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Experimental and theoretical study on time dependence of the quasi-piezoelectric d_{33} coefficients of cellular piezoelectret film

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ABSTRACT

The voided charged polymer film, also called piezoelectret, has a very large quasi-piezoelectric coefficient in the thickness direction, and has emerged as a new kind of electromechanical transducer materials. Piezoelectret film is usually prepared from polymer by means of biaxial stretching and electric charging process. Due to the inherent viscosity of polymer, the quasi-piezoelectric d_{33} coefficient of cellular piezoelectret film usually depends on the pressure and time of the measurement. In this article, experiments were carried out on the time spectra of quasi-piezoelectric d_{33} coefficient in the thickness direction for cellular linear Polypropylene piezoelectret film. To study the effect of void microstructures on the time-dependence of quasi-piezoelectric d_{33} coefficient, samples of three different thicknesses were tested under two different pressures. The micromechanical theory for viscoelastic composite was extended to predict the electromechanical properties of voided charged polymer film. The voids with surplus charges, which can be piezoelectriclike under deformation, are considered as ellipsoidal heterogeneous piezoelectric inclusions, while the viscous polymer is taken as the matrix. In Laplace transformed space, the generalized Eshelby tensor is formulated for the isotropic nonpolar matrix as well as for the anisotropic matrix. The Mori–Tanaka average scheme is used to find the overall electromechanical properties. Time dependence of the effective properties in real space can be studied by Laplacian inversion. Sensitivity analysis to various parameters is investigated for time dependence of the effective properties, including effective elastic moduli and the quasi-piezoelectric coefficients. Theoretical simulation was presented and comparison with experimental results was conducted. Both qualitative analysis and quantitative comparison with experiments show that this theoretical formulation can predict the time dependence of the effective properties of voided charged piezoelectret film.

1. Introduction

Cellular piezoelectret is actually a voided charged polymer film. Two main processes are needed in preparing piezoelectret film, i.e. making voids and electric charging. To make voids, the polymer film pre-mixed with very fine mineral particles is subjected to biaxial stretching. This process introduces into the polymer film many lens-like flat tiny voids. The voided polymer film is subsequently taken to electric charging. Under high electric fields, internal breakdown

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usually happens to the air inside the voids. Opposite charges are separated and trapped on the upper and bottom sides of the internal surface of voids, which constitute electric dipoles. Upon mechanical pressure, nonuniform deformation usually occurs to the voided charged film. Accordingly, the electric dipole moments of the charged voids change, which results in the quasi-piezoelectric effect macroscopically (Bauer et al., 2004; Qiu, 2010). In the thickness direction, piezoelectret film displays remarkable quasi-piezoelectric effect. The quasi-piezoelectric d_{33} coefficient of cellular Polypropylene (PP) piezoelectret film can be as large as that of the piezoelectric ceramics, e.g. Lead Zirconate Titanate (PZT), and much larger than that of the traditional piezoelectric polymer, Polyvinylidenefluoride (PVDF) (e.g., Gerhard-Multhaupt, 2002; Bauer et al., 2004; Lindner et al., 2004). In the past two decades, cellular piezoelectret film has emerged as a new kind of transducer materials, and shows great potential in the application of flexible electromechanical sensors and transducers.

So far, many works have been devoted to the understanding of material preparation, such as electric charging technology and the designing of void microstructure, etc. (e.g., Paajanen et al., 2000; Tuncer et al., 2005; Mellinger et al., 2006; Mellinger and Mellinger, 2011). But some fundamental theoretical relations between the effective electromechanical properties and the mechanical behavior of the host polymer as well as the geometrical microstructure of voids are yet far from clear. For example, since polymer usually displays viscous deformation, the overall electromechanical properties of cellular piezoelectret film usually depend on the pressure and time of measurement (Hillenbrand and Sessler, 2004; Dansachmüller et al., 2005). A theoretical model that can predict the time-dependent behavior of a voided charged polymer film will undoubtedly facilitate the design and application of the cellular piezoelectret film, and therefore, is highly demanded. On the other hand, the micromechanical means have been used in previous studies (e.g. Haberman and Berthelot, 2007; Wan et al., 2010), explaining theoretically the influences on the effective electromechanical properties of void microstructures, such as the void shape and void fraction. But as for the temporal evolution of the overall electromechanical behavior, what is the role of the void microstructure and how the void shape and void fraction influence the time-dependent behavior? This problem still remains not fully addressed, both theoretically and experimentally. A complete understanding of the overall behavior of the voided charged polymer calls for experimental investigation and theoretical models for the evolution with time of the effective electromechanical properties.

In this article, experiments were carried out for the time spectra of piezoelectric d_{33} coefficient in the thickness direction of cellular linear PP piezoelectret film. To consider the effect of void microstructures on the time-dependence of quasi-piezoelectric d_{33} coefficient, samples of three different thicknesses were tested under two different pressures. To simulate the time-dependent behavior of the effective electromechanical properties obtained in experiments, we have developed a micromechanical theory for the voided charged piezoelectret film. Since cellular piezoelectret films are generally of the thickness of about 100 μm, with the scattered void being around 10 μm thick, it is believed that the micromechanics means based on continuum mechanics still applies to this size scale. The developed theory is motivated by the basic fact that the creep behavior of the host polymer underlies the mechanism of temporal evolution of the overall electromechanical properties. Cellular piezoelectret films with different void microstructures exhibit different evolving behavior of the overall electromechanical moduli. Hence, to address the time-dependent behavior, a micromechanical model incorporating the creep behavior of the host matrix will be most necessary. The micromechanical theory for viscoelastic composite (Wang and Weng, 1992) is extended to predict the electromechanical properties of cellular piezoelectret film. The lens-like voids with surplus charges, which behave like piezoelectric media under mechanical deformation, are considered as ellipsoidal heterogeneous piezoelectric inclusions. The polymer is taken as the viscous matrix. In Laplace transform space, the generalized Eshelby tensor is formulated for both the isotropic matrix and the anisotropic matrix. The Mori–Tanaka average scheme is used to find the overall electromechanical properties. Time dependence of the effective properties in real space can be obtained by numerical Laplacian inversion. Sensitivity analysis to various parameters is investigated for time dependence of the effective properties, including the effective moduli and the quasi-piezoelectric d_{33} coefficient. Comparison was made between theoretical simulation and experiments. Both qualitative analysis and quantitative comparison with experiments show that this theoretical formulation can predict well temporal evolution of the effective properties of voided charged piezoelectret film.

2. Experiments

Materials used in this experimental study are linear PP films (manufacturer designation PQ50) with the original thickness of 50 μm and bulk density of 550 kg/m^3. The cavities of the material are generated by the manufacturer by biaxial stretching of the extruded films containing mineral particles (such as $CaCO_3$), as shown in Fig. 1. The PP films were cut into small pieces with the dimensions of 10 cm × 10 cm, and then inflated by a pressed-gas expansion process at a given pressure of 1.9 MPa (Zhang et al., 2004a, 2004b). For the inflated films with different thickness, the elevated temperatures, 90 °C, 110 °C and 150 °C, were used in the inflation process. Thereafter samples were charged for 60 s by a corona process by utilizing a needle voltage of −20 kV. Metallization with aluminum on both sides was subsequently conducted. Finally, aging was taken at the temperature of 90 °C for 30 min. The samples used in this experiment were of a diameter of 20 mm. For the measurement of piezoelectric response in time-domain, in the present study, the induced charges Q on electrode were recorded by Keithley 6514 electrometer over the time of the force F applied on the sample provided by the weight, as shown in Fig. 2. The relation between the d_{33}-coefficient and the induced charges on the