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Dependence of the piezoelectric coefficient d_{33} on the static pressure for non-uniform polymeric layered structures

Streszczenie. *W pracy przedstawiono model opisujący zależność współczynnika piezoelektrycznego $d_{33}(p)$ od przyłożonego ciśnienia p dla niejednorodnych dielektrycznych struktur warstwowych. Wcześniejsze badania wykazały, że charakter zmian $d_{33}(p)$ dla wymienionych struktur może być określony zależnością współczynnika sprężystości warstwy miękkiej $Y_1(p)$ od obciążenia. Przedstawiony model dotyczy struktury zawierającej warstwę elastyczną w postaci włókniny, dla której zależność współczynnika sprężystości może być opisana modelem odkształcania włókien. Analiza modelu wskazuje, że zależność $d_{33}(p)$ można opisać funkcją potęgową typu $d_{33}(p)=Ap^n$. Potęgowy charakter zależności $d_{33}(p)$, z wartością -0.20 < n < -0.12 potwierdzono doświadczalnie.* ***Zależność współczynnika piezoelektrycznego d33 od obciążenia statycznego dla niejednorodnych struktur polimerowych.***

Abstract*. The paper presents a model describing dependence of the piezoelectric coefficient $d_{33}(p)$ on the pressure p applied to heterogeneous layered dielectric structures. Previous studies have shown that the $d_{33}(p)$ dependence for such structures may be determined by the pressure dependence of the elasticity modulus $Y_1(p)$ of a soft layer. The model is related to an elastic layer comprising a nonwoven, for which the $Y_1(p)$ dependence can be described by the deformation of individual fibers being in the mutual contact. Model analysis shows that the $d_{33}(p)$ dependence can be approximated by a power-type function: $d_{33}(p) = Ap^n$. The power type of the $d_{33}(p)$ dependency, with power-factor -0.20 < n < -0.12 was confirmed experimentally.*

Słowa kluczowe: efekt piezoelektryczny, dielektryki niejednorodne.
Keywords: piezoelectric effect, heterogeneous dielectrics, layered structures, piezo-laminates.

Introduction

The piezoelectric effect appearing in dielectric solids can be described by different physical mechanisms [1-3]. One of them is non-homogeneous deformation of the dielectric containing a built-in space charge [4, 5]. The non-homogeneous deformation may result from non-uniform stress as well as from non-uniformity of mechanical properties (e.g. elasticity modulus) of the material. The last opportunity leads to a construction of laminated structures combining "soft" and "hard" dielectric layers, which simultaneously exhibit sufficiently good electret properties. Practical realization of the mentioned structures is difficult, because high elasticity polymers usually do not exhibit sufficiently good electret properties. Nowadays the problem is solved by application of "dispersed" polymeric dielectrics, applied earlier in technology of electrets. One can distinguish two types of "dispersed" or porous dielectric structures which can be applied in technology of piezo-active laminates, namely: 1. Dielectric foams (containing usually oriented gas voids) [5] or 2. Fibers structures (both, woven and non-woven) [10]. In both of the mentioned cases, the "soft" layer can be treated as the composite dielectric material, consisting of the basic polymer "dispersed" in the air. The way, the basic polymer is "dispersed" in the air, influences (in a basic manner) the value of piezoelectric coefficient as well as it dependence on the applied mechanical load. A model of laminate containing a non-woven layer (as the "soft" one) is considered in the paper and the deformation of individual fibers being in the mutual contact was found to be responsible for the power-type dependence of the piezoelectric coefficient on the static load.

Piezoelectric properties of layered dielectric structures

The fundamental model of the layered, dielectric structure, which should exhibit the piezoelectric properties is shown in Fig. 1. [6].

For the structure shown in Fig. 1, comprising dielectric layers with electrical relative permittivity ε_1 and ε_2, thicknesses x_1 and x_2, elasticity moduli Y_1, Y_2 and the bipolar

Fig. 1. Model of a layered dielectric structure containig charges with densities of $+q_s$ and $-q_s$ on the interfaces between the layers

charge with density $+q_s$, and $-q_s$, spread on the interfaces between layers (1) and (2), the piezoelectric coefficient d_{33} can be determined from the formula [7]:

$$d_{33} = \frac{2q_s\varepsilon_1\varepsilon_2 x_2 x_1}{(\varepsilon_1 x_2 + \varepsilon_2 x_1)^2}\left(\frac{1}{Y_1} - \frac{1}{Y_2}\right). \quad (1)$$

Assuming, that the elasticity coefficients Y_1, Y_2 for the layers (1) and (2), respectively, satisfy the conditions:

$$Y_1 << Y_2 , \quad (2)$$

what means that the layer (1) is "soft" and the outer layers (2) are "hard", and additionally the thicknesses x_1, x_2, are in relation:

$$x_1 >> x_2 , \quad (3)$$

the equation (1) can be simplified to the form:

$$d_{33} \cong \frac{2q_s\varepsilon_1 x_2}{\varepsilon_2 x_1}\frac{1}{Y_1} . \quad (4)$$

The "soft" layer can be realized in the form of air-polymer composite as it take place in a case of foamed or non-woven (fibers) layers. In case when the "soft" and "hard" layers contain the same solid polymer, the relative permittivity ε_1 of the "soft" dielectric layer can be roughly determined from the relation [8]:

$$\varepsilon_1 = \varepsilon_a + (\varepsilon_2 - \varepsilon_a)W_p , \quad (5)$$

where the polymer content in the "soft" layer W_p (taken as the polymer fiber-air composite) can be determined from the relation:

(6) $$W_p = \frac{g_w}{x_1(1-\frac{p}{Y_1})\cdot\rho_p} = \frac{W_{p0}}{1-\frac{p}{Y_1}}$$

where g_w [kg/m2] – is the mass of the 1sq. meter of the "soft" layer material (nonwoven) (specific mass), x_1 - initial thickness of the "soft" layer (not loaded), ρ_p [kg/m^3] - is the mass density of the polymer, the non-woven is made of; W_{p0} – is the polymer content for the non-loaded "soft" layer.

Combining of expressions (4) to (6) under assumptions, that the applied pressure $p << Y_1$ and the same basic polymer of both of the layers (the "soft" layer – with a thickness x_1 and solid - with a thickness x_2 are made of the same basic polymer with relative permittivity ε_2) leads to the following relation for the d_{33} coefficient:

(7) $$d_{33} = \frac{2q_s x_2\left[1+(\varepsilon_2-1)W_{p0}\right]}{\varepsilon_2 x_1}\frac{1}{Y_1}$$

The "soft" layer, with a modulus Y_1, can be realized in the form of a fibrous or foamed layer [9,10]. Expression (7) indicates that the $Y_1(p)$ dependence will substantially define the relationship $d_{33}(p)$. The $d_{33}(p)$ dependence determines a practical use of the whole structure. The $Y_1(p)$ dependence is determined by the physical mechanism describing the "soft" layer deformation process. The last one is closely related to the structure of the "soft" layer (fibrous, foamed, etc.)

Model of nonwoven layer

In a considered simplified nonwoven "soft" layer the fibers with circular cross-section are distributed regularly as it is shown in Fig. 2. Particular fibers with diameter φ are distributed in the layer at a distance d from each other.

The force F applied to the total surface of the whole structure S results in:
- deflection of fibers on a distance d - for small stresses,
- compression of fibers and strain of their cross section - for higher stress.

In both cases the elasticity modulus of the basic polymer, the "soft" layer is made of, was assumed to be the same like that of the solid dielectric layer and equal to Y_2.

Value of the effective elasticity modulus Y_1 for the particular polymer–air "composite" (distributed dielectric – a foam like or a non-woven layer) can be determined from the basic equation:

(8) $$Y_1 = \frac{x_1}{\Delta x_1}p$$

Where Δx_1 is a strain of the x_1 layer due to the application of the pressure p.

It can be shown that for the elastic deflection of fibers in the structure, like that in fig 2. (i.e. for small stresses), the value of the effective modulus of elasticity of the "soft" layer does not depend on the pressure p [11].

In case of higher stresses, for which the fibers distributed in the volume of the "soft" layer are in a mutual contact, they begin to deform at the contact points. The deformation will finally determine value of the effective elasticity modulus.

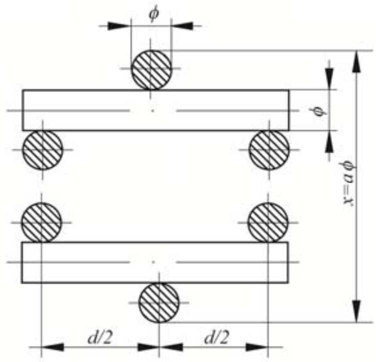

Fig. 2. The Model of nonwoven fabric with regularly distributed fibers. x_1- sample thickness, a - the number of layers, ϕ - diameter of fiber

The process of fibers deformation (at the contact point), can be modeled by deformation of rollers, whose axes of symmetry are twisted by an angle of $\pi/2$ [11] as it is shown in figure 3.

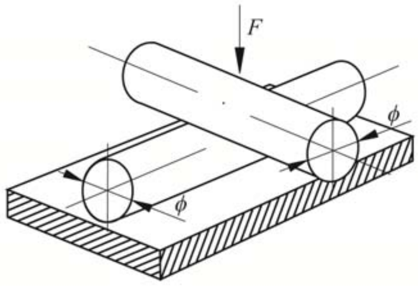

Fig. 3. Model of compression of roller-like fibers with perpendicular axes.

The model includes the following assumptions [11]:
- the roller-type fibers are homogeneous, isotropic and linearly elastic;
- the surfaces of the contact are surrounded by a smooth, and regular curvature (cylinder-roller/flat half-space);
- the diameters of both of rollers are the same and equal to ϕ;

Assuming regular distribution of fibers in the nonwoven structure, the force F acting on the fiber at the contact point can be determined from the relation:

(9) $$F = pd^2$$

where: p – is the average pressure value (in the area equal to $S=d^2$ – surface of the elementary part of the fiber's mesh - as it is shown in Fig. 2); d - average value of the distance between the fibers in a layer. Assuming regular distribution of cylindrical fibers in the layer, d value can be determined from the relation:

(10) $$d = \frac{\rho_p\phi\pi x_1}{4g_w} = \frac{\phi\pi}{4W_{p0}}$$

Deformation of the fiber $\Delta\phi$ for the arrangement shown in Fig. 3 can be described by the relation [10]:

(11) $$\Delta\phi \cong 1{,}2\phi\left(\frac{(1-\nu_p^2)}{W_{p0}^2 Y_2}\right)^{\frac{2}{3}} p^{\frac{2}{3}},$$

where ν_p – is the Poisson factor for the transverse deformation of the roller due to the longitudional strain. Assuming elastic deformation of fibers (rollers) at the contact point, the value of effective Young's modulus Y_1 of the "soft" layer can be determined using the following formula:

$$Y_1 = \frac{\phi}{\Delta\phi} p \cong \frac{\left(W_{p0}^2 Y_2\right)^{\frac{2}{3}} p^{\frac{1}{3}}}{1{,}2(1-\nu_p^2)^{\frac{2}{3}}} \qquad (12)$$

Substituting the equation (12) into (7) one can finally obtain:

$$d_{33} \cong A p^{-\frac{1}{3}} \qquad (13)$$

where:

$$A \cong \frac{2{,}4 q_s \varepsilon_1 x_2}{\varepsilon_2 x_1} \frac{(1-\nu_p^2)^{\frac{2}{3}}}{\left(W_{p0}^2 Y_2\right)^{\frac{2}{3}}} \qquad (14)$$

Expression (13) shows a power type dependence of the d_{33} coefficient on the pressure p with the exponent n = -1/3.

Samples

Investigation of the piezoelectric coefficient d_{33} dependence on the static pressure p, were performed on samples with a three layer structure, as it is shown in Fig.1. The structures includes an elastic layer made of the pneumo-thermal polypropylene non-woven sandwiched between two layers of polypropylene film. Thickness of the nonwoven fibers layers, and their specific mass are collected in Table 1. In each case, thickness of the outer film layers having was equal to x_2=26 µm. The whole structure was subjected to thermal welding with the welding point density equal to about 13 dots/ cm^2. The cross-section structure in a region between welding points is shown in Fig. 4. Structures with dimensions of about 25x25 mm were

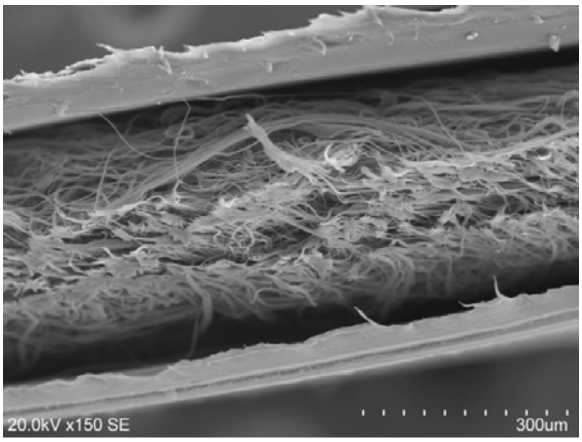

Fig. 4. The PP film - nonwoven PP - PP film sandwich in the region between welding points.

equipped with electrodes placed concentrically on both sides of the structure. Electrodes with diameter of 18 mm were made of colloidal graphite. The samples equipped with the electrodes were subjected to a formation process. The formation process was carried out at room temperature and under normal pressure for selected voltage (electric field) and time. During the activation process electrodes deposited on the structures were connected to dc high voltage power supply through the high voltage limiting resistor (1 MΩ/10 kV) and polarized by the voltage U_p applied within the time t_p.

Table 1. Characteristic parameters of nonwoven layers

Sample	g_w [g/m^2]	x_1 [mm]	W_{p0} [-]
P.1.	25	0.30	0.093
P.2.	15	0.21	0.079
P.3.	60	0.75	0.089
P.4.	45	0.55	0.091
P.5.	30	0.35	0.095

Measurements of piezoelectric coefficient d_{33}

Measurements the piezoelectric coefficient d_{33} were performed using static method [3] illustrated in Fig. 5. The sample (1) was placed between the fixed and rigid measuring electrode (2), located on an insulator (3) and the grounded, movable electrode (4). Voltage measurements were carried out using the electrometer (5) RFT-6302 with an attached measurement capacitor (6). The total capacity of the voltage measurement system (including measurement capacitor, the cable and voltmeter input capacitances and sample capacitance) was equal to C_T=1.57 nF. Whole the electrode system was placed in a grounded Faraday cage (7) for shielding from external electric fields. The controlled stress was applied to the samples using the weights placed on the pan (8), mechanically coupled to the movable electrode (4). Mass of the weights was determined using a quartz scale (9).

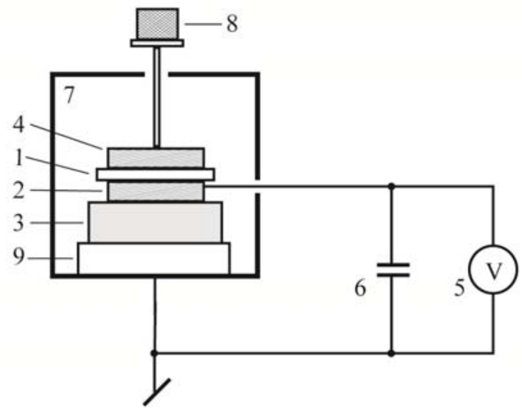

Fig. 5. An arrangement for measuring the piezoelectric coefficient d_{33} using static method (explanations in the text)

Value of the coefficient d_{33} was determined from the relationship:

$$d_{33}(p) \cong S_E C_T \frac{\Delta U(p)}{\Delta mg} \qquad (14)$$

Where C_T - is the total capacity of the voltage measuring system, $\Delta U(p)$ - the voltage change caused by changing mass of the load Δm, S_E - surface of the measuring electrode (part of the sample surface subjected to the stress), g - gravity.

Results

Results of d_{33} coefficient measurements, obtained for samples activated in different conditions – in different polarization voltage are shown in Fig. 6. The obtained $d_{33}(U_p)$ dependency allows to optimize activation (polarization) conditions, in particular polarization voltage U_p, and indicate possibility of appearing of two mutually competing phenomena during the activation process. One of them is a process of partial discharges occurring in gas voids (present in non-woven layer) which is observed for voltages higher than the threshold voltage, which in case of investigated structures was in the range of 2 - 3 kV (see

Fig. 6). The increase of polarization voltage leads to sharp increase in the intensity of partial discharges and the accompanying increase in effective density of the surface charge q_s on the inner surface of gas voids. The process intensifies until the state in which partial discharges will cover all the voids, what can be observed on $d_{33}(U_p)$ dependency as a "saturation" effect. The second process, which can lead to a decrease in the value of d_{33} coefficient for higher voltages U_p, is associated with increasing probability of the electrical breakdown of the thin dielectric layer surrounding the whole gas void and thus a reduction in the effective charge density q_s and finally with the maximum appearance on the $d_{33}(U_p)$ dependency.

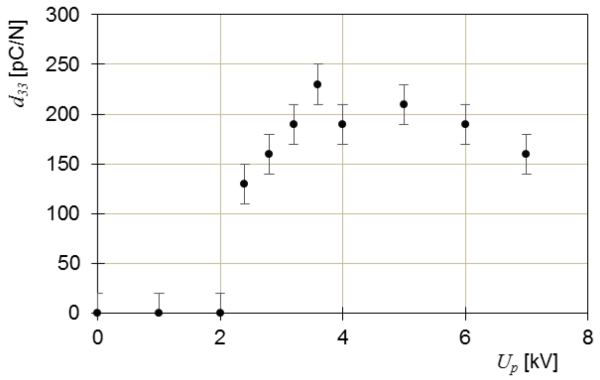

Fig. 6. Dependence of d_{33} coefficient on the activation voltage U_p for the structure P.1. (see Tab. 1)

The dependence presented in Fig. 6 was a typical for all of investigated structures and allow to determine the activation conditions. Structures for further research were activated at a voltage U_p = 5.5 ± 0.2 kV, within the time t = 30 ± 0.2 s. Results of d_{33} coefficient measurements obtained for structures described in Tab. 1. are shown in Fig. 7. The results obtained for the additional static load to be changed in the range 4 - 60 kPa showed a power-type $d_{33}(p)$ dependency, what confirms applicability of the presented model. However, the experimentally determined value of the exponent n for all of tested structures was

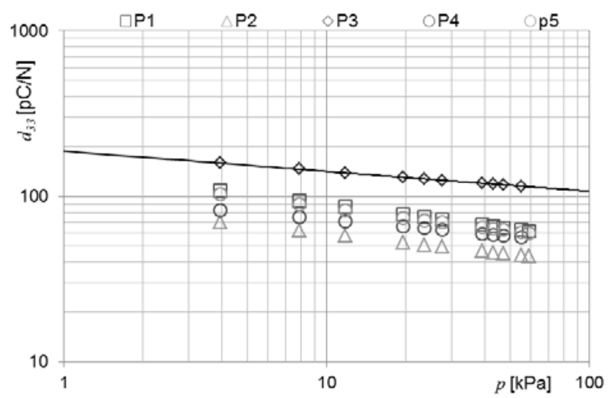

Fig. 7. Theoretical - a solid line, and experimental - measuring points) dependencies of d_{33} coefficient on the pressure p.

found to be on the level n=-0.16±0.04, and deviates from the value determined by the model (n=-0,33 - see Eq. 13).

Summary

Results of measurements and considerations concerning the described and investigated piezo-active structures allow to formulate the following conclusions:

- the presented theoretical model, describing $d_{33}(p)$ characteristic, (considering deformation of fibers under external stress) leads to a power-type dependence with the exponent value n = - 0.33;
- experimental studies confirmed the power-type of the $d_{33}(p)$ dependence, but the value of the exponent n was found to be less than that determined by the model, and was in the range - 0.20< n<- 0.12;
- lower values on the n-factor may result from other dependency of the permittivity ε_1 on the pressure p than the given by expressions (5) and (6);
- the value of d_{33} coefficient seems not to follow the dependence on the W_{p0} and x_1 values - as it is required by Eq. (14). It may suggest that the values of surface charge density q_s (in gas voids) may also depend on the same parameters;
- the model developed for the elastic layer with regularly distributed fibers (of the same diameter), seems to properly describe the $d_{33}(p)$ dependency for the real structure, i.e. structure with chaotic arrangement of fibers and the distribution of their diameters.

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REFERENCES

[1] Wada Y., Hayakawa R., *Piezoelectricity and Pyroelectricity of Polymers*, Jap. Journ. of Appl. Phys., 15 (1976), No. 11, 2041.

[2] Hilczer B., Malecki J., *Electrets*, Studies in Electrical and Electronic Eng., Vol. 14, Elsevier, Amsterdam, 1986, p. 166

[3] Mort J., Phister G., Electronic Properties of Polymers, Ed, J. Wiley & Sons, NY 1982.

[4] Sessler G.M., Hillenbrand J. *Electromechanical Response of Cellular Electret Films.* Proc. 10th Int. Symp. on Electrets, 22-24, Sept. 1999, Delphi, Greece, p. 261-264.

[5] Gerhard-Multhaupt R. *Less can be more. Holes in polymers lead to a new paradigm of piezoelectric materials for electret transducers*. IEEE Trans. Diel. Electric. Ins. 2002; v.9: p. 850-859.

[6] Kacprzyk R., Kisiel A., *Piezo-electric properties of polypropylene laminates with a non-woven layer,* J. Electrostatics 71 (2013) p.400-402.

[7] Kacprzyk R, Motyl E, Gajewski J B, Pasternak A. *Piezoelectric properties of non-uniform electrets*, J. Electrostatics. 1995; v. 35: p.161-166.

[8] Nelakanta P.S. *Handbook of Electromagnetic Materials*. CRC Press, Boca Raton, NY, 1995.

[9] R. Kacprzyk, *Piezoelectric Properties of Non-uniform Electrets,* Proc. IEJ-ESA Joint Symp. on Electrostatics, 25-26 Sept. Kyodai Kaikan, Kyoto, Japan (2000) pp. 108-115.

[10] R. Kacprzyk, *Non-conventional application of unwoven fabrics.* J. Electrostatics, 56 (2002) p.111-119.

[11] Johnson, K. L, *Contact mechanics*, Cambridge University Press, 1985, p.107-152.