samples were then cut to 2 × 2 cm for evaluation of their radiative cooling ability.

We evaluated the radiative cooling ability of the resulting devices by measuring the device temperatures and the blank substrate (close to ambient temperature) where the blank substrate was a 2 × 2 cm Al plate. In order to confirm an aluminum substrate to measure a radiative cooling ability, we measured an aluminum substrate (as blank) and ambient temperature. The temperature difference between the aluminum substrate and ambient temperature was less than 0.1°C. Thus the aluminum substrate had little effect on the measuring temperatures of radiative cooling devices. Figure 4 presents the temperatures of the devices and the blank. The device temperatures at the film thicknesses of 6.5 and 13.2 μm were consistently lower than the ambient temperature. In contrast, the device temperature at a film thickness of 35.2 μm was consistently higher than the ambient temperature. The average temperature differences of devices from the ambient temperature with film thicknesses of 6.5, 13.2 and 35.2 μm were −0.82, −0.44 and +0.83°C, respectively. A radiative cooling effect was observed for the devices with film
thicknesses of 6.5 and 13.2 μm, the results that were closely consistent with the previous reports.3) In contrast, a warming effect was observed for the device with film thickness of 35.2 μm. Increasing devices’ film thicknesses caused an increase in their absorption of the IR radiation, where the radiation was self-emission from the devices and emissions from their surroundings (i.e., walls, persons, and so on). Thus, radiative cooling effects were observed only for the devices with film thicknesses of 6.5 and 13.2 μm (thin films).

4. Conclusion

We fabricated radiative cooling devices by deopsiting Si$_2$N$_2$O particle coatings on Al substrates, and evaluated the devices’ cooling ability. The present investigation indicates that radiative cooling devices or warming devices can be designed using Si$_2$N$_2$O particles and the cooling or warming ability of the devices is dependent on the thickness of the Si$_2$N$_2$O coating layer.

References

1) W. B. Grant, Appl. Opt., 29, 451–462 (1990).
2) C. G. Granqvist and A. Hjortsberg, J. Appl. Phys., 52, 4205–4220 (1981).
3) A. P. Raman, M. A. Anoma, L. X. Zhu, E. Rephaeli and S. Fan, Nature, 515, 540–544 (2014).
4) T. S. Eriksson and C. G. Granqvist, J. Appl. Phys., 60, 2081–2091 (1986).
5) H. Miyazaki, S. Yoshida, Y. Sato, H. Suzuki and T. Ota, J. Ceram. Soc. Japan, 121, 242–245 (2013).
6) L. Besra, T. Uchikoshi, T. S. Suzuki and Y. Sakka, J. Am. Ceram. Soc., 91, 3154–3159 (2008).
7) H. Miyazaki, A. Ichikawa, H. Suzuki and T. Ota, Adv. Mater. Sci. Eng., 2016, 9387651 (2016).