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# Piezoelectric properties of the new generation active matrix hybrid (micro-nano) composites

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#### ABSTRACT

A hybrid piezoelectric composite structure is obtained by addition of nano-sized BaTiO<sub>3</sub>, SiO<sub>2</sub> to the micro-sized PZT and polymers composition. Although the PZT material itself has excellent piezoelectric properties, PZT-based composite variety is limited. Piezoelectric properties of PZT materials can be varied with an acceptor or a donor added to the material. In addition, varieties of PZT-based sensors can be increased with doping polymers which have physical-mechanical, electrophysical, thermophysical and photoelectrical properties. The active matrix hybrid structure occurs when bringing together the unique piezoelectric properties of micro-sized PZT with electron trapping properties of nano-sized insulators (BaTiO<sub>3</sub> or SiO<sub>2</sub>), and their piezoelectric, mechanic and electromechanic properties significantly change. In this study, the relationship between the piezoelectric constant and the coupling factor values of microstructure (PZT-PVDF) and the hybrid structure (PZT-PVDF-BaTiO<sub>3</sub>) composite are compared. The *d*<sub>33</sub> value and the coupling factor of the hybrid structure have shown an average of 54 and 62% increase according to microstructure (PZT-HDPE-SiO<sub>2</sub>) have exhibited about 68 and 52% increase according to microstructure composite, respectively.

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#### 1. Introduction

Active composite materials are mostly used in radio engineering, electronics and optoelectronics areas due to their pyro-piezo properties. For example, they are seen in applications like the protection of spacecraft from radiation, taking images of submarines, seismical and geological research, alternative energy sources and medical-biological areas [1–3].

The piezoelectric composites 0-3 and 1-3 can be obtained by randomly mixing polymer matrixes with ceramic-shaped piezoelectrics which have various particle sizes (PZT-5H, PZT-5A, etc.). These active matrix composites can be produced at specific shapes, and also their piezoelectric properties are protected [4–7].

A hybrid piezoelectric composite (HPC) is made of a structure with at least two groups. They occur as a result of the integration of low or high density polymers (PE—polyethylene, PVDF—polyvinylidene fluoride, PP—polypropylene) and nanosized metal oxides (BaTiO<sub>3</sub>, SiO<sub>2</sub>, TiO<sub>2</sub>) in the first group and low or high density polymers (PE, PVDF, PP) and micro-sized tetragonal The composite is called a nanocomposite in the first group, and the composite in the second group is called a micropiezo composite. The hybrid piezoelectric is acquired as a result of the integration of nanocomposite and micropiezo composite structures [8]. It can be said that the variety of composition at PZT family

structured PZT (lead zircon titanate) ceramics in the second group.

may not be quite enough in the development of piezo-pyro and electric materials. Not only piezo-pyroelectric properties, but the dielectric constants also increase in the materials with PZT. Nevertheless, this situation does not contribute to the efficiency of the material. Therefore, there is no chance for the noticeable increase of characteristic efficiency of the piezo-pyroelectric and electroceramic materials [9]. The situation of the micro-sized materials is not enough, especially for high efficiency and durable sensor applications [10].

In this study, electromechanical properties of the hybrid piezocomposite and micro-sized piezocomposite obtained with discharge plasma systems are investigated.

#### 2. Materials and method

Micro and hybrid piezoelectric composites are produced by using the discharge plasma technique. PZT-5A and 5H are used in

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Fig. 1. Process of the hybrid piezocomposite.



Fig. 2. Crystallization form used by the discharge plasma procedure.

the composites as active piezoelements, and the PVDF and HDPE (high density polyethylene) are used as a matrix. Nano-sized  $BaTiO_3$  and  $SiO_2$  are used in the hybrid structure as a nano-structured dope material. The process of the hybrid piezocomposite and the discharge plasma technique for the crystallization is shown in Fig. 1 and Fig. 2, respectively.

The processing of the hybrid piezocomposite is as follows:

(a) Formation of the substrate (microcomposite) by mixing with PZT and polymer and then pressing.

- (b) Formation of nanocomposite by mixing  $SiO_2$  or  $BaTiO_3$  with polymer and then pressing.
- (c) Formation of the hybrid structure by putting the microcomposite between two nanocomposites (the sandwich structure, Fig. 3a).
- (d) Pressing the sandwich structure under specific temperature and pressure.
- (e) Crystallization via discharge plasma.
- (f) Fastening the electrodes to the piezocomposite (Fig. 3b).
- (g) Polarization of the hybrid piezocomposite.
- (h) Transformation into piezosensor of the hybrid structured piezocomposites (Fig. 3c).

#### 3. Results and discussion

The  $d_{33}$ ,  $k_{33}$  tan  $\delta$ , Y (Young's modulus) and dielectric properties of the micro and the hybrid structured piezoelectric composites are shown in Table 1.

To reduce the fragility and to increase the elasticity of the materials, PZT-based materials with polymer composites are generated. It can be seen that the Young's modulus of the PZT + polymer composite is lower than that of single PZT material. As the Young's modulus of the material has a lower value, the elasticity of the material has the highest values. It is observed that the values of  $\varepsilon_{33}$ ,  $d_{33}$  and  $k_{33}$  are reduced as the polymer has low dielectric constant while the elasticity of the composite structure is increased. In this case, the hybrid structure is generated by doping nano-structure SiO<sub>2</sub> or BaTiO<sub>3</sub> into the PZT + polymer composite. The electromechanical properties of this hybrid composite structure have values higher than that of  $\varepsilon_{33}$ ,  $d_{33}$  and  $k_{33}$  of the PZT + polymer composite [8].

Although the single PZT materials have piezoelectric properties, they cannot be used singly in technological applications. The characteristics of PZT can be diversified by doping an acceptor and a donor into the PZT. However, this situation is limited. The variety of sensors based on PZT is increased by doping a polymer into the PZT, with the added benefit that the elasticity properties of the sensor are healed. Furthermore, it is seen that electron trapping ( $d_{33}$ ) and the coupling factor ( $k_{33}$ ) of the hybrid structure are increased due to fact that the hybrid has nano-sized SiO<sub>2</sub> or BaTiO<sub>3</sub> [9].

Thermostimulated depolarization (TSD) graphs of PVDF and PZT+PVDF materials are shown in Fig. 4. The availability of the PVDF polymer is not suitable owing to their low piezoelectric properties. Thus, they must be mixed with PZT material at a specific rate. PZT+PVDF polymer composite structure has elasticity and also piezoelectric properties.

As seen in the 1st peak in Fig. 4, the discharge temperature of only PVDF is  $110 \,^{\circ}$ C. There are two different peaks of the discharge temperature of the PZT + PVDF composite. One of them is at  $110 \,^{\circ}$ C due to PVDF; the 2nd peak in Fig. 4 is at about  $150 \,^{\circ}$ C, due to PZT; a 3rd peak in Fig. 4 belongs to the PZT + PVDF composite polarized under high voltage. As seen in the 1st peak in Fig. 4, the current value of only PVDF is not sufficient. However, the current value of the PZT + PVDF composite is 100 times larger than the current value of PVDF.

In Fig. 5, the TSD graph of the hybrid composite with  $BaTiO_3$  is shown. It can be seen that peaks similar to that of PVDF and PZT shown in Fig. 4 are obtained. In addition to these peaks, there is another third peak at 270 °C. In this peak at 270 °C, the current intensity (electron trapping) is increased by adding nano-sized  $BaTiO_3$  into PZT+PVDF composite. Thus, the hybrid composite (PZT+PVDF+BaTiO<sub>3</sub>) has better piezoelectric properties [8,10].

A TSD graph of the hybrid composite with nano-sized  $SiO_2$ (PZT + PVDF + SiO<sub>2</sub>) is shown in Fig. 6. The discharge temperature of

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