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# Fabrication and characterization of aluminum nitride thick film coated on aluminum substrate for heat dissipation

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## ABSTRACT

For effective heat dissipation in high-power LED applications, aluminum nitride (AlN) thick films as thermally conductive dielectric layers were developed, which were deposited on an Al substrate by aerosol deposition (AD). The aerosol-deposited AlN thick films on Al substrates have advantages over conventional polymer-based dielectric substrates or ceramic substrate mounted heatsink systems including an epoxy adhesive, such as excellent heat dissipation capacity and low thermal resistance. AD is an effective method to fabricate high-quality AlN thick film without the  $Al_2O_3$  phase because the film is formed at room temperature. Highly dense and well-adhered, pure AlN thick films with thicknesses up to  $30 \mu$ m were deposited on an Al substrate. AlN-Al\_2O\_3 and AlN-polyvinylidene fluoride (PVDF) composite films were also deposited on an Al substrate in order to investigate the effect of  $Al_2O_3$  and polymer on the microstructure and thermal properties. Among the films, pure AlN thick film exhibited the highest dielectric strength, the highest thermal conductivity, and the lowest thermal resistance. Therefore, it can be expected that the aerosol-deposited AlN thick film on Al substrate could be used as a powerful heatsink. © 2016 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

#### 1. Introduction

With enhanced performance requirements for more compact, miniaturized, and high-density electronic packages, considerable attention has been given to the heat dissipation problem of electronic systems [1]. The efficient removal of heat from the semiconductor junction to the ambient environment has become a crucial issue of electronic packages to offer optimal performance without functional failure. In the past, the thermal design of electronic packages was the most important topic related to thermal management. However, while module and device heat fluxes have been drastically increased, thermal design margins are continuing to decline for heat dissipation, which has led to an increase in the demand for advanced thermal management materials [2].

Effective thermal management materials require a low coefficient of thermal expansion (CTE), as well as low density materials with high thermal conductivity for thermal stability and compatibility with other materials. Ceramic-based thermal management

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materials can largely meet these requirements, while offering dielectric isolation to prevent an electrical short. Due to its excellent properties in chemical stability and thermal conductivity, aluminum nitride (AIN) has been regarded as an attractive ceramic material applicable to the heat dissipation substrate for highpower or high current devices [3,4]. Today, the most commonly used substrate for the high-power light emitting diode (LED) is bulk AIN ceramics. In the LED package, the bulk AIN substrates are typically attached to a metal heatsink via a TIM, which yields a high thermal resistance between the AIN substrates and heatsink due to the low thermal conductivity of polymer-based TIM [5]. The removal of a TIM layer can give the most efficient thermal path between the AIN substrates and the heatsink.

The direct deposition of AlN thick film on a metal heatsink is the most likely way to remove the TIM layer and enhance the heat transfer to the heatsink. In particular, the thickness of AlN thick films can be also effectively reduced by the direct deposition of AlN thick film on a metal heatsink. Among the various coating techniques, plasma spraying is a well-established thermal spraying method used to produce a thick ceramic coating. However, this technique has several drawbacks such as low density, poor adhesion to substrate, and phase decomposition/thermal oxidation due to high-temperature exposure. Due to these drawbacks, it is

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difficult to obtain the high quality of AlN films which show high thermal conductivity [6].

Aerosol deposition (AD) is a type of powder spray coating method which is capable of depositing dense and strongly adherent ceramic coating layers at room temperature (RT) [7–9]. Solid particles are used as the starting materials and the coating layer is formed by the collision of highly energetic particles onto a substrate. Because AD is carried out at RT, the starting powder and the resultant coating have the same chemical composition; the coating composition can, hence, be precisely controlled by manipulating the powder composition [7,10]. Therefore, it is expected that AD can be very suitable for fabricating the AlN thick film which has minimal oxide phases such as aluminum oxide  $(Al_2O_3)$ . Actually, several studies have reported the fabrication of AlN film by AD [11,12]. However, the AD process has commonly been reported to generate high compressive stress in the film [13] and, as a result, the thickness of the aerosol-deposited AlN film cannot exceed about 10 µm without controlling the particle size distribution or the incorporation of stress-relieving polymer phases in the film [11,12]. At an AlN film thickness below 10  $\mu$ m, the film cannot be applied to a dielectric layer of high-power devices since the film thickness is too low. Polymer phases, incorporated in the film, can drastically deteriorate the thermal conductivity of AIN film, owing to their low thermal conductivity.

In this study, we fabricated high-quality AlN thick films with thicknesses up to 30  $\mu m$  on an Al substrate by AD, without a TIM layer. For comparison, AlN films with different compositions were additionally prepared: AlN-Al\_2O\_3 film and AlN-polyvinylidene fluoride (PVDF) ceramic-polymer composite film. The AlN-Al\_2O\_3 film was fabricated to investigate the effect of the formation of Al\_2O\_3 on the thermal properties in AlN films. PVDF is the most favorite material which shows the reliable fabrication of ceramic-polymer composite in AD films [14]. The physical, electrical, and thermal properties of the films deposited on Al substrate were characterized and the effect of film composition on the film characteristics was also investigated.

# 2. Material and methods

## 2.1. Preparation of AlN powders for coating

A commercially available AlN powder (Eno Material, Qinhuangdao, China; oxygen content: 0.8 wt%) with a volumetric mean diameter (d<sub>50</sub>) of approximately 3.0  $\mu$ m was used as the starting powder (d<sub>50</sub>) of approximately 3.0  $\mu$ m was used as the starting powder the coating process. The starting powder was milled using zirconia balls and 99.9% ethanol in a planetary ball mill for 4 h. The slurry was then dried and heat-treated at 500–1000 °C for 2 h under nitrogen atmosphere. Among the powders, only AlN powder heated at 800 °C was successfully used for high-quality AlN thick film, which was designated as AlN(800N<sub>2</sub>).

For AlN-Al<sub>2</sub>O<sub>3</sub> film fabrication, AlN-Al<sub>2</sub>O<sub>3</sub> powder was prepared by thermal oxidation of AlN powder. The starting AlN powder was planetary ball milled for 4 h and heat-treated at 500–1000 °C for 2 h under air atmosphere. Among the powders, the AlN powder heated at 850 °C in air [AlN(850Air)] was the most suitable for film formation. In addition, AlN-polyvinylidene fluoride (PVDF, 99.9% purity, Sigma-Aldrich Co., Milwaukee, WI, USA) ceramic-polymer composite powder was prepared for the AlN-PVDF composite film. For the deposition of this film, the as-received AlN powder itself was used without the ball milling treatment. The as-received AlN powder was mixed with PVDF powder at a volume ratio of 10% and ball-milled for 1 h in a nylon jar using zirconia balls and 99.9% ethanol. The mixed slurry was then dried in a rotary evaporator to obtain AlN-PVDF composite powder.

#### 2.2. Aerosol deposition (AD)

Ceramic thick films, including AlN [AlN(800N<sub>2</sub>)], AlN-Al<sub>2</sub>O<sub>3</sub> [AlN(850Air)], and AlN-PVDF were deposited on an aluminum substrate by AD using the corresponding powders. The substrates, which were used for the coating deposition, were commercially available 5052 aluminum (Al) plates with dimensions of 12.7 mm in diameter and 1.5 mm in thickness. Full details of the apparatus utilized in the AD system have been described elsewhere [8]. The powder was poured into the aerosol chamber and the Al plate was placed in the deposition chamber. After evacuating the chamber with a rotary vacuum pump coupled to a mechanical booster pump, nitrogen gas as a carrier gas was flowed into the aerosol chamber at a flow rate of 30 L/min. The powder was mixed with the carrier gas in the aerosol chamber and then ejected onto the Al plate through a slit-type nozzle with a  $35 \times 1 \text{ mm}^2$  rectangular opening. The film was formed over the entire surface of the Al plate by scanning the Al plate on the motorized X-Y stage. Film thickness was controlled by changing the number of scan repetitions.

#### 2.3. Characterization

The powders used for coating were characterized by a laser diffraction particle size analyzer (HELOS/BF, Sympatec GmbH, Clausthal-Zellerfeld, Germany) and transmission electron microscopy (TEM, JEM-2100F, JEOL Co., Tokyo, Japan). The crystal structure and phase composition were determined using an X-ray diffractometer (XRD, D-MAX 2200, Rigaku Co., Tokyo, Japan). Scanning electron microscopy (SEM, JSM-5800, JEOL Co., Tokyo, Japan) and TEM were used to observe the surface and cross-sectional morphologies of the films deposited on the Al substrates. The TEM sample preparation of the films was performed using a focused ion beam (FIB, AURIGA, Carl Zeiss, Germany). The density of the AlN-coated Al samples was measured by the liquid immersion method using xylene.

The electric breakdown voltages of the deposited films were measured using a withstanding voltage tester (TOS 5101, Kikusui Electronics Corp., Yokohama, Japan). The voltage was applied at a rate of 500 V/s and five samples were tested. The thermal diffusivity of the AlN-coated Al substrate was determined at room temperature by the laser flash method using a laser flash unit (LFA 457, MicroFlash, Netzsch Instruments Inc., Germany). The thermal conductivities of the AlN-coated Al samples were calculated from the thermal diffusivity, specific heat capacity, and density of the samples. Thermal transient measurement of the samples was also carried out with the Thermal Transient Tester (T3Ster, Mentor Graphics Inc., OR, USA) in connection with the DynTIM tester (Mentor Graphics Inc., OR, USA).

### 3. Results and discussion

The control of the starting powder particle size has been reported to be very important for depositing a high density ceramic thick film on a substrate using AD [15]. A critical particle size range is typically considered within which the particles can result in strongly adherent, dense ceramic thick film on a substrate. The particle size distributions of the powders are given in Fig. 1. The as-received AlN powder exhibited a broad distribution of particle sizes ranging from 0.4 to  $25 \,\mu$ m. When the AlN film was deposited on the Al plate via AD using the as-received AlN powder, a coating layer was not formed and the surface of the Al plate was damaged and eroded due to the existence of large particles with a diameter greater than 10  $\mu$ m.

One of the characteristic feature of aerosol-deposited film is

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