PIEZOELECTRIC PROPERTIES OF THE ACTIVE-MATRIX HYBRID COMPOSITES

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A hybrid piezoelectric composite structure is obtained by addition of nano sized $BaTiO_3$, SiO_2 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 ($BaTiO_3$ or SiO_2), and their piezoelectric, mechanic and electromechanics 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–$BaTiO_3$) composite are compared. The d_{33} value and the coupling factor of the hybrid structure have shown an average of 54 and 62% increase according to microstructure composite, respectively. In addition, the d_{33} value and the coupling factor of the hybrid structure (PZT–HDPE–SiO_2) have exhibited about 68 and 52% increase according to microstructure composite (PZT–HDPE), respectively.

Keywords: PZT $BaTiO_3$ SiO_2 Hybrid Piezoelectric Properties Piezoelectric sensor
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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, seismically 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 piezoelectric which have various particle sizes (PZT-5H, PZT-5A, etc.). These active-matrix composites can be produced at specific shapes, and their piezoelectric properties are protected [4–7]. A hybrid piezoelectric composite (HPC) is made of a structure with at least two groups. They occur because of the integration of low- or high-density polymers (PE—polyethylene, PVDF—polyvinylidene fluoride, PP—polypropylene) and nanosized metal oxides ($BaTiO_3$, SiO_2, TiO_2) in the first group and low- or high-density polymers (PE, PVDF, PP) and micro-sized tetragonal structured PZT (lead zircon titanate) ceramics in the second group. The composite is called a nanocomposite in the first group, and the composite in the second group is called a micro piezo composite. The hybrid piezoelectric is acquired because of the integration of nanocomposite and micro piezo composite structures [8]. It can be said that the variety of composition at PZT family 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 electro ceramic 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. MATERIAL AND METHODS

We consider the matrix composites (0-3 type) based on PKR-7M (tetragonal) type piezoelectric ceramics from family of lead zirconate titanate, thermoplastic polymer polyethylene of high density PEHD with a melt fluidity index of 1.3 g / 10 min (load-2.0 kg, temperature 190°C) and silicon dioxide SiO_2 dielectric. Selection of carbon-chain polymer-polyethylene is related to the fact that it is characterized with high reproducibility due to its composition, structure, and physical and chemical properties. A PKR-7M (PZT-5H) piezoceramic was selected due to its high piezoelectric modulus $d33 = 760 \cdot 10^{-12}$ C/N),), Young's modulus ($Y_{11}{}^{E} = 0.57 \cdot 10^{11}$ Pa) and dielectric permittivity ($\varepsilon_{33}/\varepsilon_0$ = 5 000). The silicon dioxide SiO_2 particles were used in spherical shape with density of 22 g/m^3, specific surface area of 200 m^2 /g, and the electrical conductivity of $10^{-12} \cdot m^{-1}$. The plasma crystallization of composite promotes arising the active centers with physical and chemical nature in the polymeric phase. Duration of discharge exposure was varied from 15 to 30 min. depending on the properties and volumetric content of polymer and piezoelectric ceramic in composite. A thickness of the gas gap, where micro-discharges were initiated was 0.5 - 4 mm. The voltage applied to the gas-insulator-composite system

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was 3 − 20 kV [6-10]. Selected SiO_2 nanoparticles possess significant surface activity and enough high activity. The high surface energy of nanoparticles leads to unusual surficial properties and reactions. The temperature and pressure for compressing of composites were selected within 437-463 K and 30 MPa respectively. The thickness and diameter of the piezo- composite were chosen as $250 \cdot 10^{-6}$ m, and $(160-200) \cdot 10^{-6}$ m respectively. Micro and hybrid piezoelectric composites are produced by using the discharge plasma technique. PZT-5A and 5H are used in the composites as active piezo elements, 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.

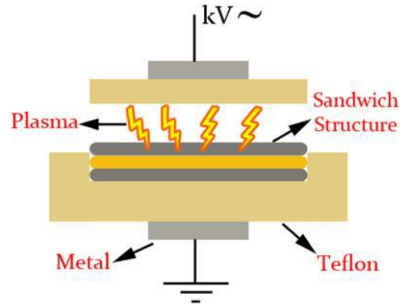

Fig. 1. Crystallization form used by the discharge plasma procedure.

3. RESULTS AND DISCUSSION

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_2 or $BaTiO_3$ [9].

In Fig. 2, the TSD graph of the hybrid composite with $BaTiO_3$ is shown. Peaks like that of PVDF and PZT shown in Fig. 2 are obtained. In addition to these peaks, there is another third peak at 270 °C. In this peak at 270 °C, the current intensity is increased by adding nano sized $BaTiO_3$ into PZT + PVDF composite. Thus, the hybrid composite (PZT + PVDF + $BaTiO_3$) has better piezoelectric properties [8,10]. A TSD graph of the hybrid composite with nano sized SiO_2 (PZT + PVDF + SiO_2) is shown in Fig. 3. The discharge temperature of the hybrid composite with nano-sized SiO_2 is like that of the hybrid composite with nano-sized $BaTiO_3$, as described above. The current values (electron trapping) of PZT + PVDF + SiO_2 are higher than that of PZT + PVDF + $BaTiO_3$.

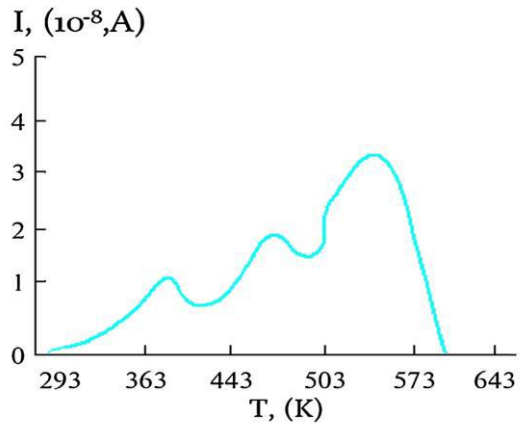

Fig. 2. TSD graph of the hybrid piezoelectric composite doped with nano sized $BaTiO_3$.

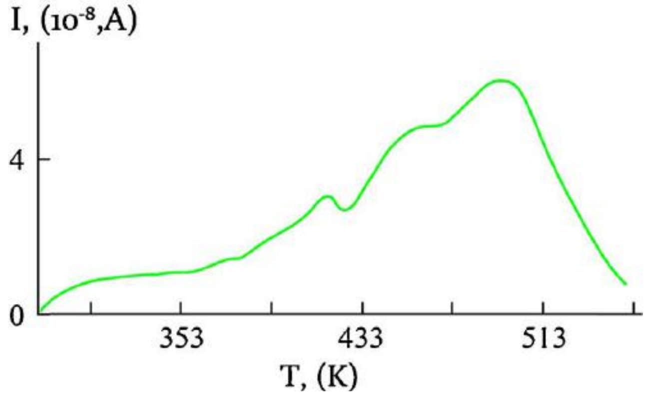

Fig. 3. TSD graph of the hybrid piezoelectric composite doped with nano sized SiO_2.

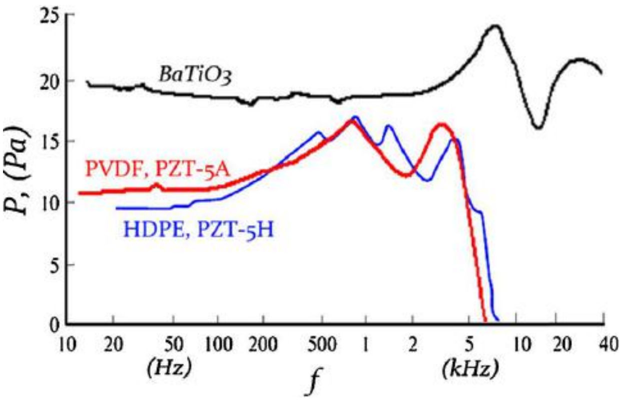

Fig. 4. Frequency characteristic of the micro piezoelectric composite and the hybrid piezoelectric composite doped with nano sized $BaTiO_3$.

The frequency characteristics of the hybrid and micro composites are shown in Fig. 4. The frequency of PZT and different polymers mixed with microgranular composites changes between 10 Hz and 5 kHz. No response is taken after this value. However, the hybrid composites have wide frequency characteristic. It is seen that the frequency characteristic, which has a decisive frequency between 10 Hz and 5 kHz, increases by 40 kHz.

4. CONCLUSIONS

In this study, it is seen that the hybrid (micro and nano) structured piezocomposites have higher piezoelectric properties compared to micro-sized piezocomposites. In addition, the frequency characteristic of the $BaTiO_3$-doped hybrid structured composite (PZT-5A + PVDF + $BaTiO_3$) is higher than that of the SiO_2-doped hybrid structured composite (PZT-5H + PVDF + SiO_2). The hybrid-based piezocomposites which are obtained by discharge plasma system can be used in acoustic applications at 5Hz–40 kHz. The hybrid structured piezocomposites, tailored specifically for the application areas and placed into molds of different structures, can transform into a piezo sensor.

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