Dielectric Barrier Discharges in Micrometer Sized Voids

1 Introduction: Ferroelectrets

Piezo- and pyroelectricity of mechanically and electrically heterogeneous polymers were first described three decades ago [1], but at that time, the experimentally observed effects were too small to be of use in most applications. This changed with the advent of electrically charged cellular polymers or polymer foams [2] (now frequently called ferroelectrets or piezoelectrets) in the mid-1990s. Since then, the field has expanded rapidly, as documented in several review articles [3–6]. Ferroelectret films have been produced in a variety of ways, including blow-extrusion of charge-storing polymers (e.g., polypropylene (PP)), voiding of various polymers with supercritical carbon dioxide [7, 8]), and most recently as as template-based regular structures [9–12]. Typical void heights are of the order of 1...30 µm.

The voided structure is then electrically charged, either in a corona discharge [15], or in direct contact with metallic thin-film electrodes [16]. As the field in the gas-field voids exceeds the threshold for Paschen breakdown, a dielectric barrier discharge results, in which pairs of electrical charges are created and deposited on opposite surfaces of the internal void (Fig. 1). It is these “engineered” electrical dipoles that give the material its piezoelectric properties.

Ferroelectrets have a variety of applications such as sensors (intrusion detection, smart packaging), actuators (wall-sized loud speakers, active noise canceling) and energy harvesting (proposed...
applications such as shoe inserts and tire implants). Some applications of ferroelectrets have recently appeared on the market, most notably bed sensors for monitoring the movement of hospital patients [17].

While there has been considerable research to widen the materials base of ferroelectrets, significant work remains to be done in order to understand the charging process, and recent research has opened up significant questions. Investigation of the light emission during the charging of cellular PP films [6, 18, 19], confirmed the hypothesis that charges are generated in dielectric barrier discharges. The quasi-polarization of the internal voids (positive charge on one surface, negative charge on the opposite surface) can be reversed by sufficiently high electric fields and the resulting “effective charge density versus field” curves exhibit hysteresis behavior [18]. It was shown that back-discharges (initiated by the electric field of the deposited space charges) destroy approx. 75% of the deposited space charge [18]. Minimizing the back discharges could thus increase the piezoelectric $d_{33}$ coefficient by a factor of up to 4.

Very recently, earlier electromechanical models [20–22] were extended by taking into account a realistic distribution of void heights in cellular polypropylene, using a “thick stack” of voids with different height classes (Fig. 2 (left)). While the calculated space charge hysteresis curves were in very good agreement with experimental data, the onset of piezoelectric activity was observed at significantly higher electric fields than predicted by Townsend’s model of Paschen breakdown [14]. Using modified Paschen constants, however, good agreement between observed and calculated $d_{33}$ coefficients as a function of the applied charging voltage was reached, as shown in Fig. 2 (right). It is evident that the commonly accepted Paschen curve for electric breakdown in air poorly describes the critical electric field for dielectric barrier discharges in micrometer-size cavities. Interestingly, the observed departure (higher $E_c$ at small gap sizes) is opposite to the results of recent particle-in-cell simulations [23].

When applied to charging conditions at various pressures in a dry nitrogen atmosphere, the model predicted that the piezoelectric charge density could be more than doubled by optimizing the pressure [13, 24]; Fig. 3 (left) shows that this result is in excellent agreement with the experimental data. Moreover, the calculations yielded the amount of space charge deposited on the internal void

![Figure 3](image-url)
surfaces, as shown in Fig. 3 (right). Two features stand out:

- Small voids (height < 7 µm) do not get charged at all. Since these voids exist in large numbers in most voided materials, being able to electrically charge them promises large gains in the piezoelectric activity.
- Large voids can potentially be charged to a high surface charge density; however, when the external electric field is turned off at the end of the charging cycle, back discharges limit the amount of residual charge density to approximately 0.3 mC/m² at atmospheric pressure.

Hence, improving charge deposition in micrometer-sized voids is the primary goal of this project.

## 2 Proposed Work: Microplasma Discharges and Optimized Charging

Recent research has indicated that conventional charging methods have only a limited efficiency [13, 18], since (a) only voids larger than 6-8 µm are charged, and (b) back discharges destroy a substantial amount of the space charge in the larger voids. Improving the performance of ferroelectrets requires careful study of the microplasma discharges in the closed voids using a combination of electromechanical techniques with optical spectroscopy and imaging. The goals of our project are:

- Measure breakdown fields in µm-size cavities as a function of void height and gas composition
- Use these measured breakdown fields (instead of the currently used values that are based on a modification of Paschen’s law) to verify our recently developed model [13].
- Using the model to predict (and experimentally verify) approaches to maximize the charging efficiency by optimizing pressure, gas composition and the applied voltage vs. time.

A “model cavity” consisting of a single void with an adjustable µm-size air gap will be built and subjected to high-voltage profiles while the light emission is imaged and spectroscopically analyzed with an electron-multiplying CCD camera (Andor IXON DU-987), featuring a readout noise of less than 1 electron per pixel (Fig. 4). Simultaneously, the breakdown currents due to barrier discharges will be amplified with a FEMTO DLCPA-200 current amplifier and subsequently recorded with a NI PCI-6221 data acquisition board. These experiments will yield breakdown fields for a variety of gases, indicating to what extent the Townsend model for Paschen breakdown is applicable to dielectric barrier discharges in small gaps. The breakdown data will be added to our model describing the hysteresis behavior the effective space charge density $\sigma_{\text{eff}}$ [13, 14]. This model will then allow predictions about the electromechanical behavior of other polymer foams, leading to ferroelectrets with higher performance. The model cavity will be built using one of the recently published templating techniques [9–12], where patterned spacer films are sandwiched or laminated between
non-patterned electret films (typically, biaxially oriented polypropylene, cycloolefin copolymers or various fluoropolymers, cf. Fig. [5].

Based on the predictions of the charging model, parameters such as gas pressure and gas composition will be varied to maximize the deposited space charge. In addition to dc charging voltages, short pulses generated by a high-voltage switch (Willamette High Voltage PHVSW-015V, rise-time 10 ns) will be used, as recent research on pulsed microplasma discharges has indicated an improved plasma-generation efficiency [25,26], and it is likely that this approach will also increase the space charge yield.

3 Resources

All instrumentation listed is available in Dr. Mellinger’s research laboratory at the Department of Physics, Central Michigan University. In addition, the applicant will be able to use resources and instrumentation in the Department of Chemistry and the School of Engineering, which are both participating in the interdepartmental “Science of Advanced Materials” Ph.D. program.

References