CELLULAR POLYPROPYLENE FERROELECTRET FILM: PIEZOELECTRIC MATERIAL FOR NON-CONTACT ULTRASONIC TRANSDUCERS

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Abstract

Electromechanical properties of the cellular polypropylene ferroelectret films (EMFIT) evidence their high potential for the air-coupled ultrasonic applications. The disadvantage with respect to their low coupling factor could be compensated by the extremely low acoustic impedance. A figure of merit is proposed to compare and estimate piezoelectric materials intended for the air-coupled ultrasonic applications. Electron beam evaporation technology was adapted to the EMFIT films and films with two-sided Au and Al electrodes were prepared without reducing or suppressing of the electromechanical properties. Finally, air-coupling ultrasound transducers based on the EMFIT films were developed and characterized.

Keywords: air-coupled ultrasound, ultrasonic transducers, electromechanical properties, piezoelectric materials, impedance spectroscopy

1 INTRODUCTION

A non-contact ultrasonic technique, where air is the only coupling medium, is a promising approach adding to the number of contact methods [1-5, 8]. It allows for an automated testing without any additional treatment of the material and under ambient conditions. The main problem of air-coupled ultrasound (ACU) techniques is the very high impedance mismatch between air and material under test, air and transducer. The mismatch is responsible for the high attenuation of the information-carrying signal between transmitter and receiver. Accounting the four air-ceramic interfaces [2, 3, 6] the attenuation can be as high as -300 dB. While the losses caused by the impedance mismatch between air and material under test are unavoidable, the losses due to the impedance mismatch between air and transducer can be reduced in principle. Improvement of the classical piezoelectric or electret transducer design (matching layers, membranes, back plate holes, etc.) and use of the pulse-compression techniques has resulted in the decrease of the attenuation down to -200 dB [2, 6, 7]

Smart polymer foams seem to be much more interesting and promising for the ACU applications. The EMFIT film (Emfitech Ltd, Finland) [10-12] is an example of smart polymer foams with a unique combination of the strong electromechanical response and very low density (i.e., low acoustic impedance). This electret material with many built-in electric dipoles behaves similar to ferroelectrics and is called therefore a ferroelectret [13-15]. Soft ferroelectrets recently attract a great interest of researcher due to the both unusual physical properties and tentative applications [10, 11, 13]. In this paper we report on the results of our impedance spectroscopy study and discuss perspectives of the cellular polypropylene ferroelectret films for the air-coupled ultrasonic applications.

2 EXPERIMENTAL

Taking into account the extreme softness and susceptibility of the ferroelectret film, one can suppose a significant influence of the electrode mass on the electromechanical properties. We studied the evaporation of a few kinds of the commercially available cellular polypropylene ferroelectret EMFIT films (Emfitech Ltd, Finland) [12]: HS-04-10BR and HS-03-20BR, a 70 µm thick cellular film, and HS-03-20BR Al1, a 70 µm thick cellular film with a one-sided aluminium (Al) electrode evaporated directly to the film.

We used gold (Au) and aluminium (Al) as electrode material. To ensure a sufficient high conductivity of our electrodes and to avoid additional undesired energy losses, we limited the minimal thickness of electrodes to about 100 nm. Subsequently we checked the influence of the chromium (Cr) sub-layer. The latter also could improve the adhesion to the polymer. The following samples, denoted with (i) in the following, of the EMFIT films with evaporated electrodes on both surfaces were investigated in details by means of impedance spectroscopy (see tab. 1):

No. sample	film name	thickness	electrodes	Cr-sub layers
(1)	HS-04-10BR	200 nm	Au	10 nm Cr sub-layer
(2)	HS-03-20BR	100 nm	Al	
(3)	HS-03-20BR	300 nm	Al	
(4)	HS-03-20BR	100 nm	Al	10 nm Cr sub-layer
(5)	HS-03-20BR	300 nm	Al	10 nm Cr sub-layer

Tab. 1. Five samples consisting of different film thickness and electrode materials.

The frequency dependences of the impedance (real part R and imaginary part X) and admittance (real part G and imaginary part B), as well as of serial capacitance C_s and dielectric losses $tan\delta$ were measured using a computer controlled Hewlett-Packard 4194A Impedance Analyser, 100 Hz – 40 MHz. The measurements were mainly performed in the range of electromechanical resonance corresponding to the thickness mode, i.e. between 100 KHz and 1,5 MHz. The main electromechanical properties, such as resonance frequencies of the parallel (F_p) and serial (F_s) resonance, sound velocity (V_3), coupling factor (K_{33}), quality factor (Q), mechanic ($tan\delta_m$) and dielectric ($tan\delta$) losses, dielectric permittivity ($\varepsilon^T{}_{33}$), acoustic impedance (Z_A) were calculated from the experimental data [1, 9]. Because of the small difference between F_p and F_s ($F_p \approx F_s$), the coupling factor was additionally estimated by accurately measuring the difference between the mechanically free (C^T) and mechanically clamped (C^S) capacitance [9]:

$$C^T = C^S(1+K^2{}_{33}). \tag{1}$$

To study the sensitivity, sensors with an embedded EMFIT film were used as transducers in the air-coupled ultrasonic experiment, which was performed by means of Panametrics Pulser / Receiver 5800 and Yokogawa Digital Oscilloscope DL 7100.

3 RESULTS AND DISCUSSION

The resonance characteristic is depicted in Fig. 1. This includes the main frequency dependent properties such as $C(f)$, $tan\delta(f)$, $R(f)$, and $G(f)$. Contrary to Fig. 1 no frequency dependence is usually observed for $X(f)$, $B(f)$ which are mainly defined by the capacitive response. Taking into the account that for all measured samples $X >> R$ and $B >> G$, we find that

$$Z(f) \approx X(f) \sim (f{*}C)^{-1},\ Y(f) \approx B(f) \sim (f{*}C). \tag{2}$$

A small difference between the parallel and serial resonance frequencies, evidences the low electromechanical coupling *($F_p \approx F_s$*, see also Fig. 1, right a and b). According to equation (1) this was confirmed by estimating the coupling factors from the *C(f)* step-down due to mechanical clamping. Here we found K_{33} = 0.06 for the HS-04-10BR film and K_{33} = 0.05 for the HS-03-20BR film.

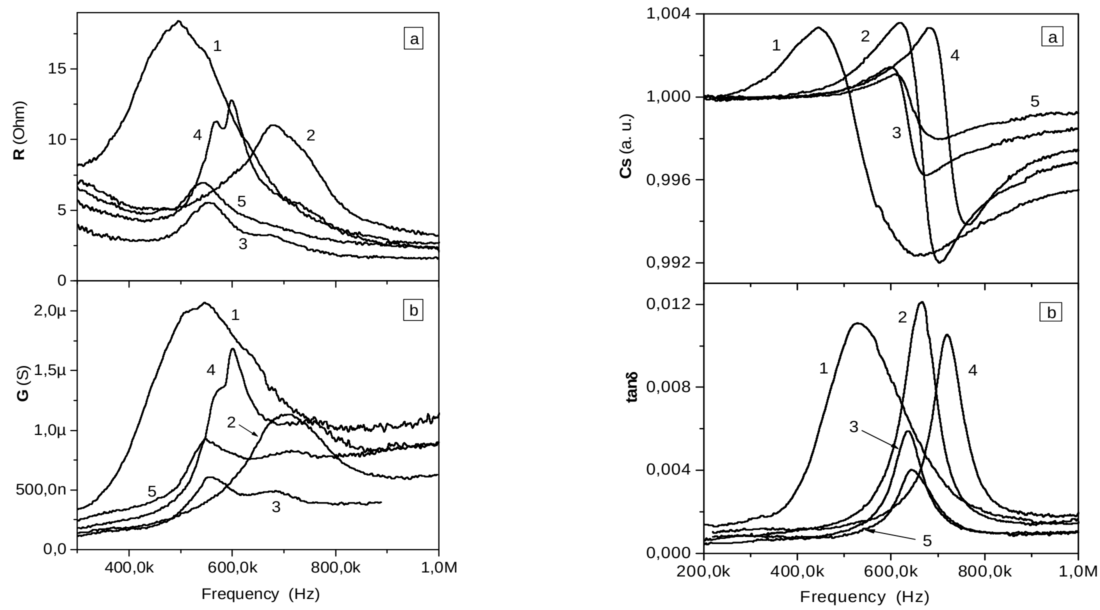

Fig. 1. Comparison of the EMFIT films with different evaporated electrodes. *C(f)*, *tanδ(f)*, *R(f)*, and *G(f)* as function of the operation frequency. Sample no. (1) – (5) denote the different films according to tab.1. Left: Films without embedding them into the sensor, right: Film was embedded into a sensor.

These experimental findings reveal an extreme softness of the ferroelectret film along the 33-direction, perpendicular to the surface. The in-plane electromechanical properties are less developed. The anisotropy is caused by the layered structure and the lens-like shape of the voids [11, 13, 14]. The resonance anomalies are weak in comparison to that of ferroelectric polymers or piezoceramics [9] and are superimposed on the dominating pedestal formed by the impedance (or respectively admittance) of low-loss capacitor.

The dielectric loss of the EMFIT films are very low (Fig. 1) and achieves the maximal values of $tan\delta = 0.005 - 0.012$ at the resonance. No significant dielectric dispersion was observed in the frequency interval ranging from 10 kHz to 40 MHz. However, slight anomalies could be observed due to electromechanical resonances. The dielectric permittivity of the EMFIT films is very low (ε^T_{33} = 1.1−1.2) indicating a great volume fraction of the air-filled voids. We observed no dielectric dispersion and low losses implying that the conductivity contribution of the additional metal-layer and low frequency polarization mechanisms at ultrasonic frequencies (at f > 10 kHz at least) does not play an important role. This is a promising feature for further sensor development and analysis. Mechanical losses are also low ($tan\delta_m$ = 0.05) corresponding to a quality factor which is as high as Q = 1100.

Evaporated electrodes and embedding the films within a sensor can be considered as a mechanical load for the extremely soft EMFIT films and result in the decrease of parallel and serial resonance frequencies observed by the shift of $R(f)$ or $G(f)$ maxima respectively (see Fig. 1, right). Taking into account the small thickness of the EMFIT films (70 µm), the resonance frequency is very low (600 − 850 KHz) and corresponds to the extremely low sound velocity V_3 = 85 − 120 m/s. This value apparently shows the inhomogeneous structure of the film. Together with the low density (ρ =

330 kg/m^3), the extremely low sound velocity results in the anomalously low acoustic impedance of the EMFIT film: $ZA = \rho V_3 = (0.028 - 0.040)106$ kg/m^{2}s = 0.028 − 0.040 M_{Rayl}. It is only two order of magnitude higher than the acoustic impedance of air (Z_{air} = 0.00042 M_{Rayl}), but three orders of magnitude less than the impedance of ceramic or crystal piezoelectrics and two order of magnitude less than the impedance of composite or polymer piezoelectrics. Generally, EMFIT film demonstrates acceptable electromechanical properties in comparison with other piezoelectric materials (Tab. 2):

Material / Property	Ferroelectret film EMFIT	Ferroelectric polymer film PVDF	Ceramic (PZT) -polymer composite	Ceramics PZT
Density ρ [kg/m^3]	330	1780	1000-5000	4000-8000
Dielectric permittivity [10 kHz], $\varepsilon_{33}{}^T$	1.12 – 1.23	12	50-500	150-3500
Dielectric losses [10 kHz], $tan\delta$, 10^{-3}	1	10	1-100	1-10
Coupling factor K_{33}	0.06	0.11-0.15	0.65	0.35-0.55
Quasi-static piezoelectric coefficient d_{33} [pC/N]	25 – 700* 80	20-25	50-300	70-600
Sound velocity V_3 [m/s]	85	2200	~3000	4000-6000
Acoustic impedance Z_A [MRail]	0.028	3.9	6.5	25-37
$FOM = 10^4K^4/ Z_A{}^2$	165	0.19	42	~0.6
Operating temp. [°C]	-20…+50	...+80	…+150	…+300

Tab. 2. Properties of piezoelectric materials in Comparison, *Values of the piezoelectric coefficient from literature.

For the air-coupled ultrasonic applications, two parameters play the major role: the coupling factor and the acoustic impedance. Coupling factor K defines an effectivity of the energy transformation by the ultrasonic transducers:

$$K^2 = \Delta W/W, \tag{3}$$

where ΔW is a transformed energy and W is an applied energy. Acoustic impedance Z_A defines the transmitting energy attenuation because of the impedance mismatch between the transducers and air. The sensitivity of the pair of air-coupled ultrasonic transducers can be estimated by the ratio W_2/W_1, where W_1 is the electric energy (or power) applied to the transmitter and W_2 is the electric energy measured at the receiver. Considering two processes of energy transformation and two transducer-air interfaces leads to

$$W_2/W_1 = K^2T_1T_2K^2, \tag{4}$$

where T_1 and T_2 are the transducer-air and air-transducer transmission coefficients. In the case of normal sound incidence we obtain

$$T_1 = 4\, Z_A Z_{air}/(Z_A + Z_{air})^2 \quad \text{and} \quad T_2 = 4\, Z_{air} Z_A/(Z_{air} + Z_A)^2, \tag{5}$$

$$\text{as far as} \quad Z_A >> Z_{air} \qquad \text{and}\; W_2/W_1 \approx 16\, K^4 Z_{air}^2/Z_A^2. \tag{6}$$

Consequently, the value of K^4/Z_A^2 characterizes efficiency of piezoelectric materials in the air-coupled ultrasonic applications and can be used as a figure of merit:

$$\text{FOM} \;=\; 10^4 K^4/Z_A^2, \tag{7}$$

where Z_A is in [M_{Rayl}]. Low coupling factor complicates the application of EMFIT film, but taking into the account the figure of merit (i.e. both important parameters, K and Z_A), EMFIT film has advantages in comparison with piezoelectric polymer film (PVDF), composites or ceramics (see Table 1).

Both material and thickness of the electrodes influence the electromechanical properties of the EMFIT film (Figures 1, 2). By increasing the electrode thickness from 100 nm to 300 nm results in a decreasing resonance frequency, in a diffusing and reducing the resonance anomalies and in decreasing of the normalized mechanically clamped capacitance C^S. This is valid for both Al electrodes and Cr/Al electrodes. The change of the normalized mechanically clamped capacitance C^S reflects (according to the equation (1)) the change of the coupling factor K_{33}. Samples (2) and (4) have the same C^S as the films without electrodes and corresponds to the coupling factor K_{33} = 0.05. In sample (3) the coupling factor reduces down to K_{33} = 0.04 and even to K_{33} = 0.032 in sample (5). In the case of sample (1), the normalized mechanically clamped capacitance C^S and the coupling factor are the same as for the films without electrodes (K_{33} = 0.06). When embedding the EMFIT films in a sensor we observed a further decrease of the resonance frequency. We then also find diffused and reduced resonance anomalies (Fig. 1, left), but the clamped capacitance C^S does not change. Consequently, being embedded in the sensors, EMFIT films with thin evaporated electrodes, samples (1), (2) und (4), remain the value of coupling factor K_{33} = 0.05 − 0.06 characteristic for the free films without electrodes.

For the first use in an air-borne ultrasonic experiment we also built a simple NDE-measurement setup, in which we proved high sensitivity and high spatial resolution of our EMFIT transducers. The air-coupled ultrasonic pulse-echo experiment (Fig. 4) was carried out with a step wedge sample. Measuring the delay time we found a good signal-to-noise ratio and a good spatial resolution. Quantitative NDE investigation of the air coupled systems' performance will follow.

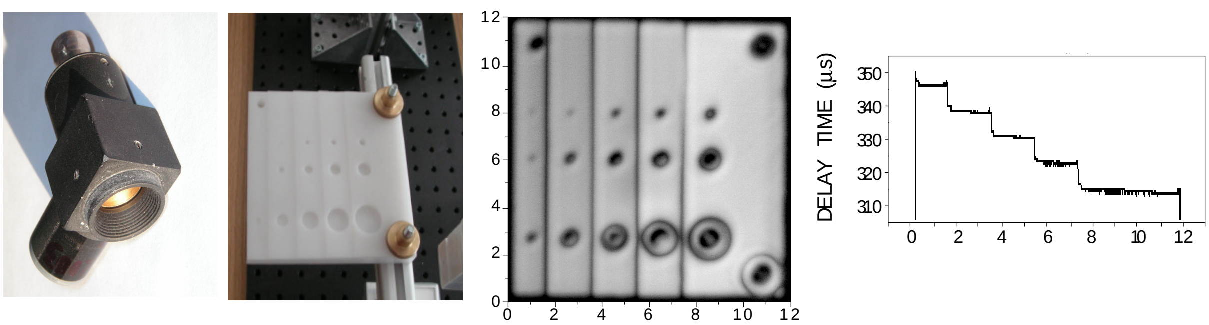

Fig. 2: From left to right: EMFI transducer, sample under test (step wedge), 2D-scan (scanning area is 12 cm x 12cm) as function of delay time, line scan of delay time shows sharp steps.

4 CONCLUSIONS

Our impedance spectroscopy study of the commercially available EMFIT films reveals useful electromechanical properties the air-coupled ultrasonic applications: high-frequency electromechanical resonance (thickness mode above 500 kHz), very low acoustic impedance $Z_A = 0.028 - 0.040\ M_{Rayl}$, low dielectric and mechanic losses, absence of dielectric dispersion out of electromechanical resonance. Low coupling factor $K_{33} = 0.06$ complicates the application, but taking into account both important parameters, K_{33} and Z_A, EMFIT films has advantages in comparison with PVDF films or piezoelectric composites. High frequency ultrasound sensors (transducers), based on the EMFIT films with thin evaporated Al- and Au electrodes, were developed and tested. Although we used a simple NDE measurement setup, high sensitivity of the EMFIT transducers was proved in air-coupled ultrasonic pulse-echo experiments.

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