Highly Oriented and Stress Modified Thick AlN Films Deposited on Low Thermal Expansion Alloy Substrates for Flexible Electronics in Harsh Environment

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Abstract. Highly oriented and stress-modified thick aluminium nitride (AlN) films were deposited by reactive AC magnetron sputtering on 4 inch substrates and foils made of the low thermal expansion alloy “42Alloy” for flexible electronics in harsh environment. As a result of modification of film depositing conditions for reducing residual stress, a flat three micrometer thick AlN film was deposited successfully on the 50 μm thick 42Alloy foil, and its full width at half maximum of AlN(002) was 5.00 degree. And, it was confirmed that the AlN films on 42Alloy foils have piezoelectricity by measurement of a tip displacement of the cantilever-shape test structure.

1. Introduction
A piezoelectric transducer is an important electrical-mechanical converting component for micro electro mechanical systems (MEMS). Aluminum nitride (AlN) is expected to be one of the most attractive piezoelectric materials which have the practical figures of merit and relatively high electromechanical coupling for sensor applications and microgenerators. A structural layer constitution of an AlN film on a flexible substrate such as metal foils which features high fracture toughness and high operating temperature can enlarge the advanced sensor application in the field of the harsh environment including combustion pressure sensors, piezoelectric vibration energy harvesters and other flexible electronics [1-4]. In the previous study, stainless steels “SUS304” and Ni-based superalloy “Inconel600” foils were used as a flexible substrate [1-3]. The large mismatch in the coefficient of thermal expansion (CTE) between these materials and AlN causes the adverse thermal stress.

In this work, for achieving highly oriented and stress-modified thick AlN films, nickel-based alloy “42Alloy” was selected for the low CTE mismatch between the material and AlN as shown in Table 1. Also, the AlN films were deposited by reactive alternating current (AC) magnetron sputtering with tailoring Ar/N2 flow rates. 100 nm thick Ni films were adopted as the buffer layer to improve the c-axis orientation of AlN films. And the properties of films were characterized for investigating the effects of these layer constitutions.
Table 1. Comparison of coefficient of thermal expansion and mechanical properties of the materials [5]

| Material | Coefficient of thermal expansion [ppm/K] | Young modulus [GPa] | Poisson coefficient [-] |
|----------|------------------------------------------|----------------------|------------------------|
| AlN      | 4.7                                      | 330                  | 0.22                   |
| Si       | 3.9                                      | 160                  | 0.27                   |
| 42Alloy  | 4.2 at RT - 300 degC                     | 145                  | Unclear                |

2. Experimental procedure

Experimental procedures and results are described below. First of all, one micrometer thick AlN films were deposited on 0.5 mm-thick mirror polished 42Alloy and 0.525 mm-thick Si wafers as reference with or without 100 nm Ni buffer layer in order to search for optimum conditions as shown in table 2. The role of the nickel buffer layer is to improve the orientation by local epitaxial growth of AlN using Ni (111) plane. And, the reason for using a thick substrate is to avoid errors which give the evaluation of stress and crystallinity due to distortion of the substrate. These 42Alloy substrates and foils consist of 41 % nickel (Ni), 0.53 % manganese (Mn), 0.37 % cobalt (Co), 0.06 % silicon and iron (Fe). After that, we conducted experiments to reduce residual stress as shown in figure 1, and the deposition condition was applied to the 50 μm-thick mirror polished 42Alloy foils. The results of morphology of the 3μm AlN film by FE-SEM and photograph of the 4 inch wafer are shown in figure 2.

The fabrication process and characterizing flow of a piezoelectric cantilever for the measurement of the piezoelectric response is described below. First, 200 nm Au/ 100 nm Ni/ 15 nm Ti was deposited by RF-magnetron sputtering as the upper electrode. Next, X-ray diffraction (XRD) patterns and x-ray rocking curves (XRC) were measured by x-ray diffractometer (Bruker D8 ADVANCE) as indicated in figure 3 and 4. Finally, the SUS foils were diced into cantilever-shape and fixed with flexible flat wires by fixture glasses as shown in figure 5. The dimensions of a cantilever were 10 mm length and 1 mm width as diced. And the effective length was 6.5 mm with the fixture.

Piezoelectric vibration was generated by applying sine wave voltage between upper and bottom electrodes. The tip displacement as a function of applied voltage and its frequency in figure 6 and 7 was measured by the setup using laser vibrometer (Onosokki LV-1710) and digital lock-in amplifier (NF Corporation LI 5640).

Table 2. Summary of initial test of AlN deposition

| Layer constitution                          | Thickness of AlN films [nm] | FWHM of AlN(002) [deg.] | Stress of AlN films [MPa] |
|--------------------------------------------|-----------------------------|-------------------------|--------------------------|
| Mirror polished 0.525 mm Si substrate      | 999                         | 1.90                    | 174                      |
| Mirror polished 0.5 mm 42alloy substrate   | 992                         | 4.93                    | -335                     |
| 100 nm Ni/ Mirror polished 0.5 mm 42alloy substrate | 1016                      | 3.90                    | -1338                    |
Figure 1. XRD pattern of the AlN films on 42Alloy and Si substrates.

Figure 2. The photograph of the 3μm AlN film on 100 nm Ni/50μm 42Alloy foil (a) and its morphology by FE-SEM (b).

Figure 3. XRD pattern of the 3μm AlN film on 100 nm Ni/50μm 42Alloy foil.

Figure 4. X-ray rocking curves of AlN(002) of 3μm AlN film on 100 nm Ni/50μm 42Alloy foil

Figure 5. Schematic diagrams of piezoelectric cantilevers. The as-diced element (a), the cross-section with fixture glass (b).
3. Results and discussion
The improvement of orientation of AlN and increase of compressive stress by Ni buffer layer was observed as summarized in table 2. Figure 1 shows that the residual stress was controlled by the flow rate of Ar of deposition gas on the 42 alloy substrate as well as Si up to around -200 MPa as compressive stress. A flat three micrometer thick AlN film was achieved on 100 nm Ni/ mirror-polished 50 μm thick 42Alloy foil whose diameter was 4 inch. The crystals of AlN films were arranged with diameters of 50 nm to 200 nm as shown in figure 2. Figure 3 and 4 shows that the highly oriented AlN film was obtained and its full width at half maximum of AlN(002) was 5.00 degree. This value is better than the result on the nickel-based superalloy “Inconel 600” [3]. The shape of the rocking curve changed depending on the direction of XRC measurement. It is thought that there was the distribution in tilt of the (002) axis of AlN crystals. Dynamic piezoelectric response by the tip displacement as a function of the frequency of applied voltage and linear piezoelectric actuation at 400 Hz were shown in figure 6 and 7.

4. Conclusion
We deposited successfully highly oriented and stress modified thick AlN films on 100 nm Ni/ 50 μm-thick mirror polished 42Alloy foils for flexible electronics in harsh environment. In case of using this kind of alloy substrates, Ni buffer layer improves the (002) axis orientation of AlN films. And, we confirmed the piezoelectric response of these AlN films on 42Alloy using cantilever structure and measurement setup.

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