High-Power Piezoelectric Characteristics at Large-Amplitude Vibration of Bismuth Layer-Structured Ferroelectrics, $\text{SrBi}_2\text{Ta}_2\text{O}_9 - \text{Bi}_3\text{TaTiO}_9$ Solid Solution

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High-power piezoelectric characteristics of $\text{SrBi}_2\text{Ta}_2\text{O}_9 - \text{Bi}_3\text{TaTiO}_9$ solid solution ($\text{Sr}_{x-1}\text{Bi}_{4-x}\text{Ti}_{2-x}\text{Ta}_x\text{O}_9$, $1 \leq x \leq 2$, SBTT2- x) ceramics were investigated. Important piezoelectric constants for obtaining high vibration velocity are both piezoelectric strain constant d and mechanical quality factor Q_m . In case of bismuth layer-structured ferroelectrics (BLSFs), they have been characterized as small d value but high Q_m . In this study, High-power piezoelectric characteristics of SBTT2- x with high Q_m more than 8000 in (33) vibration mode were measured using the Electrical Transient Response Measurement. At large-amplitude vibration, the Q_m of these samples did not decrease with increasing $v_{0,p} < 2.5$ m/s.

Key words: Bismuth layer-structured ferroelectrics, Lead-free, Piezoelectric properties, High-power piezoelectric characteristics.

1. INTRODUCTION

Recently, many high-power piezoelectric ceramic devices, such as ultrasonic motors and piezoelectric actuators, have been developed. Hard $\text{Pb}(\text{Zr,Ti})\text{O}_3$ [PZT] ceramics or $\text{Pb}(\text{Mn}_{1/3}\text{Nb}_{2/3})\text{O}_3$ - $\text{Pb}(\text{Zr,Ti})\text{O}_3$ [PMN-PZT] [1-3] ceramics with a high mechanical quality factor, Q_m , are often used in high-power piezoelectric applications. However, PZT ceramics contain a large amount of Pb. Therefore, lead-free piezoelectric materials for replacing PZT have recently been demanded from the viewpoint of environmental protection. At present, many lead-free piezoelectric ceramics such as potassium niobate-based materials [4-7], $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ -based materials [8-10], bismuth layer-structured ferroelectrics (BLSF) [11-14] have been extensively investigated and their ultrasonic devices have been demonstrated [16-21]. The family of BLSFs is attracting attention as a lead-free piezoelectric material for resonator, filter, sensor, and high-power applications. The general crystal structure of BLSFs is $(\text{Bi}_2\text{O}_2)^{2+}(\text{A}_{m-1}\text{B}_m\text{O}_{3m+1})^{2-}$, where A represents Bi^{3+} , Pb^{2+} , Sr^{2+} , Ca^{2+} or other cations at the 12-coordinated site; B represents Fe^{3+} , Ti^{3+} , V^{5+} , Ta^{5+} , W^{6+} or other cations at the 6-coordinated site; and m is an integer corresponding to the number of BO_6 octahedra in pseudo perovskite blocks. In the crystal structure of BLSFs, perovskite blocks, $(\text{A}_{m-1}\text{B}_m\text{O}_{3m+1})^{2-}$, are interleaved with $(\text{Bi}_2\text{O}_2)^{2+}$ layers along the c-axis, and ferroelectricity arises mainly in the perovskite blocks. Recently, the high-power piezoelectric characteristics of some BLSF ceramics have been reported [11-14]. In these reports, the $v_{0,p}$ of the (33) vibration mode for textured BLSFs such as $\text{SrBi}_2\text{Nb}_2\text{O}_9$ and $\text{CaBi}_4\text{Ti}_4\text{O}_{15}$ ceramics linearly reached approximately 2.5 m/s without large resonance frequency change. One of the reasons for these results is that these BLSFs have both high Q_m and large electromechanical coupling factor k_{33} . Thus, they seem

to be good candidates for high-power ceramic devices realizing large amplitude and a linear stability for vibration velocity. It is speculated that resonant vibration of BLSF piezoelectric ceramics are affected by their Q_m performances. In this study, we focused on lead-free BLSF ceramics of $\text{SrBi}_2\text{Ta}_2\text{O}_9 - \text{Bi}_3\text{TaTiO}_9$ solid solution. These ceramics have ultra high Q_m above 5000 much larger than the value of PZT ceramics. It is expected the high value of Q_m contribute vibration velocity at high-power driving. And the performance of high Q_m materials for high-power applications were studied.

2. EXPERIMENTAL PROCEDURE

Ceramic samples of $\text{SrBi}_2\text{Ta}_2\text{O}_9 - \text{Bi}_3\text{TaTiO}_9$ solid solution ceramics ($\text{Sr}_{x-1}\text{Bi}_{4-x}\text{Ti}_{2-x}\text{Ta}_x\text{O}_9$, $1 \leq x \leq 2$, SBTT2- x) ($x=1.0\sim 2.0$) (abbreviated as SBTT2- x) lead-free ceramics were prepared by a conventional solid-state reaction. [15] Especially, $x = 1.250$, 1.375 and 1.500 composition were studied because of their high Q_m . In addition, to compare other piezoelectric compositions, PZT-based ceramics were prepared.

The starting raw materials were Bi_2O_3 of 99.99% purity, and, SrCO_3 , TiO_2 and Ta_2O_5 of 99.9% purity. They were mixed by ball milling for 10 h and calcined at 850 °C for 2 h. After calcination, the ground and ball-milled powders were pressed into disks 20 mm in diameter. The disks were processed in a cold isostatic press (CIP) at 150 MPa to obtain dense ceramics. After the treatment, the disks were sintered at 1180-1220 °C for 2 h in air. The sintered ceramics were cut and polished into rectangular specimens of $2 \times 2 \times 5$ mm³ for the (33) mode. Electrodes were made with fired-on Ag paste for measurements of electrical properties such as dielectric, ferroelectric, and piezoelectric properties. Then, the samples were poled in silicone oil bath heated at

200-300 °C for BLSF ceramics and 120 °C for PZT-based ceramics. A DC electric field of 5.0-8.0 kV/mm for lead-free compositions and 1.0-3.0 kV/mm for PZT ceramics were applied to the samples for 5-10 min during the poling process. Piezoelectric properties at a small amplitude were measured by the resonance-antiresonance method on the basis of IEEE standards, using an impedance analyzer (HP 4294A). High-power characteristics were measured using (33) mode samples. A piezoelectric transducer was driven by a function generator (NF WF1943A) and a power amplifier (NF HAS4052). The vibration velocity v_{0-p} was measured using a laser Doppler vibrometer (Ono Sokki LV1710) with an oscilloscope (Tektronix TDS3054B). The mechanical quality factor Q_m at a large amplitude was measured using an electrical transient response of burst voltage, which was developed by Umeda *et al.* [19]. The vibration velocity v_{0-p} of the high-power piezoelectric driving was determined by driving under the condition of constant voltage at resonant frequency that observed maximum current by tracing the frequency. To measure continuous properties, 5 minutes driving of each applied electric field were held for becoming a stationary state of the piezoelectric transducers. In the measurement of sample temperature under the continuous driving, optical thermometer (HIOKI Temperature Hightester 3445) was used.

3. RESULTS AND DISCUSSION

The crystal structure of the obtained ceramics was confirmed by X-ray diffraction analysis to show a single phase of the bismuth layer structure with $m=2$ for SBTT2- x . The relative densities of the sintered ceramics were all higher than 95% for each sample, as measured by the Archimedes method. The optimum sintering temperature for each composition was determined to yield the highest relative density.

Table I shows the dielectric and piezoelectric properties of (33) mode samples for SBTT2- x and PZT-based ceramics. All prepared samples were poled sufficiently indicated by their high $\theta_{\max}$ to determine their piezoelectric properties. The SBTT2-1.250 indicated a low k_{33} of 0.159, dielectric loss, $\tan\delta$ at 5 kHz of 0.25% and a high Q_m of 7800. Focusing on the PZT-based ceramics, they have so excellent characteristics of k_{33} , Q_m and $\tan\delta$ at 5 kHz of 0.720, 1000 and 0.33%, respectively.

Figure 1 shows the applied field E_a dependences of the vibration velocity v_{0-p} for the SBTT2- x and PZT-based ceramics under the long time driving. It is found that the v_{0-p} of SBTT2-1.250 linearly increased up to approximately 2.0 m/s at 6 V/mm. On the contrary, PZT ceramics were less than 1.0 m/s under 10 V/mm for both measurements. These results are reasonable because SBTT2- x ceramics have high Q_m . It is expected that high Q_m contributes to v_{0-p} properties at high-power piezoelectric driving. In the case of PZT-based ceramics, at the small electric field drive condition, PZT-based ceramics has a large slope of v_{0-p} as compared with SBTT2- x ceramics. However, the resonant vibration of PZT-based ceramics disrupts the stability of vibration velocity, and vibration velocity does not increase with applied electric field [11]. So, these materials are generally used at a vibration velocity below 1.0 m/s in

the practical uses.

Figure 2 shows the v_{0-p} dependences of resonant frequency change for the SBTT2- x and PZT-based ceramics. In the case of SBTT2- x ceramics, the resonant frequency shifted slightly over the v_{0-p} of 2.0 m/s. In the case of PZT-based ceramics, the transducers have a large shift of the resonant frequency as compared with those of SBTT2- x ceramics. Speculating the reason of these differences, it is expected that the temperature changing of piezoelectric transducers were affected to the sample stiffness.

Figure 3 shows the sample surface temperature dependences of vibration velocity for SBTT2- x and PZT-based ceramics. It is indicated that SBTT2- x has small temperature change. Their small temperature changes contribute to the small resonance frequency shift. On the contrary, in case of PZT, the transducers generated heat and raised temperature on their surfaces. Additionally, it is expected that temperature in samples is higher than on their surfaces and the resonant frequency were shifted furthermore. Anyhow, it is expected that the resonant frequency shift for long time driving shown in figure 2 are affected by their temperature rises. However, it is expected that the resonance frequency change were not only determined by temperature rising of samples.

Figure 4 shows the dissipation power density dependence of vibration velocity for SBTT2- x and PZT-based ceramics. SBTT2- x can generate large vibration by using small power density as compared with PZT-based ceramics. At this point, it is expected that most of the input power into the samples finally goes to heat energy for long time drive conditions. As power supply was turned on, the electrical power into transducers mainly are converted to vibration and dielectric loss energy. However, for long time driven, vibration power generated as electric power on is decay and finally goes to heat generation, in spite of the case of driving the high Q_m samples. So, most of the supplied power energy into samples are changed to heat energy from the mechanical and electrical loss.

Figure 5 shows the variations in Q_m as a function of v_{0-p} for SBTT2-1.250 and PZT-based ceramics. In case of the SBTT2-1.250 ceramics, there is small change of Q_m with increasing v_{0-p} . Other SBTT2 compositions ($x=1.375$ and 1.500) also showed similar tendency in Q_m with increasing v_{0-p} . However, in the case of PZT-based ceramics, their Q_m s decrease drastically as increasing v_{0-p} . It is expected that the degradation of Q_m were

Table I Dielectric and piezoelectric properties of (33) mode samples for SBTT2- x and PZT-based ceramics

	$x=1.250$	$x=1.375$	$x=1.500$	PZT-based
$\varepsilon_{33}^T/\varepsilon_0$	119	106	127	1400
k_{33} [%]	15.9	15.1	16.5	72.0
d_{33} [pC/N]	16.6	15.0	17.7	337.1
Q_m	7800	6650	4980	1000
N_{33} [Hz·m]	1950	1910	1940	1970
$Z_{\min}$ [Ω]	3640	5780	4180	90
$\tan\delta$ [%]	0.25	0.26	0.29	0.33
$\theta_{\max}$ [deg.]	88.6	87.5	87.6	89.7

Figure 1 Vibration velocity dependences of applied field for SBTT2- x and PZT-based ceramics.

Figure 2 Resonant frequency change dependences of vibration velocity for SBTT2- x and PZT-based ceramics.

Figure 3 Surface temperature dependences of vibration velocity for SBTT2- x and PZT-based ceramics.

Figure 4 Dissipation power density dependence of vibration velocity for SBTT2- x and PZT-based ceramics.

Figure 5 Mechanical quality factors dependence of vibration velocity for SBTT2-1.250 and PZT-based ceramics.

Figure 6 Force factors dependence of vibration velocity for SBTT2- x and PZT-based ceramics

concerned with the coercive field, E_c of each sample. The small decrease in Q_m under a large-amplitude vibration for SBTT2- x ceramics is attributed to the high E_c , which is related to the suppression of domain wall motion. Nagata *et al.* reported the E_c of SBTT2- x ceramics indicated by measurement of P - E hysteresis loop. [15] The E_c of SBTT2- x ceramics were estimated to be larger than 70 kV/cm, which is larger than that of PZT-based ceramics. It is speculated that a large coercive field contributes the stabilities such as decline slope of Q_m and the magnitude of mechanical loss at small and large amplitude vibration.

Figure 6 shows the force factor A measured by electric transient response as a function of vibration velocity for SBTT2-1.250 and PZT-based ceramics. The A of SBTT2-1.250 is very small as compared with PZT-based ceramics. Also, the compositions of $x=1.375$ and 1.500 in SBTT2- x ceramics showed similar A values to those of $x=1.250$. It is indicated that the vibration properties of SBTT2- x has large strain with small force because of their small input electrical power. For more improvement about the force factor on SBTT2 ceramics, it is expected that small impedance at resonance frequency were required to generate the force of a piezoelectric strain such as PZT-based ceramics as shown in Table I.

Finally, Table II summarizes the high-power piezoelectric properties at continuous driving around 0.6 m/s for SBTT2- x and PZT-based ceramics. SBTT2- x ceramics can reach about 0.6 m/s of v_{0-p} by small dissipation power density with small change in resonance frequency, temperature and Q_m as compared with other ceramics.

Table II High-power piezoelectric properties at continuous driving around 0.6 m/s for SBTT2- x and PZT-based ceramics

Parameter	Composition $x=1.250$ 0.87 m/s	$x=1.375$ 0.55 m/s	$x=1.500$ 0.79 m/s	PZT-based 0.63 m/s
Frequency Change [%]	-0.023	-0.016	-0.018	-1.037
Dissipation power density [mW/mm ³]	0.48	0.32	0.48	2.74
Temperature [°C] (R.T. + ~ °C)	+1.2	+0.9	+2.1	+21.8
Mechanical quality factor, Q_m	6560	6370	4920	400
Force factor, A [N/V]	0.0024	0.0022	0.0028	0.033

4. CONCLUSIONS

High-power piezoelectric characteristics at continuous driving were studied in the bismuth layer-structured ferroelectric $\text{SrBi}_2\text{Ta}_2\text{O}_9 - \text{Bi}_3\text{TaTiO}_9$ solid solution ($\text{Sr}_{x-1}\text{Bi}_{4-x}\text{Ti}_{2-x}\text{Ta}_x\text{O}_9$, $1 \leq x \leq 2$, abbreviated as SBTT2- x). SBTT2- x ceramics has the stability of frequency and temperature change because of their small dielectric loss and large coercive field above 70 kV/cm. It is expected that this coercive field contributed domain wall stability such as Q_m and will affect the stability at high-power piezoelectric driving for the long time conditions. The (33) mode vibration of the SBTT2- x ceramics was stable at a high vibration velocity of 2.0 m/s, and the resonant frequency was almost stable with increasing vibration velocity. This is the advantage for high-power applications using at limited conditions, e.g.

high temperature. Therefore, SBTT2- x ceramics is a promising candidate for lead-free high-power applications such as ultrasonic transducers.

5. ACKNOWLEDGEMENT

The authors would like to thank Professor Mikio Umeda of Nagaoka National College of Technology for his technical support and discussion on high-power measurements.

6. REFERENCES

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(Received March 7, 2011; Accepted August 9, 2011)