



Development of Nitride Thin Film and Micro Devices for Piezoelectric Vibration Energy Harvesters

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論文内容要約

Energy harvesters that scavenge electricity from ambient energy resources have attracted a great deal of attention as promising on-site power source for wireless sensor nodes, which will construct "*sensors communication society*" that can be indispensable infrastructure of the Internet of Things (IoT). In response to the growing social demand for IoT, extensive research has been done so far, and various types of energy harvesters have been developed. Among them, piezoelectric-based vibration energy harvesters (VEHs) have been considered as one of the most practical devices for their excellent features: high power density, high potential of size reduction, and long lifetime.

The most important constituents determining performance of the vibration energy harvesters are piezoelectric materials, often provided in the form of thin films. Lead zirconate titanate (PZT) has been the most widely used one for VEHs due to its superior piezoelectric coefficient ($d_{31} \sim 150 - 200 \text{ pm/V}$). However, its high dielectric constant ($\varepsilon_{r} \sim 1000$) restricts figure of merit (FoM) for power generation since the FoM is inversely proportional to ε_{r} . In addition, PZT contains a large amount of toxic lead (Pb) that should be avoided in commercial devices. For these reasons, lead-free piezoelectric materials with higher FoM than PZT are strongly desired.

This study aims to 1) develop lead-free piezoelectric materials with higher FoM than PZT and 2) realize micro-VEHs showing the highest power generation capability among VEHs so far developed. Furthermore, in response to rapidly growing social demand to high-performance VEHs, high-throughput combinatorial approach is adopted.

In chapter 2, I conducted high-throughput investigation of lead-free MgHf co-doped AlN ($(MgHf)_xAl_{1-x}N$) thin films that is one of the most promising piezoelectric materials showing high piezoelectric coefficient. At the beginning of this research, I focused on finding growth conditions to obtain high crystallinity AlN films using an ion beam sputtering. The conditions studied here were substrate temperature, existence of bottom electrodes, and sputtering power. Then, effect of the MgHf doping on the piezoelectric coefficient of (MgHf)_xAl_{1-x}N films was rapidly and systematically studied using combinatorial approach.

Concentration of MgHf was continuously changed and extended up to x = 0.44. A monotonic increase in longitudinal piezoelectric coefficient (d_{33}) with the dopant concentration was observed and (MgHf)_{0.44}Al_{0.56}N exhibited the highest value 13.7 pm/V, which is 3.5-fold higher than that of the pure AlN films.

In Chapter 3, I implemented high-throughput investigation of the FoM for MgHf co-doped AlN films. For this purpose, 10 cantilever-shaped micro measurement devices $(0.2 \times 1.5 \times 0.05 \text{ mm}^3)$ with $(MgHf)_xAl_{1-x}N$ films (thickness = 700 nm) were fabricated along composition gradient of the $(MgHf)_xAl_{1-x}N$ composition spread film. The measurements found that the transverse piezoelectric coefficient (d_{31}) and ε_r increased with increase in the dopant concentration; d_{31} from 2.3 pm/V to 6.8 pm/V and ε_r from 10.3 to 13.5. As a result, FoM increased significantly from 6.5 GPa for x = 0 to 31.5 GPa for x = 0.44.

In Chapter 4, I evaluated output performance of energy harvesters with $(MgHf)_xAl_{1-x}N$ films. The energy harvesters were fabricated in the same way as the cantilever-shaped devices discussed above. The output power increased with increase in the dopant concentration (thus with FoM) and the maximum value of 3.74 µW was obtained for x = 0.44 at the resonance frequency of 2422 Hz and the input acceleration of 0.9 *g*. Corresponding normalized power density (NPD) of 18.4 mW.cm⁻³.g⁻² was the highest value among thus far developed piezoelectric VEHs. This is the first time NPD of a lead-fee material based VEH can exceed the highest value provided by PZT-based devices.

In Chapter 4, I challenged to further improve performance of the $(MgHf)_xAl_{1-x}N$ -based VEHs by adding a Si/W proof-mass to the $(MgHf)_xAl_{1-x}N/Si$ beam. For x = 0.44, the NPD reached to 61.9 mW.cm⁻³.g⁻² and resonant frequency decreased to 908 Hz. The NPD value was 30-fold than that of the pure AlN-based VEHs and even 6-fold than that of the best PZT-based VEHs.

In summary, we have developed highly efficient piezoelectric (MgHf)_xAl_{1-x}N thin films that can replace PZT and improve performance of the piezoelectric VEHs. Moreover, this study established a novel way to apply high-throughput combinatorial approach to piezoelectric materials discovery and devices fabrication/evaluation. Our breakthrough achievements on (MgHf)_xAl_{1-x}N-based VEHs has opened the way for sensor nodes powered by micro VEHs, which will finally lead to the sensors communication society.