Low pressure and time storage influences on the electrets stability of HDPE composite films

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Abstract. The influence of both low pressure and time storage on the surface potential decay of high density polyethylene (HDPE) composite films with different weight concentrations of the zeolite particles - 0 wt.%, 2 wt.% and 6 wt.% were studied. The samples were charged in a corona discharge by means of a corona triode system for 1 minute under room conditions. Positive or negative 5kV voltage was applied to the corona electrode and 1 kV voltage of the same polarity as that of the corona electrode was applied to the grid. After charging, the electret surface potential was measured by the method of the vibrating electrode with compensation. Two groups of tests were performed. In the first group after charging, the electrets were placed into a vacuum chamber where the pressure was reduced step by step in the range from 1000 mbar to 0.1 mbar. At each step the samples were stored for 1 minute. Then the electrets were removed from the vacuum chamber, the surface potential was measured again and the normalized surface potential was calculated. It was established that the low pressure had led to the surface potential decay of the electrets. The influence of the low pressure was analyzed by the equation that describes processes of desorption from the electret surface accompanied with surface diffusion. In the second group after charging, the electret surface potential was measured with the time of storage for two months. The experimental results obtained show a significant change in the electret behaviour of the composite films after the incorporation of zeolite particles with different concentration into the HDPE matrix. It was established that the surface potential decay depends on the corona polarity and the particle concentration.

1. Introduction

Considerable scientific interest has been shown in the surface potential decay of corona charged polymeric materials such as polypropylene (PP), polyethylene (PE), polyethyleneterephthalate (PET) and etc. that has been stimulated by industry demand related to the adjustment of Xerox-photography processes, assessment of cable insulation, and evaluation of electrostatic risks or monitoring of aged insulators [1-3]. Over the last few years, however, together with these materials another relatively new direction has been developed. It is the investigation of different composite electret films with specific properties obtained by the incorporation of different particles into polymer matrices [4]. Polymer zeolite composites have attracted wide interest because of enhancing polymer properties and extending their applications [5]. In [6] composites of low-density polyethylene (LDPE) and zeolite H-b particles at different zeolite contents up to 10 wt% were investigated. It was established that after incorporating
zeolite H-b particles, the polyethylene composite films exhibit considerable degradation in mechanical properties and they have shown a good dispersion of zeolite particles in the PE matrix. Lopes and co-authors [7, 8] have investigated dielectric properties of composite polymers of α-PVDF doped with different NaY zeolite particles. It was established that dielectric properties and conductivity can be tailored by zeolite inclusion in polymer matrix and the presence of the zeolite particles increases ac conductivity of the matrix.

In the literature, however, there are no data available about the influence of the incorporation of zeolite particles on the charge decay of electret polymer films at different conditions. The aim of the present paper is to investigate the influence of both low pressure and time storage on the surface potential decay of HDPE composite films with different concentrations of the zeolite particles.

2. Experimental

2.1. Preparation of the composite films
The high density polyethylene (HDPE) brand PE2NT11-285D and the zeolite particles with a density of 2.4 g/cm³ and a particle diameter of less than 50 μm were used. Mixing of the polymer with the zeolite particles had been carried out on the mixer (Brabender Plastograph EC Plus) for 7 minutes at a temperature of (170 ± 5) °C. Polymer composite samples were prepared as 250 μm films under a hydraulic press at a temperature of (170 ± 5) °C and holding time of 5 min.

2.2. Corona treatment and surface potential measurement
Samples of 30 mm diameter were cut from the HDPE composite films. Charging of the samples in a corona discharge was carried out by means of a conventional corona triode system (figure 1) consisting of a grounded plate electrode, a corona electrode and a grid placed between them. The distance between the corona electrode and the grid was 10 mm and the distance between the grid and the grounded plate electrode was 3 mm. The samples were charged for 1 minute under room conditions. The voltage of the corona electrode was ±5 kV. The electret surface potential was limited by the grid potential, which was set at 1 kV with the same polarity as the corona electrode voltage.

![Figure 1](image)

Figure 1. Scheme for obtaining electrets: 1. high voltage source; 2. corona electrode; 3. grid; 4. plate grounded electrode; 5. sample on a metal pad; 6. voltage divider.

The electret surface potential was measured by the method of the vibrating electrode with compensation [1] and the estimated error was better than 5%. After charging, the initial surface potential \( V_0 \) was measured and the electrets were divided in two groups according to the kind of treatment – different low pressure and time storage.

2.3. Low pressure treatment
After charging the samples, the initial electret surface potential \( V_0 \) was measured and then the electrets were placed into a vacuum chamber. The pressure in the vacuum chamber was reduced step by step in the range from 1000 mbar to 0.1 mbar. At each step the samples were held at respective
constant pressure for 1 minute. After that the electrets were removed from the vacuum chamber, the surface potential $V$ was measured again and the normalized surface potential $V/V_0$ was calculated.

3. Results and discussion

3.1. Time storage influence on electrets surface potential decay

The dependences of the normalized surface potentials $V/V_0$ on the storage time for positively and negatively charged HDPE composite films with different weight concentrations of the zeolite particles (0 wt.%, 2 wt.% and 6 wt.%) had been studied for two months. These dependences are presented in figures 2 and 3 respectively. The surface potential was measured once a week except for the first 20 days when the charge was rapidly decaying. Steady state values of the surface potential were established for all samples investigated after 30 days storage time.

**Figure 2.** Time dependences of the normalized surface potential for positively charged HDPE composite films with different concentration of the zeolite particles.

**Figure 3.** Time dependences of the normalized surface potential for negatively charged HDPE composite films with different concentration of the zeolite particles.
Each point in the figures is a mean value from 6 samples. The calculated standard deviation was better than 5% from the mean value with confidence level 95%.

The results illustrated in figures 2 and 3 demonstrate the following peculiarities:

- For all investigated samples the normalized surface potential are initially sharp decaying exponentially for the first 10 days, then are slowly decreasing to the 30\textsuperscript{th} day and are practically stabilized to the 60\textsuperscript{th} day.

The value of the surface potential of electrets depends on the amount of trapped charges in the different localized surface states of the samples. In the initial period of time after the corona charging, the surface potential rapidly decreases due to the release of the weakly captured charges from the shallow energy states. Then the surface potential stabilizes to a steady state value caused by the tightly captured charges in the deep energy traps.

- The steady state values of the normalized surface potential for positively charged HDPE composites are higher than those for negatively charged ones independently of the concentration of the zeolite particles incorporation in the HDPE matrix;

- The normalized surface potential of HDPE composite films with different concentrations of the zeolite particles (2 wt.% and 6 wt.%) decays slower than the one of the pure HDPE films independently of the corona polarity.

Therefore, the incorporation of the zeolite particles in the HDPE matrix independently of the concentration leads to an increase of the electret surface potential value and to obtaining the stable electrets.

For all samples the experimental results fitted very well to an exponential dependence of the type:

\[ y = y_\infty + (1 - y_\infty) \exp\left(-\frac{t}{\tau}\right), \]  

where \( y = V/V_0 \) is the normalized surface potential, \( y_\infty = (V/V_0)_{st} \) is the steady state level which is reached for \( t \gg \tau \) and \( \tau \) is a relaxation time. Equation (1) is analogous to an equation characterizing the kinetics of adsorption-desorption processes [10]. In our case, these processes are not parallel but in series. Firstly, during the corona discharge, an adsorption takes place, as a result of which the accumulated surface charge determines \( V_0 \) at \( t = 0 \). Subsequently, the release of the captured charges from the energy states takes place and its development is described by the second term in equation (1).

Above a certain value of the time (\( t \gg \tau \)), the steady state level of the normalized potential \( y_\infty \) can be determined. Table 1 shows two obtained parameters \( y_\infty \) and \( \tau \) by fitting to the equation (1) and the determination factor R2 for all the samples as well as experimental values of the steady state level \( (V/V_0)_{st}^{exp} \) measured on 60th day.

The very good coincidence between the values of the parameter \( y_\infty \) and the experimental measured normalized surface potential on 60th day \( (V/V_0)_{st}^{exp} \) as well as the determination factor R2

| Concentration of the zeolite | Corona polarity | \( y_\infty \) | \( \tau \) | \( R^2 \) | \( (V/V_0)_{st}^{exp} \) |
|-----------------------------|----------------|----------|------|------|----------------|
| 0 wt.%                      | Positive       | 0.47     | 11.6 | 0.99 | 0.48          |
|                             | Negative       | 0.36     | 10.0 | 1.00 | 0.37          |
| 2 wt.%                      | Positive       | 0.71     | 7.2  | 0.97 | 0.70          |
|                             | Negative       | 0.46     | 11.4 | 1.00 | 0.47          |
| 6 wt.%                      | Positive       | 0.71     | 5.8  | 0.98 | 0.70          |
|                             | Negative       | 0.44     | 10.4 | 1.00 | 0.45          |
that is very close to 1 for all the samples prove an equation 1 true. It is cleaned that relatively weakly-trapped charged is released over tens of days as the relaxation time $\tau$ is around 10 for all samples. Moreover the relaxation time does not depend of the zeolite concentrations for negatively charged HDPE composite films while it decrease with increasing of the zeolite concentrations for positively charged HDPE composite films.

3.2. Low pressure influence on electrets surface potential decay

The dependences of the normalized surface potential $V/V_0$ on the low pressure for positively and negatively charged HDPE composite films with different weight concentrations of the zeolite particles (0 wt.%, 2 wt.% and 6 wt.%) have been studied. Dependences of normalized surface potential on normalized pressure for HDPE composite electrets charged in positive or in negative corona are presented in figures 4 and 5.

**Figure 4.** Dependences of the normalized surface potential on the normalized pressure for positively charged HDPE composite films with different concentration of the zeolite particles.

**Figure 5.** Dependences of the normalized surface potential on the normalized pressure for negatively charged HDPE composite films with different concentration of the zeolite particles.
The symbol $V_0$ in the figures marks the initial value of the surface potential measured just after electrets charging, and the symbol $p_0$ marks the atmospheric pressure. Each point in the figures is a mean value from 6 samples. The calculated standard deviation was better than 10% from the mean value with confidence level 90%.

The results presented in figures 4 and 5 show that:

- Independently of the concentration of zeolite particles three parts are observed in each curve. There are two regions of high and low pressure where the surface potential is constant. For each curve a relatively narrow region of pressures exists where a sharp decay of surface potential occurs. A similar behavior has also been observed in [9].
- The steady state values of the normalized surface potential at a pressure of 0.1mbar for the samples charged in a positive corona are higher than those for the samples charged in a negative corona independently of the zeolite particle concentrations.
- The steady state values of the normalized surface potential at a pressure of 0.1mbar do not depend of the incorporation of zeolite particles in HDPE matrix.

The results obtained (figures 4 and 5) were analyzed by the equation that describes the processes of desorption from the electrets surface accompanied with surface diffusion. This equation (2) was used and described earlier in [11]:

$$\theta = a + \frac{1}{2} b \left( 1 + \text{erf} \left( \frac{x-c}{\sqrt{2}d} \right) \right).$$

Here $\theta = V/V_0$ is the normalized surface potential, $x = \log p/p_0$ and $a$, $b$, $c$ and $d$ are parameters. The parameter $a$ is the minimum value of the normalized surface potential, $b$ is the difference between the maximum and the minimum values of the normalized surface potential, $c$ is the midpoint of the pressure range where the surface potential decay occurs and $d$ is the half-width of the range of sharp potential decay.

Values of the parameters $a$, $b$, $c$ and $d$ for HDPE composites charged in a positive or a negative corona to an initial surface potential value of 1000 V are presented in table 2.

The data obtained in table 2 for HDPE/zeolite composite materials could have practical benefits in its use under conditions of reduction pressures. If you know the initial surface potential value $V_0$ of a certain HDPE/zeolite composite material, data from table 2 could be used to determine the pressure range in which there will be a potential sharp decay - from parameter $d$, and the minimum value of the surface potential to which it will decrease under reduced pressure - from parameter $a$.

**Table 2.** Values of the parameters $a$, $b$, $c$ and $d$ obtained by fitting to the equation 2.

| Concentration of the zeolite | Corona polarity | a     | b     | c     | d     |
|-----------------------------|----------------|-------|-------|-------|-------|
| 0 wt.%                      | Positive       | 0.50±0.01 | 0.47±0.01 | -1.28±0.02 | 0.42±0.02 |
|                             | Negative       | 0.42±0.01 | 0.48±0.02 | -1.49±0.05 | 0.63±0.07 |
| 2 wt.%                      | Positive       | 0.47±0.01 | 0.51±0.01 | -1.14±0.02 | 0.46±0.02 |
|                             | Negative       | 0.42±0.01 | 0.52±0.02 | -1.26±0.04 | 0.68±0.05 |
| 6 wt.%                      | Positive       | 0.44±0.01 | 0.50±0.02 | -1.03±0.04 | 0.58±0.05 |
|                             | Negative       | 0.39±0.01 | 0.46±0.01 | -1.29±0.04 | 0.69±0.05 |

**4. Conclusion**

Steady state values of the surface potential of the HDPE/zeolite composite electrets were established for all investigated samples after 30 days storage time. Therefore, this composite electrets material will
be good for practical applications after 1 month when their electrical charge is stabilized.

Our experimental data show that the low pressure at which HDPE/zeolite composite electrets have been stored should be the important factor determining electrets charge decay. This is extremely important, and should be taken into account in the study of the stability of electrets sensors and devices that are used at room temperature and function under real conditions involving various low pressure levels.

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