# Elastic Behaviour of Multilayer Piezoceramic $BaTi_{1-x}Sn_xO_3$ in the Lower MHz Region

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# Abstract

Small plates of piezoceramic  $BaTi_{1-x}Sn_xO_3$  with double layer structure were sintered. The Sn concentration in the layers was nearly uniform with the x-values of 0 or 0.1. The plates were poled and bonded to steel and brass with epoxy resin. The pulse and continous wave (cw) responses of these mechanical systems were examined and compared to a uniform barium titanate ceramic plate. The reflected ultrasound pulse signals for the layered transducers had a greater bandwith than the uniform barium titanate ceramic plate. The results of the bonded thickness vibrators in the pulse and cw operation mode are discussed. © 1999 Elsevier Science Limited. All rights reserved

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# 1 Introduction

Piezoelectric transducers are important for ultrasonic devices in nondestructive testing and other applications. Multilayer piezoceramic plates can be used as ultrasonic transducers. A bandwith enlargement should be possible if they are properly designed.

## 2 Experimental Procedure—Sample Preparation

The ceramic powders with a nominal composition of  $BaTi_{1-x}Sn_xO_3 + 1 \mod\%$  TiO<sub>2</sub> (x = 0, 0.10) were produced by the classical mixed-oxide technique. After mixing and calcining (1100°C, 2 h) of appropriate amounts of BaCO<sub>3</sub> (Leuchtstoffwerk Breitungen GmbH, no. 3018), TiO<sub>2</sub> (Merck, no. 808) and SnO<sub>2</sub> (Alfa Johnson Matthey, no. 87779), the powders were press-granulated using poly(vinyl alcohole) and successively uniaxially pressed into the same press mould  $(25 \times 16 \text{ mm}^2)$  for creating the desired layered structures. Then the samples were sintered at 1350 or 1400°C for 1 h. The samples exhibit a coarse-grained microstructure (average grain size 80  $\mu$ m). To obtain fine-grained material (average grain size 1  $\mu$ m) a sintering temperature of 1300°C (2 h) was chosen. The sintering was performed with a heating and cooling rate of 10 K min<sup>-1</sup> in both cases.

The samples for the investigations have rectangular plate shapes with a thickness of approximately 1 mm and lateral dimensions from 7 to 11 mm. The surfaces are ground carefully to have a parallel plate. Electrodes of aluminium are evaporated on both plate surfaces. The samples were poled with an electric field of  $2 \text{ kV mm}^{-1}$  field strength.

## 3 Experimental Procedure—Measurement Setup

The samples were used as transducers both in the pulse excitation and in the continous wave (cw) excitation mode. The first method is similar to a formerly described method.<sup>1</sup> The transducer samples are bonded to a steel parallelepiped with the silver epoxy 3021A (Epoxy Products, Fürth, Germany). This epoxy contains very fine silver particles and is suitable to provide also an electric conduction to the steel. This steel block is connected to the ground potential. An effective pulse voltage of approximately 2 volt was applied to the other side of transducer. The transducer works also as sensing element for the reflected ultrasound and is therefore connected to a broadband amplifier followed by a digital oscilloscope. The steel block thickness and hence the sound path length is 30 mm. The radio frequency (rf) pulses are generated by an arbitrary waveform generator 8550 from Kontron. It is switched to the burst mode and

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produces a burst of five rf sinusoidal vibrations with a pulse repetition rate of 1 kHz. The frequency of the rf vibrations is varied from 1.7 to 3.2 MHzand the amplitudes of the corresponding first echo pulses are recorded.

A brass wedge body was coated with a sound absorbing back material made from conventional hardened epoxy filled with tungsten powder of  $5\,\mu$ m particle size for the cw measurement. The transducer is coupled to the uncoated end of the brass wedge with the same epoxy like in the pulse measurement. The ultrasound generated by the



Fig. 1. Measurement setup for the continous wave ultrasound investigations.

transducer should now be strongly damped from the back material in this arrangement. The cw response of this transducer is examined with an impedance analyzer hp 4192A equipped with the test fixture hp 16047C. Figure 1 displays the described setup.

Brass and steel are used as sound transmission media because of their similar acoustic impedances compared to the ceramic transducers. The used silver epoxy bonding agent has also an rather high acoustic impedance and gives a good acoustic matching.

# 4 Results

We could not find remarkable differences in the behaviour of fine grained or coarse grained ceramics. Thus we show the results for the fine grained ceramics as an example. Both transducers had lateral dimensions of 6 and 7 mm and 1 mm thickness. The results for the pulse measurements are found in Fig. 2. The highest amplitude at the resonance was measured for pure BaTiO<sub>3</sub> ceramics. The result for the layered sample depends on the position of the layers relatively to the bond. The maximum amplitude decreases by 15% but at low frequencies the amplitude is still higher than for pure barium titanate if the side containing Sn points to the bond. Only approximately the half amplitude is measured if the side with Sn is apart from the bond. Thus the enhanced bandwith at a fairly high amplitude for the second case could be interesting for practical purposes.



Fig. 2. Amplitude of the first reflected echo pulse from the back side of a steel block of 30 mm thickness in dependence from the exciting frequency.



**Fig. 3.** Complex dielectric permittivity in dependence from the exciting frequency for a single-layer sample of BaTiO<sub>3</sub> ceramics bonded to a brass wedge with backing.



Fig. 4. Complex dielectric permittivity in dependence from the exciting frequency for a two-layer sample bonded to a brass wedge with backing; side with Sn near the bonding.

Figures 3–5 contain the values calculated from the cw measurements. We have chosen the complex clamped dielectric constant  $\varepsilon$  to avoid sample geometry dependent impedance notations. The value  $\varepsilon'$  denotes the real part of  $\varepsilon$  and is connected with the capacitive sample current. The imaginary part  $\varepsilon''$  describes the losses. A sharp resonance and relatively low losses are found for the pure barium titanate transducer in Fig. 3. The resonance becomes broader and the damping increases slightly for the layered sample with the Sn near the bond (Fig. 4). A medium resonance but high losses are found for the last case with Sn apart from the bond (Fig. 5). This is



Fig. 5. Complex dielectric permittivity in dependence from the exciting frequency for a two-layer sample bonded to a brass wedge with backing; side with Sn apart from the bonding.

in accordance to the findings with the pulse measurements because high losses correspond to low pulse echo amplitudes.

### **5** Conclusions

A good explanation for the reported effects is still not possible yet. The piezomodul of poled pure BaTiO<sub>3</sub> ceramics is greater than that of BaTi<sub>0.9</sub>Sn<sub>0.1</sub>O<sub>3</sub> ceramics.<sup>2</sup> The strongest gradient of the piezomodul is possible with BaTiO<sub>3</sub> at the 'air side' of the transducer arrangement. The lower gradient of the piezomodul of the BaTi<sub>0.9</sub>Sn<sub>0.1</sub>O<sub>3</sub> 'bond side' brings this arrangement nearer to an ideal one-gradient transducer realized partly in the ultrasound generation in a quartz rod using a microwave cavity.<sup>3</sup> Such a one-gradient transducer should have a greater bandwith because it has no natural resonances if it is perfectly acoustically matched to the sound propagation medium. We could not find any calculations of such a gradient transducer but some interesting hints.4-6 The transducers and the bonding material should be matched acoustically to the sound propagation medium. More data on the material parameters like piezomodules, densities, elastic stiffnesses and their spatial distribution are necessary for a better description of these transducers. Measurements of triple layer transducers are now in progress. A continous variation of material properties should be possible with a temperature gradient during

sintering with a special microwave sintering technique. This could lead to new types of ultrasound gradient transducers. The combination of layered or gradient transducers with backing opens another field of optimization of piezoelectric transducers.

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