Investigation of thermal conductivity and dielectric properties of AlN-based ceramics with ZnO addition by impedance spectroscopy analysis

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ABSTRACT
The effect of ZnO on phase composition, microstructure, thermal and dielectric properties of AlN ceramics was studied. The X-ray diffraction indicated that ZnAl2O4 and ZnO phases were detected in samples. With 0.5 wt% ZnO added, the thermal conductivity of AlN sample increased to 193.7 W/m·K. Whereas, the thermal conductivity dropped sharply due to excess ZnO. Moreover, the addition of ZnO increased the permittivity and altered the relaxation mechanism of AlN ceramics. The fitting data and activation energy from impedance spectroscopy revealed the variation of defect mechanisms in samples. The addition of 0.5 wt% ZnO reduced the concentration of aluminum vacancy in AlN grains to improve thermal conductivity. Nevertheless, excess ZnO dissolved into the AlN lattice and created additional defects, causing the decrease in thermal conductivity and electrical resistivity for AlN ceramics. Overall, suitable addition of ZnO can effectively increase the thermal conductivity and alter the dielectric properties of AlN ceramics.

1. Introduction

Aluminum nitride (AIN) ceramics have attracted considerable attention because of their unique and excellent properties. With high thermal conductivity and high breakdown field strength, AlN ceramics are applicable to power electronic devices and packaging materials [1,2]. With high hardness, high bending strength and good thermal impact resistance, AlN ceramics are considered as the ideal material in the aerospace field [3,4]. Furthermore, as high resistivity, low permittivity and thermal expansion coefficient compatible with Si, AlN ceramics are applicable substrate materials for large scale integrated circuit [5,6].

High sintering temperature (above 1950°C) is required to pure AlN ceramics for densification due to its low self-diffusion coefficient [7]. Meanwhile, oxygen impurity in the AlN particles is detrimental to thermal conductivity. Thus, sintering additives are introduced to AlN ceramic for the reduction in sintering temperature and performance improvement. The sintering additives of AlN ceramics consist mainly of rare-earth and alkaline-earth fluorides or oxides, such as Y2O3 [8], Dy2O3 [9], Sm2 O3 [10], CeO2 [11], CaO [12], YF3 [13], and CaF2 [14]. These sintering additives can react with oxygen impurity (Al2O3) in AlN particles to form aluminates. Due to the lower melting point, aluminates could form liquid phases surrounding AlN grains at a relatively low temperature during sintering [15]. The liquid phase can wet and completely surround the solid particles. Small solid particles dissolve in the liquid phase and precipitate on the surface of large particles. Under the action of surface tension in the liquid phase, the solid particles reach a compact arrangement. And then the sintering temperature is reduced and oxygen impurities are removed.

Due to the applications of AlN-based ceramics as multilayer electronic substrates or heat sinks in power electronic devices, thermal conductivity and mechanical properties are important performances. To improve performance, various other additives have been introduced to AlN ceramics. Du [16] prepared the AlN ceramics at 1650°C with CaF2-Y2O3-Li2CO3 as additives, and Li2CO3 improved the wettability of the liquid phases. Lee [17] prepared the AlN ceramics with CaZrO3 added, and the flexural strength was greatly increased to 560 MPa. Liu [18] reported the AlN ceramics with 5 wt% Si3N4 added. The flexural strength increased from 333.3 MPa to 389.5 MPa, and fracture toughness increased from 2.4 MPa·m1/2 to 3.4 MPa·m1/2. Yun [19] reported the AlN ceramics with 1.49 vol% graphene nanosheets added, and the flexural strength and fracture toughness increased to 441 MPa and 5.09 MPa·m1/2.

The dielectric properties are also significant performances of AlN-based ceramics for high-frequency and vacuum electronic devices, such as microwave windows and attenuators. For a microwave window material, low dielectric constant and dielectric loss are...
required. For a microwave attenuation material, a high dielectric loss is required. Many works have reported the effect of different additives on thermal and mechanical properties of AlN ceramic. Whereas, research on the effect of additives on the dielectric properties is still rare. As a common direct-gap semiconductor, Zinc oxide (ZnO) has wurzite structure similar to AlN. In addition, ZnO could react with Al₂O₃ under certain condition [20,21]. Therefore, ZnO could be an effective additive to adjust thermal conductivity and dielectric properties for AlN ceramic. In this paper, AlN ceramics were synthesized with Y₂O₃-YF₃ as sintering aids. With the addition of ZnO, the thermal conductivity and dielectric properties of AlN samples presented attractive trends. Impedance spectroscopy was introduced to analyze the variation of defect mechanisms in AlN ceramics, which could effectively combine thermal conductivity with dielectric properties [22].

2. Material and methods

The AlN samples were prepared using AlN powders (JC, Toyo Aluminum, Japan) with additives of Y₂O₃ (99.99%, Sinopharm Chemical Reagent Co., China), YF₃ (99.9%, Sinopharm Chemical Reagent Co., China) and ZnO (99.9%, Aladdin, China). The main composition of materials in this work was AlN-2 wt%Y₂O₃-1 wt%YF₃-x wt%ZnO, where x = 0, 0.5, 1, 2. The raw powders were stoichiometrically weighed and milled with ZrO₂ balls in ethanol. After dried at 70°C, the mixed powders were pressed into discs, using paraffin binder. Then, the green discs were isostatic cold pressed at 140 MPa. The sintering of the samples was carried at 1700°C for 3 h with N₂ atmosphere in a graphite furnace.

The phase composition was analyzed by X-ray diffractometer (XRD-7000, Shimadzu Corp., Japan). Microstructure observations were carried out on a scanning electron microscope (SEM, EM-30Plus, COXEM Co., Korea). The grain sizes were calculated by intercept method in SEM photographs. Archimedes’ law was applied to measuring the densities. The values of thermal diffusivity were measured by a laser flash analysis method and multiplying the values of specific heat capacity, density and thermal diffusivity could obtain the thermal conductivity of the samples. The dielectric properties measurement (from 30°C to 700°C) and impedance spectroscopy (from 400°C to 600°C) were examined by an impedance analyzer (4294A, Keysight Technologies, USA) and a temperature-control system (VDMS-2000, Partulab, China).

3. Results and discussions

The XRD patterns show phase composition of AlN-2 wt%Y₂O₃-1 wt%YF₃-x wt%ZnO samples in Figure 1. The XRD photographs of all samples showed

\[
\text{ZnO} + \text{Al}_2\text{O}_3 \rightarrow \text{ZnAl}_2\text{O}_4
\]  

Figure 1. The XRD patterns of AlN-2 wt%Y₂O₃-1 wt%YF₃-x wt%ZnO ceramics sintered at 1700°C.

Figure 2 shows the SEM images on fracture surfaces of the AlN samples. All samples exhibited high density wurtzite grain structure. No pores or flaws were detected in Figure 2. The light phases were observed in the grain boundary, which were denoted as aluminate Y₃Al₅O₁₂ and ZnAl₂O₄. Most aluminate phases were concentrated at the triple junctions of the grains. Grain boundary phases increased with increasing x value, and continuous channel phases were detected along the grain boundaries at x = 2. The average grain size (AGS) and distribution curves are shown in Figure 3 for AlN samples. The ZnO-added samples showed more homogeneous distributions. With the addition of ZnO, the average grain size decreased from 4.7 μm to 3.4 μm. The increase in grain boundary phases inhibited the grain growth. Therefore, the addition of ZnO increased the grain boundary phases, decreased the grain sizes and improved the uniformity of AlN grains.

The statistics of density and thermal conductivity for all AlN samples are shown in Figure 4. All samples showed high densification, which was consistent with the microstructure shown in Figure 2. As ZnO (5.66 g/cm³) has a higher theoretical density than AlN (3.26 g/cm³), densities of the samples increased with ZnO added. For the ZnO-added AlN ceramics, the thermal
Figure 2. The fracture surface SEM of AlN samples: (a) $x = 0$, (b) $x = 0.5$, (c) $x = 1$ and (d) $x = 2$.

Figure 3. Grain size distributions of AlN samples: (a) $x = 0$, (b) $x = 0.5$, (c) $x = 1$ and (d) $x = 2$. 
conductivity showed an interesting variation tendency in Figure 4. The thermal conductivity of the ZnO-added AlN sample at $x = 0.5$ increased to 193.7 W/m·K, which was about 7% higher than the AlN sample at $x = 0$. With more ZnO added, the thermal conductivities significantly decreased at $x = 1$ and 2. From the results of XRD and SEM in Figures 1 and 2, the addition of ZnO increased the grain boundary phases and decreased the grain size of the AlN ceramics. These factors were detrimental to the improvement of thermal conductivity, which resulted in the decrease of AlN samples at $x = 1$ and 2. However, ZnO could react with oxygen impurity in AlN particles, which was a favorable factor for thermal conductivity. This might account for the increase of thermal conductivity at $x = 0.5$. To further explore the effect of ZnO on thermal conductivity, we combined it with the trend of electrical properties for the ZnO-added AlN ceramics.

Figure 5 shows the temperature dependence of dielectric properties for AlN-2 wt%Y$_2$O$_3$-1 wt%YF$_3$-x wt%ZnO ceramics. With $x$ increased from 0 to 2, the permittivity ($\varepsilon_r$) of the samples at 1 MHz increased...
from 8.75 to 9.91 at room temperature. The electron polarization in AlN ceramic was enhanced by the addition of ZnO. Both permittivity and loss showed upward trends over the measured temperature range, and two step-like peaks were detected in Figure 5. These peaks indicated the dielectric relaxation response of the defects in AlN samples. The position of loss peak at high temperature shifted to low temperature with ZnO increased. The polarization mechanism in AlN ceramic changed with the addition of ZnO. As these dielectric relaxation peaks are indistinguishable in Figure 5, the electric modulus ($M^*$) was introduced in Figure 6. $M^*$ is defined as follows [23]:

$$M^* = 1/\varepsilon^* = M' + jM''$$  \hspace{1cm} (2)

Figure 6 shows the variation of $M^*$ with temperature for AlN-2 wt%Y$_2$O$_3$-1 wt%YF$_3$-x wt%ZnO ceramics. $M''$ can clearly demonstrate the difference of dielectric relaxation mechanism in ZnO-added AlN samples. Two peaks ($P_{a1}$ and $P_{a2}$) were detected in Figure 6(a), indicating the relaxation responses of defects at AlN grains and grain boundaries, respectively. The hopping motion of defects ($\nu_a$) at grains and grain boundaries resulted in the dielectric relaxation [22,24]. Two relaxation peaks were also detected in the AlN samples at $x = 0.5$ and 1. With the increase in ZnO addition amount, the relaxation peak intensity caused by AlN grain was gradually weaker than that caused by grain boundary in Figure 6(b-c). The results of XRD and SEM indicated that the addition of ZnO changed the composition of grain boundary phases in AlN ceramics, which enhanced the dielectric relaxation. However, only one peak was detected in Figure 6(d) for the sample at $x = 2$. Compare the position of $P_d$ with other relaxation peaks, $P_d$ was identified as the relaxation responses in AlN grains. The activation energy required for the dielectric relaxation process at $P_{a1}$, $P_{b1}$, $P_{a1}$ and $P_d$ in Figure 6 was calculated in Figure 7. The logarithm of frequency ($ln f$) and the reciprocal of temperature ($1000/T$) exhibited a linear relationship, following the Arrhenius equation [37],

$$f = f_0 \exp(-E_a/k_B T)$$  \hspace{1cm} (3)

where $k_B$ is the Boltzmann constant and $E_a$ is the activation energy required for the dielectric relaxation process. The calculated $E_a$ values of $P_{a1}$, $P_{b1}$, $P_{a1}$ and $P_d$ were 1.21 eV, 1.25 eV, 1.23 eV and 1.40 eV, respectively. The $E_a$ value of $P_d$ was different from the others, indicating that the relaxation mechanisms in AlN grains of the sample at $x = 2$ was different. The excess ZnO had obvious influences on the defect mechanism in AlN grains. Therefore, the addition of ZnO showed remarkable influence on the dielectric properties of AlN ceramics.

**Figure 6.** Variation of $M^*$ with temperature for ZnO-added AlN samples: (a) $x = 0$, (b) $x = 0.5$, (c) $x = 1$ and (d) $x = 2$. 
Defects are important factors affecting the thermal conductivity and electrical properties. Impedance spectroscopy was introduced to further analyze the defect mechanism in ZnO-added AlN ceramic. Figure 8 shows impedance data for all AlN samples measured from 400°C to 600°C. The impedance data $Z'$ and $Z''$ formed compressed semicircles to characterize the electrical properties of the AlN samples. Figure 8 showed that the resistance values increased with the increase of ZnO at $x = 0.5$–1. Nevertheless, the resistance value dropped sharply at $x = 2$, even less than the sample at $x = 0$. Impedance spectroscopy could be divided to different electrical responses by a simplified circuit model shown in Figure 8(b). In this circuit model, two parallel R|CPE equivalent circuits were connected in series, which reflected the electrical responses of grains and grain boundary in AlN ceramics [25]. CPE is defined as follows:

$$Z_{\text{CPE}} = \frac{1}{[(j\omega)^n Y_0]}$$

where $Y_0$ is the value of capacitance and $n$ is an empirical exponent from 0 to 1. Based on the circuit model and impedance data in Figure 8, the fitting parameters at 500°C of all samples were calculated in Table 1. With the increase in ZnO, the capacitance values of grain boundaries increased. This was consistent with the enhancement of electron polarization shown in Figure 9. The grain boundary resistance ($R_{gb}$) of the sample at $x = 2$ was much lower than the other samples, attributed to the excess ZnO in the grain boundaries shown in Figure 1. ZnO is a semiconductor material and its resistance is lower than AlN and aluminate. The ZnO phase remaining in the grain boundary decreased the value of $R_{gb}$ for the sample at $x = 2$. Furthermore, the grain resistances ($R_g$) of the samples showed an attractive variation in Table 1. The $R_g$ of the samples at $x = 0.5$–1 were larger than the sample at $x = 0$, but the $R_g$ of the samples at $x = 2$ dropped sharply. This phenomenon indicated that the defect mechanism in AlN grains was influenced by the addition of ZnO. The values of activation energy in grains for all AlN samples were calculated in Figure 9 to analyze the effect of ZnO.

Figure 9 shows the variation of electrical conductivities ($\sigma$) with temperature for AlN grains. The $\sigma$ is dependent on temperature with Arrhenius equation [26]

$$\sigma = \sigma_0 \exp \left( -\frac{E_a}{k_B T} \right)$$

where $k_B$ is the Boltzmann constant, $\sigma_0$ is the pre-exponential factor and $E_a$ is the activation energy. The calculated values of $E_a$ for AlN grains were also added in Table 1. From Figure 9(a-c) and Table 1, the calculated values of $E_a$ for the AlN grains at $x = 0, 0.5, 1$ were 1.19 eV, 1.21 eV and 1.23 eV, respectively. It was consistent with the activation energy required for
dielectric relaxation in Figure 7. The similar $E_a$ values indicated that the defect mechanisms in grains of the samples at $x = 0$–1 were the same. As the principal impurity in AlN grains, oxygen dissolved in AlN lattice and created the main defect $V_A^O$. Thus, the defects in all AlN grains at $x = 0$–1 were $V_A^O$. From Figure 8 and Table 1, the larger values of $R_p$ at $x = 0.5$–1 indicated the lower concentrations of conductivity-enhancing $V_A^O$ [27] in AlN grains. Thus, the decrease in $V_A^O$ resulted in the weakening of the relaxation peak intensity in AlN grain for the samples at $x = 0.5$–1, as shown in Figure 6. Meanwhile, $V_A^\gamma$ is the major defect about thermal conductivity in AlN ceramic because of its large value of scattering cross section $\Gamma$ [28]. The lower concentrations of $V_A^\gamma$ resulted in the less phonon scattering in the sample at $x = 0.5$. Thereby, the thermal conductivity at $x = 0.5$ was higher than the sample without ZnO added in Figure 4. Unlike the others, the value of $E_a$ for the AlN grains at $x = 2$ was 1.58 eV in Figure 9(d). The unique $E_a$ suggested the defect mechanism in AlN grain of the sample at $x = 2$ was different with the others. The crystal structure of AlN and ZnO is both wurtzite structure in this work. Due to the similar crystal structure, ZnO could be doped into the AlN lattice [29]. In the AlN sample at $x = 2$, the excess ZnO dissolved into AlN lattice during sintering, generating additional defects in AlN grains. The reaction is as below:

$$3\text{ZnO}_2 \rightarrow 3\text{O}_3^+ + V_A^\gamma + 3\text{Zn}_2^+ + V_N$$

(6)

Therefore, the unique $E_a$ in Figure 9(d) was attributed to the additional defects created by ZnO dissolved in AlN grains. The relaxation peak $R_p$ in Figure 6(d) and the unique $E_c$ in Figure 7(d) were also caused by the additional defect mechanism in AlN grains for the sample at $x = 2$. Moreover, the defects created by the excess ZnO in grains created more phonon scattering. This also resulted in the sharply decline of thermal conductivity for the sample at $x = 2$ in Figure 4.

Consequently, the suitable addition of ZnO effectively improved the thermal conductivity and affected the dielectric properties of AlN ceramics. The addition of ZnO could further reduce oxygen impurity concentration in AlN ceramics. The decrease in $V_A^O$ improved thermal conductivity and changed the polarization mechanisms in AlN ceramics. Nevertheless, the excessive addition of ZnO had adverse effect on thermal conductivity and obviously changed dielectric properties of AlN ceramics. The excess ZnO not only remained in the grain boundary but also dissolved into the AlN lattice during sintering. It created additional defect mechanisms, resulting in the decrease in thermal conductivity and electrical resistivity for AlN ceramics.

Figure 8. Complex impedance spectra ($Z'$-$Z''$) of AlN-2 wt%Y$_2$O$_3$-1 wt%YF$_3$-x wt%ZnO samples from 100 Hz to 10 MHz measured at (a) 400°C, (b) 500°C and (c) 600°C.

| Sample | $R_p/$MΩ | $Y_p$pF | $n$ | $E_a$/eV | $R_A^0$/MΩ | $Y_A^0$pF | $n$ |
|--------|----------|---------|-----|----------|-------------|-----------|-----|
| $x = 0$ | 0.81     | 176.6   | 0.82| 1.19     | 1.79        | 352.1     | 0.80|
| $x = 0.5$ | 3.35     | 142.3   | 0.79| 1.21     | 2.22        | 1281      | 0.72|
| $x = 1$ | 5.62     | 56.8    | 0.83| 1.23     | 3.53        | 2027      | 0.73|
| $x = 2$ | 0.28     | 627.0   | 0.71| 1.58     | 0.42        | 5311      | 0.64|
4. Conclusions

AlN-2 wt%Y2O3-1 wt%YF3-x wt%ZnO (x = 0, 0.5, 1, 2) ceramics were synthesized in this work. The evolution of phase composition, microstructure, thermal and dielectric properties of AlN samples were discussed. With the addition of ZnO, ZnAl2O4 and ZnO phases were detected in AlN ceramics. As the increase in grain boundary phases, the grain sizes of ZnO-added AlN samples decreased and the distributions were more homogeneous. The thermal conductivity of the AlN sample at x = 0.5 increased to 193.7 W/m·K, but that of the samples at x = 1–2 dropped sharply. Moreover, the addition of ZnO showed a significant effect on the dielectric properties of AlN ceramics. The permittivity ($\varepsilon_r$) of the AlN ceramics increased with ZnO added, indicating the polarization was enhanced. Two relaxation peaks were detected in the samples at x = 0, 0.5 and 1, but only one peak detected in the sample at x = 2. Impedance spectroscopy and the fitting data revealed lower concentrations of $V'_\text{Al}$ in the sample at x = 0.5, resulting in the improvement of thermal conductivity. Nevertheless, the activation energy for the grain indicated that the excess ZnO dissolved into the AlN lattice and created additional defects, causing the relaxation peaks in the sample at x = 2. The excess grain boundary phases and additional defect mechanisms resulted in the decrease in thermal conductivity and electrical resistivity for AlN ceramic at x = 2. Overall, the suitable addition of ZnO can effectively improve the thermal conductivity and alter dielectric properties of AlN ceramics.

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Disclosure statement

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