Piezo-electric properties of polypropylene laminates with a non-woven layer

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Abstract. In the paper piezoelectric properties of non-uniform dielectric structures with elastic layers made from unwoven fabric are presented. Piezoelectric properties of the samples were studied through measurement of piezoelectric $d_{33}$ coefficient, and carried out in dependence on static pressure. The characteristics $d_{33}=f(p)$ obtained for the laminates were hyperbolic type. In order to elucidate this kind of relationship, a theoretical model has been proposed. Based on the new model, the calculated $d_{33}$ piezoelectric coefficients showed good agreement with the experimental results. According to the model developed, the $d_{33}(p)$ dependence is related to the changes of the unwoven structure elastic modulus $Y$ under the influence of static pressure. The magnitude of this change is strongly influenced by the structure of the non-uniform layer, including the fiber diameter, the number of fibers and their arrangement, and the fiber elasticity modulus.

1. Introduction
During the last years non-uniform materials with foamed or voided structure became the subject of intense research due to their advantages over well-known piezoelectric polymers and ceramics [1]. Flexible cellular polymer films and multi-layer structures may exhibit very high value of a piezoelectric coefficient $d_{33}$ that is one order of magnitude higher than that of reported for traditional ferroelectric polymers like PVDF and comparable to the values obtained for the best ceramic materials [2, 3]. In terms of small loads layered structure can reach a value of $d_{33}$ even above 1000 pCN$^{-1}$.

Analysis of piezoelectric properties of electromechanical films, as well as of nonuniform layered structures made of dielectrics having very good electret properties, indicates the possibility of using them in the construction of various types of sensors, electro-acoustic and electro-mechanical transducers used for example to convert ambient energy into electrical energy (the harvesters) [4]. Except of very good piezoelectric properties such materials are characterized by the low cost, thus they can be the basis for the development of large-area transducers.

The possible applications of non-uniform structures [5] indicate that they require determining the piezoelectric coefficient dependence on the applied pressure. This relationship was investigated during studies reported in the presented work.

Studies on the nature of the piezoelectric phenomenon occurring in various materials have shown that it may be the result of several mechanisms [6, 7]. Among them it is distinguished piezoelectric effect related to the nonuniformity of the mechanical properties of the dielectric structure, while the

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occurrence of space charge. A model describing piezoelectric effect in non-uniform multilayer structures was discussed previously in the literature [8, 9], but to explain the results presented in the paper was insufficient. Therefore a new model of piezo-active laminate with non-woven layer has been developed.

2. Piezoelectric effect in non-uniform dielectric structures

Let’s consider three-layer non-uniform “sandwich-like” structure, consisted of two solid films with non-woven (fibers) layer between them. The analysed structure is symmetrical and has an arrangement of layers: hard/ soft/ hard. The charge densities at the interfaces hard/ soft are assumed to be opposite sign and equal in module i.e. \( |q_s1|=|-q_s2|=q_s \).

In the case of such laminate subjected to external stresses applied in the normal direction to its surface the piezoelectric coefficient \( d_{33} \) can be expressed by the relation:

\[
d_{33} = \frac{2q_e\varepsilon_2xd}{(\varepsilon_1d + \varepsilon_2x)^2} \left( \frac{1}{Y_1} - \frac{1}{Y_2} \right)
\]

where: \( d \) - thickness of “hard” layer, \( x \) - thickness of “soft” layer, \( Y_1, Y_2 \) - Young's modules of the “soft” and “hard” layers respectively, \( \varepsilon_1 \) - dielectric permittivity of non-woven material, \( \varepsilon_2 \) - dielectric permittivity of polymeric film. Assuming \( Y=Y_1<<Y_2 \), which means that the external stress \( p \) produces strain in the “soft” layer – \( x(p) \) only, and validity of the Hook’s low, in the form:

\[
x(p) = x_0(1 - \frac{p}{Y})
\]

where \( x_0 \) - the initial thickness of the non-woven layer (without pressure applied), the equation (1) can be rewritten in the form:

\[
d_{33}(p) \approx \frac{2q_e\varepsilon_2x_0}{Y\varepsilon_2x_0} \left( \frac{1}{Y} - \frac{1}{Y} \right) = d_{330}F(p)
\]

The analysis of relation (3) leads to conclusion that the piezoelectric coefficient \( d_{33}(p) \) dependence on static pressure \( p \) is determined by the shape of a function \( F(p) \). The course of the reduced value of coefficient \( d_{33} \) (related to the value of \( d_{330} \) when \( p=0 \)) is determined by the course of the reduced value of the function \( F(p)/ F(p=0) \). This relationship is shown in figure 1.

![Figure 1](#)

**Figure 1.** Plot of the \( F(p)/ F(p=0) \) function depending on the \( p/Y \) reduced pressure value.
3. Experimental
The samples of the three-layer structure were prepared according to the scheme depicted in figure 2.

![Figure 2. Schematic view of structure with non-woven PP layer.](image)

A non-woven polypropylene (PP) placed between two solid polypropylene films was applied as a "soft" layer. External sides of both foil-solid polymer layers were metalized with a thin layer of aluminium. The model laminate structures were prepared with different thicknesses of the "soft" - non-woven layer, ranged from 0.40 to 3.00 mm. Solid dielectric layers were made of polypropylene 25 μm thick films. The structures were made using thermal or ultrasonic welding. All of laminates exhibited good mechanical properties.

The laminate samples were electrically activated by application of breakdown method. The structures were polarized using different values of polarisation voltage. During polarization process, carried out in high electric fields, partial discharges occurring in air spaces lead to the formation of space charge distributed in the non-woven layer in a required manner, i.e. according to the assumption \(|q_{s1}|=-|q_{s2}|=q_s\). Polarization time was kept constant and equal \(t_p=300\pm10\) s. All samples were charged-polarized at the ambient temperature \(T=25\pm2^\circ C\) and relative humidity \(RH=55\pm10\%\). Piezoelectric properties of manufactured structures were investigated by measurements of the piezoelectric coefficient \(d_{33}\) with quasi-static method.

![Figure 3. Influence of static load on \(d_{33}\) piezoelectric coefficient for tested structure and theoretical calculation.](image)

The dependence of piezoelectric coefficient \(d_{33}(p)\) on static pressure \(p\) for selected sample of three-layer PP structure is presented in the figure 3. A hyperbolic type of \(d_{33}(p)\) dependence was observed for all obtained laminates, which was not consistent with graph of \(d_{33}(p)\) predicted by equation (3). For explanation this discrepancy a previous described model has been modified as follows.
4. Model of piezo-active structure with non-woven layer

The basic assumption of the new model is the dependence of elasticity modulus of the "soft"-non-woven fabric $Y$ on real contact surface of fibers $S$, given in the form:

$$ Y = \frac{S}{S_0} K_S Y_{pp} $$  \hspace{1cm} (4)

where $Y$ - Young's modulus of the "soft" layer, $Y_{pp}$ - Young's modulus of the fibres material (polypropylene), $S_0$ - the total area of transducer, $S$ - contact area of fibers, $K_S$ - structural factor (dimensionless) depending on the diameter of the fibers, their number and distribution in volume of non-woven material. Additionally it was assumed that:

$$ \frac{S}{S_0} = kp $$ \hspace{1cm} (5)

where $k$ - constant. Validity of dependence (3) for non-woven layers as well as for fabrics was confirmed experimentally [10]. In the case of three-layer symmetrical structure described above and for the previous made assumptions, the expression (1) can be rewritten in the form:

$$ d_{33} = \frac{2q_x \varepsilon_1 \varepsilon_2 x(p)d}{Y[\varepsilon_1 d + \varepsilon_2 x(p)]^2} $$  \hspace{1cm} (6)

From equations (4) and (5) one can get:

$$ Y = kpK_S Y_{pp} $$ \hspace{1cm} (7)

Substitution of expression (7) into (2) and next the result into expression (6) leads to the following equation:

$$ d_{33} = \frac{2q_x \varepsilon_1 \varepsilon_2 x_0 d(kK_S Y_{pp} - 1)}{p[\varepsilon_1 d kK_S Y_{pp} + \varepsilon_2 x_0 (kK_S Y_{pp} - 1)]^2} $$ \hspace{1cm} (8)

Taking into account that in the above equation, all parameters except $p$ are constant this relation can be written in the form:

$$ d_{33} = A \frac{1}{p} $$ \hspace{1cm} (9)

where

$$ A = \frac{2q_x \varepsilon_1 \varepsilon_2 x_0 d(kK_S Y_{pp} - 1)}{[\varepsilon_1 d kK_S Y_{pp} + \varepsilon_2 x_0 (kK_S Y_{pp} - 1)]^2} $$ \hspace{1cm} (10)

Assuming, that for the applied non-woven structures following relationship is fulfilled [10]:

$$ kK_S Y_{pp} >> 1 $$ \hspace{1cm} (11)

Equation (8) can be described by:

$$ d_{33} \approx \frac{2q_x \varepsilon_1 \varepsilon_2 x_0 d}{pkK_S Y_{pp} (\varepsilon_1 d + \varepsilon_2 x_0)^2} $$ \hspace{1cm} (12)

As can be seen from the relation (12) for the three-layer structure with non-woven fabric, for which all of the stated assumptions are met, it is expected hyperbolic dependence $d_{33}(p)$. To verify the resulting model, the calculations of piezoelectric coefficients $d_{33}$ were performed for the structure characterized by the following data: PP film thickness $d=2.5 \times 10^{-5}$ m; the initial thickness of the non-woven fabric (without static pressure applied) $x_0=4.0 \times 10^{-4}$ m; dielectric permittivity of "soft" layer material (PP non-woven) $\varepsilon_1=1.5$; dielectric permittivity of PP foil $\varepsilon_2=2.1$ the effective surface charge density...
$q_i = 2.7 \times 10^{-5} \, \text{Cm}^{-2}$. Calculations carried out for piezoelectric coefficient $d_{33} = 128 \pm 1 \, \text{pCN}^{-1}$, measured for static force $F=4.9 \, \text{N}$, applied to the total sample area $S=3.1 \times 10^{-4} \, \text{m}^2$ allow (see expression (12)) to determine value of the component $kK_s Y_{PP}$. For given data it was equal 1.48. The obtained value of $kK_s Y_{PP}$ was constant for a given sample (it was connected with its structure). Subsequently piezoelectric coefficients $d_{33}$ for various static pressure were calculated and plotted as the $d_{33}(p)$ dependence. Results obtained from measurements and theoretical calculations are presented in the figure 3. It can be seen that both of plotted curves are almost the same. These results indicate that the developed model of three-layer piezo-active structure with non-woven layer as well as all assumptions are appropriate.

5. Conclusions
Piezoelectric properties of three-layer laminates with non-woven PP layer were investigated. The results of measurements and calculations made on the basis of a new model proposed for the tested structures lead to the following conclusions:

- The tested non-uniform layered structures showed a large piezoelectric coefficient $d_{33}$ (up to several hundred pCN$^{-1}$), strongly dependent on the applied static pressure.
- For all prepared laminates experimentally obtained $d_{33}(p)$ dependences were of a hyperbolic type.
- The new proposed model of piezo-active structure with a soft layer made from non-woven fabric showed good agreement with the experimental results.
- In accordance with the model developed, the $d_{33}(p)$ dependence is related to the changes of elastic modulus $Y$ of unwoven structure under the influence of static pressure. The magnitude of this change is strongly affected by the structure of the non-uniform layer (the fiber diameter, the number of fibers and their arrangement, fiber elasticity modulus, which are given as $kK_s Y_{PP}$ component).

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