

Evaluation of Electromechanical Coupling Coefficients of Hydrothermally Synthesized (K,Na)NbO₃ Films

水熱合成(K,Na)NbO₃膜の電気機械結合係数の評価

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1. Introduction

For the development of next-generation communication systems, high-performance surface acoustic wave (SAW) filters and piezoelectric thin film resonators are required, for which piezoelectric materials with high electromechanical coupling coefficients (K^2) and low propagation loss are desired. Piezoelectric single crystals such as LiTaO₃, LiNbO₃, and quartz have been used for high-frequency devices, such as SAW filters. On the other hand, piezoelectric ceramics typified by Pb(Zr_xTi_{1-x})O₃ (PZT) have greater piezoelectricity than the above-mentioned piezoelectric single crystals and have been used in, for example, medical ultrasonic transducers, sensors, and actuators. However, since they have large acoustic loss caused by domain walls, it is difficult to apply them to high-frequency devices. In addition, since there is concern about the toxicity of Pb contained in PZT, lead-free piezoelectric materials have been actively developed.¹

(K,Na)NbO₃(KNN)-based piezoelectric material does not contain Pb and has high piezoelectricity. A hydrothermal synthesis method is attracting attention for manufacturing method. This process has potential for the low-temperature growth of lead-free materials, whereby the volatilization of elements with high vapor pressure is suppressed, such as potassium and sodium.

In this study, a thin-film resonator with a substrate (high-overtone bulk acoustic resonator: HBAR) was fabricated using KNN films prepared by a hydrothermal synthesis method, and the electromechanical coupling coefficient was evaluated. Moreover, input and output interdigital transducers (IDTs) were also fabricated on a KNN film and SAW propagation characteristics were evaluated.

2. Evaluation of coupling coefficient using HBAR

KNN films were deposited on SrTiO₃ substrates with SrRuO₃ films as the bottom electrode by a hydrothermal synthesis method. Nb₂O₅ powder was added as a niobium source to aqueous KOH-NaOH solution, and a film with a thickness h of 1.3–5.3

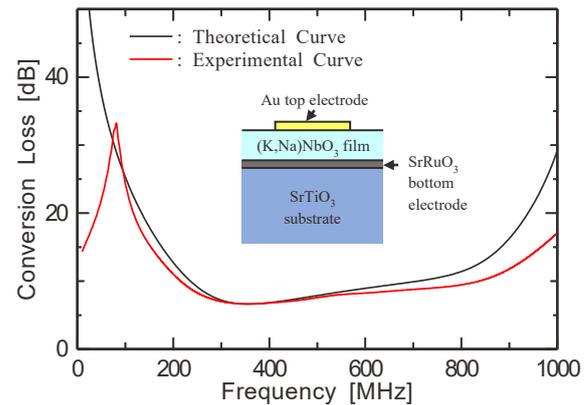


Fig. 1 Experimental and theoretical conversion losses of HBAR with amorphous Nb source, $x=0.88$, and $h=5.0 \mu\text{m}$.

μm was deposited with deposition times of 3 to 24 h at temperatures of 180 to 240 °C.² We selected two types of niobium source in this study. One is a crystalline niobium source, the other is an amorphous niobium source.² The Au thin film was vacuum-deposited on the surface of the KNN film as the top electrode, the HBAR with a top electrode/piezoelectric thin film/bottom electrode/substrate structure was fabricated. The top electrode area of the HBAR was adjusted so that the input impedance was approximately 50 Ω at the resonance frequency of longitudinal wave.

The conversion loss between the top electrode and the bottom electrode was measured using a network analyzer. When a high-frequency electric field was applied to those electrodes, a bulk wave was excited, reflected by the bottom surface of the substrate, and converted into a voltage signal by the KNN film. The insertion loss was determined by Fourier transform of the response in the time domain, and the insertion loss was multiplied by 1/2 to obtain the conversion loss.

Figure 1 shows an example of the measured curve of conversion loss for a sample with amorphous Nb source and $h=5.0 \mu\text{m}$. The electromechanical coupling coefficient K_t and phase velocity were obtained by fitting the measured curve to the theoretical curve using the Mason equivalent circuit model. Three points on each sample were measured and the average of these three was used for evaluation.

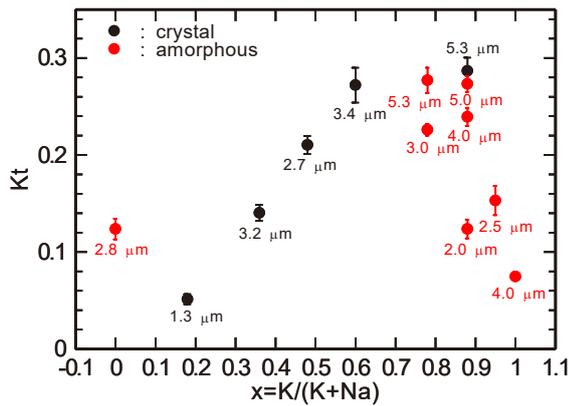


Fig. 2 Electromechanical coupling coefficient K_t evaluated for HBAR.

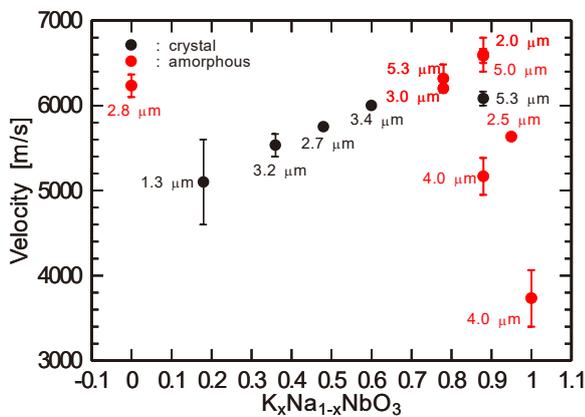


Fig. 3 Phase velocity evaluated for HBAR.

Figures 2 and 3 show K_t and phase velocity as functions of composition ratio of the solution, $x=K/(K+Na)$, for preparing KNN films, respectively. The film thickness and type of Nb source for each sample are indicated in these figures. For the crystalline Nb source, K_t increased with x , and the maximum value of 0.29 was obtained at $x=0.88$. On the other hand, when Na was not included in the film ($x=1.0$), the coupling coefficient was as small as $K_t=0.08$ for the amorphous Nb source. The phase velocity tends to increase with x and then to decrease as x approaches 1.0.

3. Evaluation of SAW propagation characteristics

With the composition ratio x fixed to 0.88 from above result, two KNN film samples, A and B, with 0.5 and 0.25 g of amorphous Nb source, respectively, and sample C with 0.25 g of amorphous Nb source without the bottom SrRuO₃ electrode film were prepared. The film thickness was in range of 15.3–16.0 μm. The input and output IDTs with a wavelength $\lambda=32$ μm, 30 finger pairs, a propagation length of $L=5\lambda$ – 50λ , and an aperture width of 50λ were fabricated using an Al-evaporated thin film.

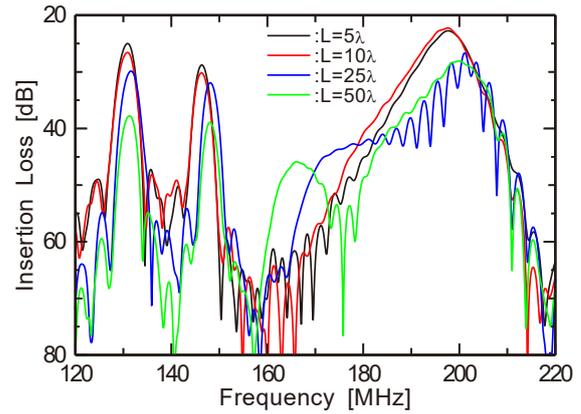


Fig. 4 Frequency response between input and output IDTs.

Figure 4 shows an example of the frequency response for sample A with the film thickness 16.0 μm. The two responses at the lower frequency among the observed three responses are considered to be the fundamental and higher modes of the Rayleigh-type SAW (R-SAW). The response at approximately 200 MHz is considered to be a bulk wave; however the details are under investigation. The propagation loss PL obtained from the slope of the minimum insertion loss as a function of L was 0.28 dB/λ for sample A. The values of PL for samples B and C were 0.27 and 0.24 dB/λ, respectively.

The electromechanical coupling coefficient K^2 of the fundamental mode was evaluated to be 0.88% for sample A from radiation admittance characteristics of the IDT. The values of K^2 for sample B and C were 0.49 and 0.59%, respectively. The above results indicate that K^2 tends to increase with the amount of Nb source. It was also observed that K^2 increased and PL was decreased by not depositing the bottom electrode.

4. Conclusions

In this study, the electromechanical coupling coefficients K_t and K^2 of KNN films prepared by a hydrothermal synthesis method using the HBAR and evaluated from SAW responses. It was clarified that K_t and phase velocity decreased at $x=1$. It was also observed that K^2 increased upon increasing the amount of Nb source, and K^2 increased when the bottom electrode was not deposited and the propagation loss decreased.

In the future, we will investigate the optimization of KNN fabrication conditions to obtain a larger electromechanical coupling coefficient and a lower propagation loss.

References

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2. A. Tateyama, *et al.*, J. Crystal Growth, **511** (2019) 1.