

# Piezoelectric cellular polymers as sensors or air-borne ultrasonic transducer: Property adjustment by foam-structure and geometry variations

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## Abstract:

Electrically charged polymers with a cellular foam structure show piezoelectric properties and are discussed as so-called ferroelectrets. Research requests are an optimization of the ultrasonic properties of ferroelectrets such as low thickness-extension resonance frequencies as well as high operating voltages for the final transducer. We present two approaches for modification of ferroelectrets which fulfill these requirements. One approach is the preparation of thicker transducer films in order to decrease the resonance frequencies, another approach is the use of dielectric layers in hybrid transducers in order to shift the onset for dielectric breakdowns within the voided structure. Beside the film processing, the electrical poling as well as the resulting transducer properties are discussed.

Key words: piezoelectric, piezoelectric sensor, piezoelectric actuator, ultrasonic transducer.

## Introduction

Piezoelectric polymers are widely used as sensor and actuator materials, while research in material development is focused on dipolar orientation processes, space-charge trapping mechanisms and electrical-mechanical interactions. The main working horse in the field of piezoelectric polymers are materials such as polyvinylidene fluoride (PVDF) and its copolymers with trifluoroethylene (P(VDF-TrFE)) and hexafluoropropylene (P(VDF-HFP)) [1-5]. PVDF and its copolymers show pronounced piezoelectric activities with piezoelectric coefficients in the range of typically 10 to 30 pC/N for the piezoelectric thickness (33) or in plane (31) effects. For application of polymers as ultrasonic transducers, often the piezoelectric 33-effect is used. However, typical transducer film thicknesses of some ten to some hundred  $\mu\text{m}$  and an elastic modulus in the range of 2 to 10 GPa leads to thickness-extension resonance frequencies in the range of some MHz to hundred MHz. In addition, an acoustic impedance of  $3 \times 10^6 \text{ kg/m}^2\text{s}$  allows an optimal acoustic matching to water but leads to a non-optimal coupling e.g. to air if such piezoelectric materials should be operated as air-borne ultrasonic transducer.

Ferroelectrets show completely other properties if they are used as ultrasonic transducers. Ferroelectrets are composites which consist of space-charge electrets arranged in a foamed structure, while the voids are usually filled with air [6-9]. The physical principle of ferroelectrets is: (i) an electrical poling of the voided polymer structure leads to trapped charges of opposite polarities located at opposite void surfaces within the cellular structure and thus to huge electrical dipoles, (ii) the soft polymer-air composite structure allows a deformation under mechanical load which leads to the change of the dipole sizes and (iii) thus to the generation

of an electrical signal during the deformation of the composite structure. The detected electrical signals are mainly linear depending on the applied mechanical load. Thus, the systems are discussed as piezoelectric transducers. Beside earlier studies on polypropylene (PP), the material basis was extended to other space-charge electrets such as polyethylene terephthalate (PET) [10], polyethylene naphthalate (PEN) [11], fluorinated ethylene propylene copolymers (FEP) [12] as well as cycloolefine polymers (COP) and copolymers (COC) [13].

The advantages of ferroelectrets are their cheap basis materials which allow large-area processing of piezoelectric transducers and especially the interesting ultrasonic properties which originate from their low-elastic modulus combined with the air-filled composite structure. It was found that ferroelectrets exhibit a huge potential as sensor and actuator materials in air-borne ultrasonic transducers [14]. This evaluation is based on the determined thickness-extension resonance frequencies in the range of some hundred kHz to about 2 MHz, combined with a good acoustic coupling to air represented by an acoustic impedance in the range of 2 to  $10 \times 10^4 \text{ kg/m}^2\text{s}$ . In addition the found piezoelectric activities are also relatively high, piezoelectric  $d_{33}$  coefficients between some ten and thousand pC/N or pm/V were reported for different kinds of ferroelectrets.

However, also for classical ferroelectrets made from PP with a cellular structure there is the interest in optimization of properties such as the reduction of the thickness-extension resonance frequency or the increase of the driving voltage during operation. Here we present two attempts which fulfill the above mentioned optimization scenarios.

**Ferroelectrets with reduced resonance frequency**

The ultrasonic properties of ferroelectrets can be adjusted by an optimal electrical poling which defines the amplitude of the actuator and sensor signal and by variations of the transducer geometry, e.g. the thickness, as well as the inner microscopic structure which defines the elastic properties of the samples. The inner voided structure is varied by means of a gas diffusion expansion of an initial foamed polymer film [15-16]. Classical PP ferroelectrets can be processed in a broad variation containing relatively flat voids to round voids corresponding to high and low composite densities, respectively. A typical dependence of the elastic and electro-mechanical properties from the voided structure is shown in Fig. 1.

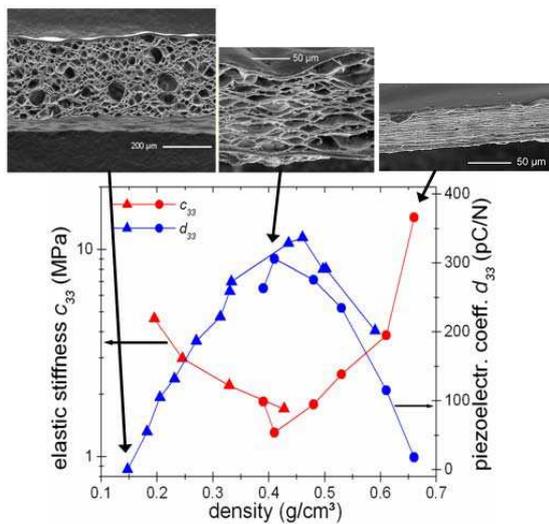


Fig. 1: Ferroelectrets with different voided structures: Dependence of the density, elastic stiffness and piezoelectric properties on the foam structure.

The underlying film thicknesses of the transducer films shown in Fig. 1 are in the range of 34 to 40 μm for high density / low expanded films and 230 μm for low density / maximum expanded films. The above mentioned films show thickness-extension resonance frequencies in the range of 600 kHz to 2 MHz, depending on the foam structure.

With preparation of initially thicker films then the above mentioned 40 μm thick transducer films we were able to produce ferroelectrets which similar void structures compared to that of the films shown above, but with thicknesses between about 90 and 340 μm. All other preparation steps such as expansion, electrical poling and metallization were employed in a similar way, like the preparation of the initially 40 μm thick films. As a result, those thicker transducer films shown thickness-extension resonances between 233 and 261 kHz and thus much lower resonance frequencies then the above mentioned initially 34 to 40 μm thick films.

**Ferroelectrets with increased operating voltage**

In order to obtain ferroelectrets with increased breakdown voltage we processed hybrid systems of classical ferroelectrets with a dielectric cover layer at one surface as schematically shown in Fig. 2. Here, the dielectric cover layer is made of polymethylmethacrylate (PMMA) processed from a liquid phase via spin coating on top of the ferroelectret film. Details about the processing and the available hybrid systems are given in [17].

Depending on the processing parameters during spin coating, the thickness of the PMMA layer can be adjusted between 1 and 16 μm. The initial thickness of the ferroelectrets before expansion was about 40 μm. The films were electrically charged in direct contact applying voltages up to 8 kV. This electrical poling leads to maximum piezoelectric activities of 165 pC/N and 195 pC/N for the pure and the hybrid ferroelectrets. More interesting is that due to the PMMA coating layer the threshold voltage for dielectric breakdowns within the voids is shifted from about 3 kV for the pure ferroelectret films to about 4 kV for the hybrid ferroelectret films. This allows the use of hybrid ferroelectrets films in ultrasonic probes applying operating voltages up to 4 kV. In addition, the dielectric layer provides a mechanical load which lowers the overall thickness-extension resonance frequency. The measured thickness-extension resonance frequencies of hybrid ferroelectret transducers coated with different thick PMMA layer are shown in Fig 3.

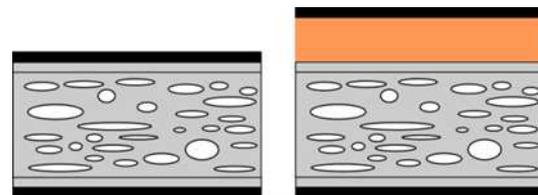


Fig. 2: Left picture: Schematic sketch of a classical ferroelectret covered with electrodes. Right picture: Schematic sketch of a hybrid system of a dielectric PMMA layer (colored orange) processed onto a ferroelectret film.

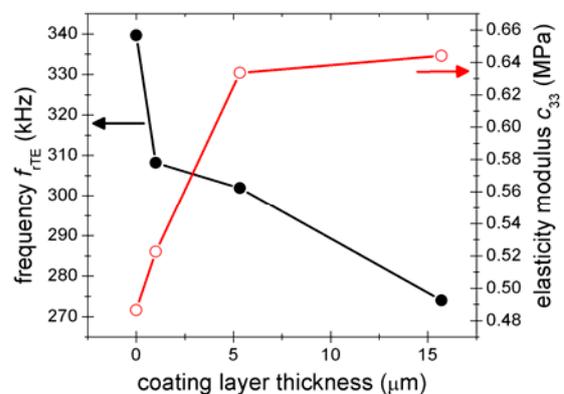


Fig. 3: Thickness-extension resonance frequencies of hybrid transducers consisting of a ferroelectret coated with different thick PMMA layer.

## Summary

In summary, two preparations of ferroelectrets with lowered thickness-extension resonance frequencies are reported. In one case, the well known preparation steps of ferroelectrets with different voided structures are applied to voided films with an initially increased thickness. Here a broad variation of piezoelectrically active films with thickness-extension resonance frequencies down to 233 kHz are received. In a second case, classical 40  $\mu\text{m}$  thick films were expanded afterwards coated with an additional dielectric layer. Depending on the thickness of the dielectric layer the thickness-extension resonance frequency is adjustable. Further more, the threshold for dielectric breakdowns within the voids is increased with the increase thickness of the coating layer which allows higher voltage operation of the ultrasonic transducer.

## References

- [1] H.S. Nalwa (Ed.), **Ferroelectric Polymers**, Marcel Dekker Inc., New York 1995.
- [2] R. Gerhard-Multhaupt, **Electrets**, in: J.G. Webster (Ed.), *Wiley Encyclopedia of Electrical and Electronics Engineering*, Vol. 6, John Wiley & Sohn, Inc., New York 1999.
- [3] M. Wegener, W. Künstler, K. Richter, and R. Gerhard-Multhaupt, **Ferroelectric polarization in stretched piezo- and pyroelectric poly(vinylidene fluoride-hexafluoropropylene) copolymer films**, *Journal of Applied Physics* 92 (12), 7442-7447 (2002).
- [4] F. Wang, P. Frübing, W. Wirges, R. Gerhard, and M. Wegener, **Enhanced polarization in melt-quenched and stretched poly(vinylidene fluoride-hexafluoropropylene) films**, *IEEE Transactions on Dielectrics and Electrical Insulation* 17 (4), 1088-1095 (2010).
- [5] P. Frübing, F. Wang, M. Wegener, **Relaxation processes and structural transitions in stretched films of polyvinylidene fluoride and its copolymer with hexafluoropropylene**, *Applied Physics A* 107, 603-611 (2012).
- [6] M. Wegener, W. Wirges, R. Gerhard-Multhaupt, M. Dansachmüller, R. Schwödiauer, S. Bauer-Gogonea, S. Bauer, M. Paajanen, H. Minkkinen, and J. Raukola, **Controlled inflation of voids in cellular polymer ferroelectrets: Optimizing electromechanical transducer properties**, *Applied Physics Letters* 84 (3), 392-394 (2004).
- [7] M. Wegener, S. Bauer, **Microstorms in cellular polymers: A route to soft piezoelectric transducer materials with engineered macroscopic dipoles**, *ChemPhysChem* 6, 1014-1025 (2005).
- [8] S. Bauer, **Piezo-, pyro- and ferroelectrets: Soft transducer materials for electromechanical energy conversion**, *IEEE Transactions on Dielectrics and Electrical Insulation* 13 (5), 953-962 (2006).
- [9] M. Wegener, **Ferroelectrets – Cellular piezoelectric polymers**, in: *New Materials for Micro-scale Sensors and Actuators – An Engineering Review*, Edited by S. Wilson and C. Bowen, *Materials Science and Engineering: R: Reports* 56, 78-83 (2007).
- [10] W. Wirges, M. Wegener, O. Voronina, L. Zirkel, and R. Gerhard-Multhaupt, **Optimized preparation of elastically soft, highly piezoelectric, cellular ferroelectrets from non-voided polyethylene-terephthalate films**, *Advanced Functional Materials* 17, 324-329 (2007).
- [11] P. Fang, M. Wegener, W. Wirges, R. Gerhard, and L. Zirkel, **Cellular polyethylene-naphthalate ferroelectrets: Foaming in supercritical carbon dioxide, structural and electrical preparation, and resulting piezoelectricity**, *Applied Physics Letters* 90, 192908 (2007).
- [12] O. Voronina, M. Wegener, W. Wirges, R. Gerhard, L. Zirkel, and H. Münstedt, **Physical foaming of fluorinated ethylene-propylene (FEP) copolymers in supercritical carbon dioxide: single-film fluoropolymer piezoelectrets**, *Applied Physics A* 90, 615-618 (2008).
- [13] E. Saarimäki, M. Paajanen, A.-M. Savijärvi, and H. Minkkinen, M. Wegener, O. Voronina, R. Schulze, W. Wirges, and R. Gerhard-Multhaupt, **Novel heat durable electromechanical film: Processing of electromechanical and electret applications**, *IEEE Transactions on Dielectrics and Electrical Insulation* 13 (5), 963-972 (2006).
- [14] V. Bovtun, J. Döring, M. Wegener, J. Bartusch, U. Beck, A. Erhard, and V. Borisov, **Air-coupled ultrasonic applications of ferroelectrets**, *Ferroelectrics* 370, 11-17 (2008).
- [15] M. Paajanen, M. Wegener, and R. Gerhard-Multhaupt, **Understanding the role of the gas in the voids during corona charging of cellular electret films – A way to enhance their piezoelectricity**, *Journal of Physics D: Applied Physics* 34, 2482-2488 (2001).
- [16] M. Wegener, W. Wirges, J. Fohlmeister, B. Tiersch, and R. Gerhard-Multhaupt, **Two-step inflation of cellular polypropylene films: Void-thickness increase and enhanced electromechanical properties**, *Journal of Physics D: Applied Physics* 37 (4), 623-627 (2004).
- [17] M. Sborikas and M. Wegener, **Piezoelectric-property adjustment of cellular ferroelectrets by foam-structure and geometry variations**. *Proceedings of the ASME 2012 Conference on Smart Materials, Adaptive Structures and Intelligent Systems, SMASIS2012-8002* (2012)