

## Transducer elements made from piezofilms of PVDF and BaTiO<sub>3</sub>/PVDF.

**Wanida Sumethagulwat** B.Sc (Physics)

Department of Physics, Faculty of Science, Prince of Songkla University

**Boonlua Pongdara** M.S. (Physics)

Department of Physics, Faculty of Science, Prince of Songkla University

**Supasarote Muensit** Ph.D (Materials Physics)

Department of Physics, Faculty of Science, Prince of Songkla University

**Keywords** : piezoelectric, dielectric, composite, sensor

### Abstract

Very thin sheets of piezoelectric materials of two types : polyvinylidene fluoride polymer (PVDF) and BaTiO<sub>3</sub> powders embeded in the matrix of PVDF were fabricated using a conventional tape casting method. Microstructure and physical properties of the materials were studied. The investigations included elastic modulus measurement , dielectric permittivity and coupling factor by means of the resonance method. The experimental results obtained indicated that the materials are attractive for a soft piezoelectric transducer material which are environmentally friendly, easily to fabricate over large area and relatively inexpensive .

### Introduction

PVDF has number of attractive properties due to its low weight, formability into thin sheets with relatively high elastic modulus. (Kawai, 1969 ; Lang, 1971), Among the perovskite ceramics, barium tita-

nate (BaTiO<sub>3</sub> , BT) is a very attractive piezoelectric material for a large area of applications such as nonvolatile memories, surface acoustic wave devices, tunable capacitors, pyroelectric detectors. This work fabricated two types of samples , PVDF and the com-

bination of BT and PVDF which was the so-called BaTiO<sub>3</sub>/PVDF composite. The latter was introduced in order to extend the range of material properties of BaTiO<sub>3</sub> ceramic which is brittle in nature. Measurements were made for the composite in comparison with PVDF. However, both materials have been demonstrated that they are attractive for the use as a soft piezoelectric transducer material or piezofilm which are suitable for converting the mechanical energy to electrical power.

### Experimental procedure and equations

The steps in the preparation of PVDF sample were as followed : PVDF powders (Fluka 81432) were dissolved in 1-methyl-2-pyrrolidone (NMP) (Fluka 69120) and a PVDF solution was obtained. Parts of the solution were taken as the matrix of the BaTiO<sub>3</sub> powders (Fluka 11729). The BT-PVDF mixture was homogeneously stirred by magnetic stirrer, slowly warmed until it became viscous and then agitated in an ultrasonic bath for an hour to ensure that the ceramic particles were distributed evenly in the polymer solution. The BT/PVDF with 0.3 volume fraction of BT was obtained. The PVDF solution and the mixture of composite were made into very thin sheets between 0.12 – 0.15 mm in thickness in a similar procedure. Each of them was casted on a glass plate before annealing at 120 °C for 6 hrs. The density  $\rho$  of the sample was calculated using an equation :

$$\rho = \phi \rho^C + (1 - \phi)^P \quad (1)$$

where  $\phi$  is the ceramic volume fraction, and the superscripts P and C refer to polymer and ceramic respectively (Lang and Das-Gupta, 2000). The heat capacity of the sample, which reflects the ability of the

material to store heat, was analyzed using a Differential Scanning Calorimeter (Perkin Elmer, DSC7). The connectivity and microstructure of the composite were investigated by using SEM (JEOL JSM5800LV). Young's modulus of the samples was measured by a Shimadzu Testing Machine.

In order to make a transducer element of 10 mm x 10 mm in crosssectional area which can be handling without the glass substrate, the four layers of each type of samples were bonded together with an epoxy resin. A thin layer of metallization (silver paste) is applied to both sides of the sheet to collect the charge and serve as electrical connections. In order to polarize the samples, the corona poling field with a dc voltage of 4 kV was applied to each samples at room temperature (25 °C) for 20 min. The dielectric constant  $\epsilon_r$  of the samples is related to the capacitance C, thickness t, area A and permittivity of vacuum  $\epsilon_0$  as :

$$\epsilon_r = \frac{tC}{\epsilon_0 A} \quad (2)$$

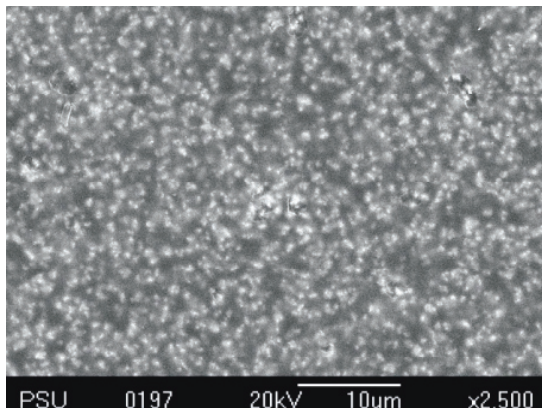
The capacitance of the samples were measured by a LCR meter (HP 4263B) and taken into equation (2) for a calculation of the dielectric constant. In the resonance measurement, each sample was applied an ac voltage by signal generator (DS 345) to excite the thickness vibration and an output voltage was recorded as a function of frequency. The main equation used for evaluation of the thickness coupling factor  $k_t$  of the sample is (Mason and Jeffe, 1954) :

$$k_t^2 = \frac{\pi}{2} \frac{f_r}{f_a} \tan \left( \frac{\pi}{2} \left( \frac{f_a - f_r}{f_a} \right) \right) \quad (3)$$

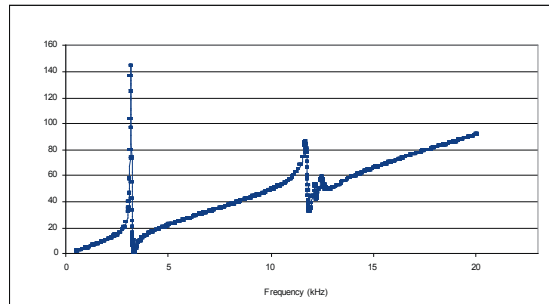
Where  $f_r$  and  $f_a$  are the resonance and an antiresonance frequencies, respectively.

## Results and discussion

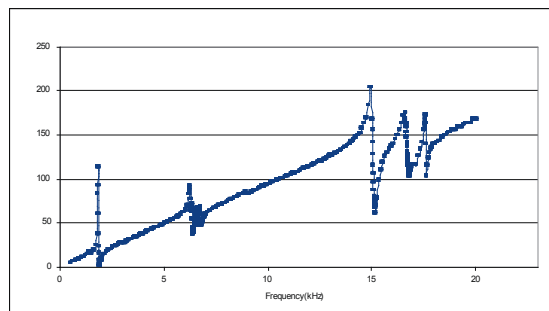
Measured values of the density, heat capacity and Young's modulus for the PVDF were respectively  $2.7 \times 10^3 \text{ kg/m}^3$ ,  $120 \text{ J/Kg } ^\circ\text{C}$  and  $6 \text{ MPa}$ . Using equation (1), the measured density of PVDF and the value of  $4.6 \times 10^3 \text{ kg/m}^3$  for BT (commercial data), the composite with  $\phi = 0.3$  has its calculated density of  $3.21 \times 10^3 \text{ kg/m}^3$ . The measured heat capacity of the composite was  $753 \text{ J/Kg } ^\circ\text{C}$ , which was relatively large. This is related to the elastic modulus of the polymer which is much higher than that of the ceramic. The ceramic powders were well distributed in the polymer matrix as seen from the SEM image in Fig.1. Values of the dielectric constant measured at 1 kHz for PVDF, BT and BT/PVDF were respectively 8, 267 and 12. This indicated that the low permittivity of the BT/PVDF was influenced by the PVDF polymer. The results from the resonance measurement were shown in Figs.2 and 3.



**Fig. 1** SEM image of the BT/PVDF composite with 0.3 volume fraction of ceramic.



**Fig. 2** Variation of the amplitude of the output voltage as a function of frequency for  $0.15 \pm 0.05 \text{ mm}$  PVDF transducer elements.



**Fig. 3** Variation of the amplitude of the output voltage as a function of frequency for  $0.15 \pm 0.05 \text{ mm}$  BT/PVDF transducer elements.

Under the applied field at frequencies below 5 kHz, the PVDF transducer element has the corresponding coupling factor of 0.33 whereas it is 0.35 for the BT/PVDF element. These values are comparable to that of commercial elements. At higher frequencies, this value was approaching to zero for the first while it maintained about 0.1 for the latter.

## Conclusions

The piezofilms made from PVDF and BT/PVDF have good physical properties, good coupling factor with good flexibility. The low-frequency coupling factor value suggests that these transducer element materials are suitable for applications as acoustic transducer (Hilczer *et al.*, 2000). The acoustic impedances of the material and related properties were addressed in future work.

## Acknowledgement

This work was financially supported by Graduate School of Prince of Songkla University, Hat Yai Thailand.

## References

- Kawai, H. 1969. The Piezoelectricity of Polyvinylidene Fluoride, Jpn.J. Appl. Phys.,8 :975-976.
- Lang, S. B. 1971. Ferroelectrics, Phys. Rev., B 4 : 3603.
- Lang, S. B. and Das-Gupta, D. K. 2000. Pyroelectricity, Fundamentals and Application : Ferroelectrics Review, 2 : 217-354.
- Mason W. P., Fellow., IRE. and Jaffe H 1954. Methods for measuring piezoelectric elastic, and dielectric coefficients of crystals and ceramic., Proc. of the IRE. p.921-932.
- Hilczer, B., Kulek, J., Markiewicz, E., and Szczesniak.2000. The Method of Matching Resonance Frequencies in Coupled Transmitter PVDF/TrFE Diaphragms. IEEE TDEI 7 (4) 498-502.