Journal of Electrostatics 76 (2015) 89–94

Contents lists available at ScienceDirect

Journal of Electrostatics

journal homepage: www.elsevier.com/locate/elstat

The electret effects of crystallized polymer–ferropiezoelectric composite under electric discharge plasma

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ARTICLE INFO

Article history:
Received 4 September 2014
Received in revised form
15 March 2015
Accepted 11 May 2015
Available online 23 May 2015

Keywords:
Electret sensor
Fluorine-containing polymer and ferroactive composite
Electric discharge plasma

ABSTRACT

The fluorine-containing polymer composite filled with piezo-ceramics such as PZT-2, PZT-5A, PZT-8 were prepared by a hot pressing method. The electret composites were treated by an electric discharge plasma process, formed by crystallization via electro-thermal treatment, and polarized. The surface charge density and the charge relaxation time of the electret composite were investigated using thermo-stimulated depolarization and their potential differences were defined via a contactless measurement method. According to the obtained experimental results, the stability of the electret charge of a composite based on F42 + PZT-5A is better than a composite based on F3 + PZT-8.

Introduction

Electrets are an important inter-disciplinary scientific subject of dielectric physics, material science, sensor-medical and bio-engineering [1–3]. Electrets have developed rapidly with the increasing number of materials, methods for investigation of charge storage phenomena of creative and existing applications ranging from air filters to microphones. In addition, electrostatic recordings, artificial muscles, acoustic-mechanical transducers, electret motors and generators are used due to their excellent electromechanical properties [4,5].

The electret is a piece of a dielectric material which has been heated and specially cooled to provide dipolar domains and thus possesses an overall fixed dipolarity. It is analogous to a permanent magnet. Instead of opposite magnetic poles, the electret has opposing electrical poles of trapped opposite charge. As a result, there exists a fixed "static" potential difference ($E_{p\text{-}d}$) between the two opposite charges of the dipolar electret. The most important target of research on the electret is to increase the potential difference of the materials used make it.

Ferroelectric materials are required by symmetry considerations to be also piezoelectric and pyroelectric. Much research on ferroelectric and piezoelectric materials have shown that the use of PZT based materials that have an inorganic structure play an important role in obtaining high efficient piezoelectric ceramics. Although the PZT materials possess a stable electrical charge, their degree of the potential difference is inadequate (10–15 V). Heterogeneity of the material is one of the principal causes leading to the formation of deep charge carrier capture centers responsible for the continual existence of electrical charge. However, it is not easy to control the degree of heterogeneity related to the composite structure of the ceramic material. The use of composite materials manufactured from polycrystalline substances and glass can be one of the ways to overcome this difficulty [6].

The electret charge value, charge relaxation time – τ (life of electret charge), charge sign inversion, presence of internal–external remnant polarization, the coexistence of two charge types (hetero-homo), and the volume distribution of charge comprise the fundamental characteristics of an electret. These parameters are conditioned both by the technological systems of electret production and external factors affecting the electret during storage and operation. The applicability of materials used to make the electret generally depends on charge relaxation time and their potential differences. The electrets which have a longer charge relaxation time are better suited for use as an electric field source. To obtain longer charge relaxation time in an electret, it is necessary for the interaction of two different charge types (hetero-homo) according to modern theory of electrically charged effects. If there is a carrier trapping based on localized states with electrons in a conductivity band, various physical and chemical conditions occur.

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http://dx.doi.org/10.1016/j.elstat.2015.05.012
0304-3886/

Owing to the existence of numerous free states in the quasi-forbidden band of polymers, the induced energy electrons move to the quasi-Fermi level, where they are trapped in free states. As the trapping process of the quasi-Fermi level continues the density of the trapped electrons becomes so high that an electrostatic interaction occurs among them. The characteristics of this process interaction between homo and hetero-charges can be so strong that one of them can generate another. Therefore, the polarization process of the electret material will create an external charge effect in itself due to the hetero-charge in the dielectric [7–13].

The electret composites with deep trapping centers on interphase boundary relate to a composite structure based on polymer matrix and piezoelectric material. Previously, the composite materials for electrets have been based on the polyolefines and ferroactive solid solutions (FASS) which have high electrically charged [14]. Also it has been shown that composites based on fluorine-containing polymers and FASS have low electrically charged parameters although the phases of the composites have both hetero and homo-charges. Those electrets that were comprised of both polymer and piezo ceramics, which have relatively high electrophysical and electro-mechanical characteristics, were manufactured mainly from fluorine containing high molecular compounds [15,16].

This study aims to improve the existing electrically charged effects in composites that are based on fluorine-containing polymer + PZT under electric discharge plasma.

Materials and methods

The composite materials which are used to produce electrets should possess some specific characteristics such as high potential difference, high charge relaxation time (lifespan), relatively high dielectric permeability, high specific electric resistance, simplicity of manufacturing technique, and they use the piezoelectric materials as ferroelectric phase with stable domain structure and high Curie temperature.

In this study, an electret (polymer + PZT) composite is made of a structure with at least two groups. One of them is the polymer matrix, i.e., PVDF or F-2 (polyvinylidene fluoride $[-CH2-CF2-]_n$), F-3 (polytrifluoroethylene $[-CF2-CFH-]_n$) and F-42 (copolymer vinylidene fluoride with tetrafluoroethylene). The other is the piezoceramic PZT structure, i.e., rhombohedral (Re), tetragonal (T) and heterogeneous (Re + T) like that PZT-2, PZT-5A, PZT-8.

Some suggested electret composites are listed below:

- Polymer matrix (PVDF) + Piezoceramic Filler (PZT-2) with rhombohedral structure
- Polymer matrix (PVDF) + Piezoceramic Filler (PZT-8) with tetragonal structure
- Polymer matrix (F-3) + Piezoceramic Filler (PZT-8) with tetragonal structure
- Polymer matrix (F-42) + Piezoceramic Filler (PZT-5A) with rhombohedral structure

The processing of the electret composites is generally as follows:

1. The electret composites are prepared in an air medium by mixing polymer and PZT and pressing them at temperatures in the ranges of 200–240 °C according to the melting temperature of fluorine-containing polymers. The thickness of the composite is from 50 to 250 μm.
2. Firstly, deeply ionized trapping centers on the interface boundaries of the electret composite are formed by crystallization via electro-thermal treatment, also are polarized.
3. Secondly, the electret composites such as F3 + PZT8, F42 + PZT5A are treated by an electric discharge plasma process. After that the aforementioned composites are formed by crystallization via an electro-thermal treatment, and they are also polarized.
4. For all resulting electret composites, the currents of thermo-stimulated depolarization (TSD) spectra are measured separately. The surface charge density and the charge relaxation time of the electret composite are determined by TSD. The potential difference value of the electret having an electric charge is measured by means of a non-contact surface potential meter (Model 279, manufactured by Monroe Electronics) at room temperature.

The electro-thermal polarization

The electro-thermal polarization (or poling) was carried out on electret composites (Polymer + PZT) in the air gap using a ceramic strip heater. During polarization, firstly the electret composites were slowly heated from room temperature to polarization temperature (T_p: 140 °C) with a controlled heating rate of 5 °C/min and maintained at 140 °C for 1 h before applying a direct current (DC) voltage. After that, a DC electrical field (E_p) of 6 MV/m and 8 MV/m was applied for 30 min at 140 °C. After the polarization process, the electrets were cooled to room temperature. The polarity of the induced surface charges depends on the applied external DC field polarity, while the magnitude of the stored charge depends on the polarization temperature (Fig. 1).

The electric discharge plasma

The electric discharges were used as non-thermal non-equilibrium plasma, which is termed surface micro-discharges (SMD). The generation of SMDs at ambient pressure is obtained from the dielectric-barrier discharge (DBD) technology which is related to the corona discharge family. A typical cylindrical DBD configuration and its equivalent electric circuit are indicated in Fig. 2. Two planar electrodes are positioned in parallel with a gap and two dielectric barriers in between. Typical materials for DBD are usually Teflon plates. Because of a capacitive coupling of the insulating Teflon (dielectric capacitance – C_d) to the gas (atmosphere) gap, DBDs can be driven by alternative feeding voltage. The total capacitance – C value can be controlled by the gap capacitance – C_g. When alternative voltage with high amplitude is applied, and, if the local electric field strength in the gas gap arrives to the ignition level, an electrical breakdown (gas discharge) occurs inside the gap medium between parallel electrodes covered by the dielectric Teflon. In this situation, plasma is formed only as a micro-discharge in such a way that it carries low current and is surrounded by an atmosphere. This

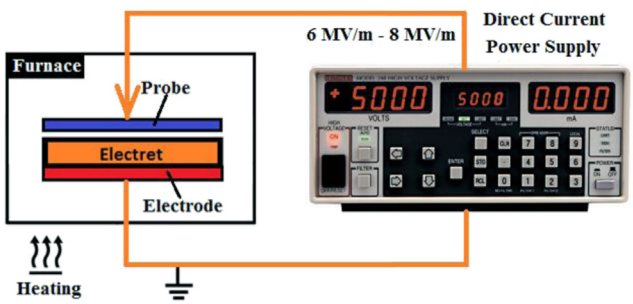

Fig. 1. Setup of the electro-thermal polarization.