High figure of merit (MgHf)Al<sub>x</sub>N thin films for miniaturizing vibrational energy harvesters

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Abstract. In this paper, we report our development of piezoelectric MgHf co-doped AlN films with high power figure of merit (FoM) for vibrational energy harvesters (VEHs). The (MgHf)Al<sub>x</sub>N films were deposited on Pt/Ti/SOI substrates by reactive ion-beam sputtering AlN and MgHf targets simultaneously. These studies revealed that we achieved a high FoM of 22.3 GPa at $x = 0.12$, increasing three-fold that of the pure AlN. Our micro-machined energy harvester exploiting the developed (MgHf)Al<sub>x</sub>N films provided the highest normalized power density (NPD) of 12.04 mW.cm<sup>−2</sup> among the state-of-the-art piezoelectric VEHs. The results opened the way for the development of high-performance VEHs using in wireless sensors network.

1. Introduction

Piezoelectric VEHs have become one of the novel approaches to self-sustainable power sources for wireless sensor networks (WSN) [1, 2]. High performance and miniaturization of VEHs continue the quest for a better piezoelectric material with higher FoM ($e_{31}^e/\varepsilon_E$).

To date, lead zirconate titanate (PZT) is the most widely used piezoelectric material for sensors and actuators [2, 3]. However, PZT is not the best material for power generation in VEHs due to the high relative permittivity ($\varepsilon_r$ of 1118 [4]), that decreases the FoM. Moreover, PZT is recently restricted to be used in society since it contains huge amount of a toxic material, lead (Pb) [3]. For such reasons, lead-free piezoelectric films such as AlN, ZnO, KNN, etc. are introduced to replace the toxic PZT materials [3-8]. Among them, AlN is one of the most promising materials for the high electromechanical-coupling factor ($k^2$ of ~ 6.5%), low dielectric loss ($\delta$ of 0.3%) and low dielectric constant ($\varepsilon_r$ of 10) [4]. In addition, the FoM for AlN is comparable to that of PZT (~ 10 GPa for AlN and ~ 13 GPa for PZT [4]).

With the aim of FoM enhancement, AlN piezoelectricity might be improved by tailoring dopants into the Al sites [3, 5-8]. In the previous studies, we proofed that co-doping Mg and Hf into Al sites enhanced approximately 3.5-time longitudinal piezoelectric coefficient ($d_{31}$) [5, 6]. Since most of piezoelectric VEHs exploit $d_{31}$-mode structure, measuring the transverse piezoelectric coefficient ($d_{31}$ or $e_{31}$), dielectric constant ($\varepsilon_r$), and Young’s modulus ($Y$) of MgHf co-doped AlN thin films become crucial.

In this study, we aimed to synthesize high-FoM (MgHf)Al<sub>x</sub>N films by reactive ion-beam sputtering with MgHf and AlN targets. We investigated extensively the impact of MgHf dopants concentration on Young’s modulus ($Y$), the transverse piezoelectric coefficient ($e_{31}$), and the FoM. Also, we developed the first VEH prototype utilizing the successful films.
2. Experimental

The (MgHf)Al$_{1-x}$N films were developed by the reactive ion-beam sputter with MgHf and AlN targets in Ar/N$_2$ ambient. The 700 nm (MgHf)$_x$Al$_{1-x}$N films with the $x$ from 0 to 0.12 were grown on Pt (100 nm)/Ti (6 nm)/SOI substrates. Base pressure of the sputtering chamber was kept at less than 1.5 x 10$^{-7}$ Torr. The AlN target was pre-sputtered before each deposition for 20 min in Ar and then for 10 min in the mixture of Ar and N$_2$. The applied RF powers for AlN and MgHf targets are 140 W and 100 W, respectively. The Ar/N$_2$ flow rate was kept at 8:32 sccm during deposition process. The detailed deposition process was presented in [6].

(MgHf)$_x$Al$_{1-x}$N thin film crystallinity was investigated and reported in our previous report [5]. The $c$- and $a$-axis lattice constants were determined from the X-ray diffractometer (Brucker-D8 advance) using $\theta$-2$\theta$ scans on the (0002) and (10$\bar{1}$2) planes in symmetric and asymmetric configurations, respectively [8]. The $Y$ of the developed films was determined from the resonant-frequency shift between (MgHf)$_x$Al$_{1-x}$N/Si and Si cantilevers [9]. The $\varepsilon_{31}$ of (MgHf)$_x$Al$_{1-x}$N was determined by measuring displacement for a unimorph cantilever under applied voltage. In this measurement, a negative polarized voltage $(|V_{pp}|=1V-20V)$ in sinusoidal waveform at frequency of 1000 Hz, being far away from the resonant frequencies, was applied on the cantilevers. The tip displacement was measured by using a Laser Doppler Vibrometer (Ono-Sokki LV-1710). To determine the FoM, the $\varepsilon$, was measured using an impedance analyzer (HP-4194A) at 10 kHz.

Figure 1 showed a scanning electron microscopic (SEM) image of the array of (MgHf)$_x$Al$_{1-x}$N/Si cantilevers with the same width of 200 $\mu$m, thickness of 50 $\mu$m and lengths of 500 $\mu$m, 1000 $\mu$m and 1500 $\mu$m. The cantilevers were fabricated with various lengths to avoid the geometrical errors.

3. Results and discussions

3.1. MgHf co-doped AlN thin film characterization

Figure 2 shows the $Y$ and the lattice constant ratio ($c/a$) depend on the MgHf concentration. The $Y$ of pure AlN decreased from 323 GPa to 305 GPa at $x=0.12$. This elastic softening is caused by the decrease in $c/a$ ratio when co-doping Mg-Hf into Al sites [10]. The $\varepsilon_{11}$ and the $\varepsilon_{31}$ in relation with the MgHf concentration were shown in figure 3. The thin films achieved $\varepsilon_{11}$ of 1.4 C/m$^2$ at $x=0.12$ that increases of 84 % compared with that of the pure AlN. The $\varepsilon_{31}$ also increased from 10.3 (for $x=0$) to 11.6 for ($x=0.12$).

The increase in the transverse piezoelectric coefficient led to the high FoM of 22.3 GPa at $x=0.12$. Figure 4 exhibits the dependence of the FoM on dopants concentration. The FoM increases three-time that of the pure AlN (from 7.2 GPa for $x=0$ to 22.3 GPa for $x=0.12$).

A comparison of FoM for (MgHf)$_x$Al$_{1-x}$N in this study with PZT and other piezoelectric materials was shown in the figure 5. It demonstrates that pure AlN has experimental FoM comparable to ZnO and PZT. However, the (MgHf)$_x$Al$_{1-x}$N provided the highest FoM at $x=0.12$ among the investigated
piezoelectric materials [2,4,7,11]. At the same dopant concentration, the FoM for (MgHf)\textsubscript{x}Al\textsubscript{1-x}N was approximately 1.9-fold that of the expensive materials of Sc-doped AlN [7]. In addition, The FoM increases three-time that of the pure AlN, being higher than that of the well-developed PZT [4].

3.2. (MgHf)\textsubscript{x}Al\textsubscript{1-x}N-based micro-energy harvesters

A micro VEH consisting of the (MgHf)\textsubscript{0.12}Al\textsubscript{0.88}N/Si cantilever of 1000 µm × 200 µm × 50 µm and the Si proof-mass of 0.6 mg was fabricated by bulk micromachining. A SEM image of the final device was shown in figure 6. The top and bottom electrodes were connected to separate terminals on a PCB plate by using a ball-wedge wire bonder (TPT-HB16) with Au wire of 25 µm in diameter.

To evaluate the device performance, the VEH was fixed on a standard shaker (G-Master APD-200FCG) and the output power spectra were measured under various accelerations as shown in figure 7. The optimal resistance for the highest output power was obtained at 800 kΩ. The device provided a maximum output power of 2.6 µW at the acceleration of 0.9 g (1 g = 10 m/s\textsuperscript{2}) and the resonance frequency of 2497 Hz. The NPD for this device was 12.04 mW.cm\textsuperscript{-3}.g\textsuperscript{-2}. This NPD increases 5.4-fold that of the best pure AlN-based VEH [12]. In addition, the achievement is the highest value among the published VEHs (figure 8).

4. Conclusion

In summary, we successfully deposited (MgHf)\textsubscript{x}Al\textsubscript{1-x}N thin films by reactive ion-beam sputtering with MgHf and AlN targets. The transverse piezoelectric properties and the Young’s modulus depending on the MgHf dopants concentration were investigated. The result showed an 84% increase in \( \varepsilon_{31} \) of (MgHf)\textsubscript{0.12}Al\textsubscript{0.88}N film leads to a threefold increase in the FoM. The VEHs utilizing (MgHf)\textsubscript{0.12}Al\textsubscript{0.88}N
with the high FoM of 22.3 GPa generated the power of 2.6 µW at the input acceleration of 0.9 g and the resonance frequency of 2497 Hz. Our device provided the highest NPD of 12.04 mW.cm\(^{-3}\).g\(^{-2}\). These results paved the way for development of high-performance (MgHf)\(_{0.12}\)Al\(_{0.88}\)N-based VEHs applying in WSN.

![Figure 7. The output power spectra of (MgHf)\(_{0.12}\)Al\(_{0.88}\)N-based VEH](image)

**Figure 7.** The output power spectra of (MgHf)\(_{0.12}\)Al\(_{0.88}\)N-based VEH

![Figure 8. The NPD of (MgHf)\(_{0.12}\)Al\(_{0.88}\)N-based VEH and the state-of-the-art piezoelectric VEHs.](image)

**Figure 8.** The NPD of (MgHf)\(_{0.12}\)Al\(_{0.88}\)N-based VEH and the state-of-the-art piezoelectric VEHs.

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