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Composite Polymeric Materials Containing Electret Components

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The experimental data are discussed concerning the polarization charge effect on physico-mechanical characteristics of polymer composite materials the components of which are in the electret state. The dependences of elastic modulus and electret charge of composites on polarization degree and filler percentage are shown. The introduction of electric polarized fillers into polymer binder leads to electret charge formation in the material and increases the elastic modulus. Thermal treatments of synthetic reinforcing fibers in stationary electric field and in corona discharge field increase the strength of reinforced plastics. The increase of strength depends on the preparation method and specimen treatment in electric field. The increase of oligomers crosslinking during electric polarization was also studied. It is shown that glass-fiber plastic specimens on the polyamide base formed between Cu and Al electrodes, keep their strength characteristics for a long time after the thermal treatment in boiling water. Such a prolonged strength preservation is conditioned by a low water absorption of electret specimens. It is proposed that polymer electrets are suitable formalizing composite materials for structural materials.

KEY WORDS Composites, polymers, electrets, mechanical properties.

INTRODUCTION

The analysis of literature related to electrets, particularly to polymeric electrets^{1–7,18,19} shows the areas for their application in mechanical engineering to be rather limited. Electrets have mainly been used in radio engineering for making microphones, as well as in the instrument-making industry for producing transducers intended to convert mechanical signals to electric ones, e.g., pressure pick-ups, movement pick-ups, vibration pick-ups, deformation pick-ups. Actually, no mention has been made of application of polymeric electrets as engineering materials in machine building. However, there is a large scientific base for a much wider application of electrets.

Bely *et al.*⁴ reported that polymers, if in contact with electrets during thermal treatment, acquire polarization charge, i.e., they become electrets as well. In an earlier work⁸ it was observed strength increases of polymeric composites in which electret

charge had been formed while they were treated thermally between closed electrodes of unlike metals. It was experimentally established⁹ that polymers in the electret state exhibit a reduced absorption of organic solvents vapors.

The strength characteristics of polymeric composites are known to depend not only on the mechanical properties and on the technology of blending of the components, but also on their technological history (e.g., mechanical and chemical treatments, application of physical fields, etc.^{5,10-12}).

The understanding of strength characteristics of polymeric composite materials whose components are in the electret state is, therefore, of a great interest.

2. EXPERIMENTAL PROCEDURE

2.1. Materials

The following polymers were used to conduct the experiments: polyvinyl butyral (PVB), pentaplast (PPI), polyamide (PA). Bakelite lacquer (BL) was used as a thermosetting binder. Fillers were quartz sand (QS) having the particle size between 20 and 60 μm and aluminum oxide (AO) with the particle size between 20 and 30 μm . The reinforcing components were glass fabric (GF) and Terlon fiber (TF) as well as Oxolon fiber (OF).

The fiber was treated by a finish, namely, by aqueous solution of polyvinyl alcohol (PVA).

The polarizing electrodes were made from copper and aluminum.

2.2. Specimens

The specimens for testing the strength characteristics were made from filled thermoplastics in the form of dog-bones having the thickness 1 mm, the design length of the working part 10–40 mm, the total length was 80 mm. The specimens were cut from films obtained by molding mixtures of polymeric powders and fillers heated above the polymer melting temperature.

The specimens intended for determining electret charge in the polymeric material were cut from the films in the form of discs having diameter of 50 mm and thickness of 1 mm.

The specimens from Terlon fiber and those from Oxolon fiber impregnated with the thermosetting binder were made in the form of plaits having the length of 80 mm and the diameter of 1 mm.

2.3. Method for Forming Electret Charge in Polymeric Composites

2.3.1. Metallopolymeric electrets. Metallopolymeric electrets were prepared by thermally treating the specimens made from a composite polymeric material between electrically closed electrodes of unlike metals. Heating was continued until the polarization temperature (T_p) was reached, which exceeded by 5 to 10°C the polymer melting temperature or the glass transition point. The specimens were cooled down to the room temperature while their contact with the electrodes was maintained.

2.3.2. *Composite polymeric materials containing dispersed electret filler.* Polymeric material of this type was prepared by blending a polymeric binder and a dispersed filler possessing electret charge. The filler was polarized in the field of corona discharge having intensity $\bar{E} = 0.5$ to 2.5 MV/m.

2.3.3. *Polarization of thermoset specimens reinforced with OF- and TF-fibers.* In the first version, polarization was carried out by thermally treating the finish-impregnated fiber in a stationary electric field of intensity $\bar{E} = 5.10-3.10$ V/m, or in the corona discharge field of intensity $\bar{E} = 0.5-2.5$ MV/m and subsequent fiber impregnation with bakelite lacquer, and thermal treatment. In the second version, the fiber was first finish-treated, then impregnated with bakelite lacquer. This was followed by thermal treatment in stationary electric field and in corona discharge.

2.4. Methods for Measuring Electret Charge

Electret charge was determined by measuring the charge surface density using the non-contact induction technique, and also thermally stimulated depolarization (TSD) of the specimens by measuring TSD current. The specimens were heated up to the temperature which exceeded the polymer melting point or its T_p .

2.5. Study of Deformational-Strength Characteristics

The deformational-strength characteristics such as ultimate tensile strength (σ_t) and elastic modulus (E) were determined from the measurements of the ultimate tensile force and maximum elongation of the specimens at rupture.

3. EXPERIMENTAL RESULTS

3.1. Effect of Dispersed Filler Charge on Strength

The dependences of the elastic tensile modulus and electret charge on the polarization degree and the filler percentage are given in Figure 1. The other conditions being equal, the strength of specimens containing electrically polarized filler is much higher than that of the reference specimens. The elastic modulus (curve 1), if the filler concentration was 5 wt %, has a distinct maximum, then its magnitude decreases up to values close to those of the reference specimens; this is indicative of an optimal filler concentration. The plot of polarization charge variation, as determined by the TSD technique (curve 3), has a maximum if the filler amounted between 1 and 5 wt %. After such a maximum has been reached, the plot approaches, in an exponential manner, a minimum value as the filler concentration is being increased up to 5 to 10 wt %.

3.2. Strength of OF- and TF-Fiber Reinforced Composites

The thermal treatment of reinforcing synthetic fiber in stationary electric field and in corona discharge field was observed to increase by 1.6–2.0 times the strength of the specimens, as compared to the reference specimens, depending on the fiber ma-

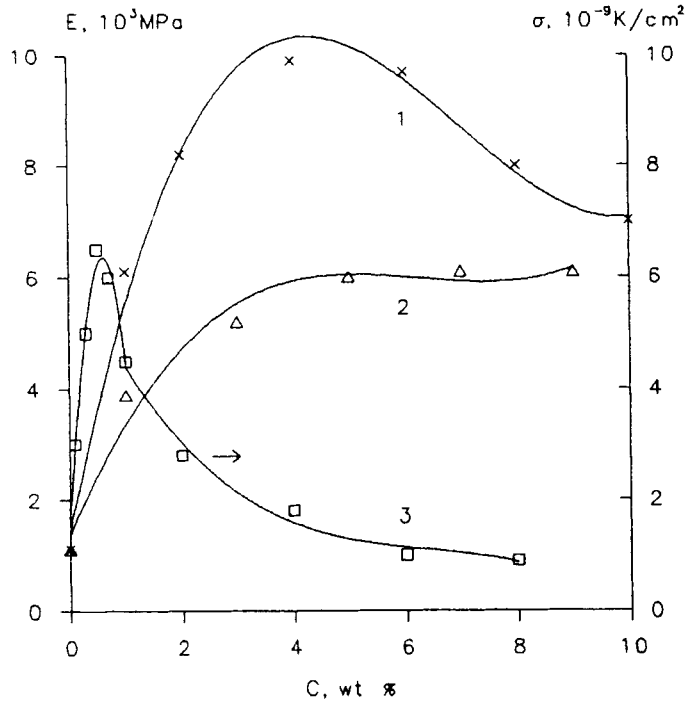


FIGURE 1 Elastic modulus (E , curves 1 and 2) and TSD charge (σ , curve 3) for specimens made from composite material based on PPI filled with Al_2O_3 versus treatment in electric field and the filler percentage. Curves 1 and 3 stand for specimens containing the filler treated in corona discharge, curve 2 stands for the reference specimens.

TABLE I

Strength of specimens reinforced with synthetic fiber depending on treatment process in electric field

Process	σ_p , MPa for different specimens and their production process			
	OF		TF	
	Electric field	Corona discharge field	Electric field	Corona discharge field
1	40.1	47.2	81.7	91.4
2	45.2	53.4	86.3	98.6
Reference specimen		25.8		53.2

terial, the production process, and the specimen's treatment in electric field (Table I).

3.3. Strength of Glass-Fiber-Reinforced Plastics Formed in Contact Between Closed Cu and Al Electrodes

The specimens of the glass-fiber-reinforced plastic based on polyamide possessing electret charge were observed to decrease their strengths to a lower degree, than the reference specimens, after 72-hours exposure in boiling water. For example, the

ultimate tensile strength of electret specimens dropped from 93 to 90 MPa, while that of the reference samples dropped from 54 to 37 MPa.

4. DISCUSSION

The maximum elastic modulus (Figure 1, curve 1) for the situation where the polymeric matrix was filled with aluminum dioxide thermally treated in corona discharge can probably be explained by the following factors: 1) perfection of the supermolecular structure of the polymeric binder owing to the filler as well as to the effect of the filler's electric field on the binder crystallization^{1,6,9,13}; 2) increased adhesion between the polymeric binder and the filler owing to surface activation of the filler particles and a greater potential difference in the double electric layer on the polymer/filler interface.^{7,12,14}

Probably, the factors mentioned above influence, in an additive manner, the properties of the polymeric composite; increasing first the material's strength after increasing the filler concentration, and then decreasing the strength in consequence of local stresses which develop in the specimens through formation of less perfect supermolecular structures in the binder.¹⁵

The maximum on the plot of the TSD charge variation (Figure 1, curve 3) at the filler concentrations between 1.0 and 1.5 wt %, as well as the exponential charge decrease to a minimum as the filler concentration grows, can be explained by the following factors.

1. Charged aluminum dioxide particles added to the polymeric binder lead to a rise in the volume electric charge in the specimen; this charge is composed of i) the charge concentrated on the polymer/filler interface, that is the layer of charge carriers tunneled into the matrix under the effect of a stronger electric field generated over filler's particles' protrusions, and also ii) the charge of the layer of oriented dipoles (residual polarization) bordering on the tunneled layer.¹ The field intensity on particles' asperities can exceed 10 to 500 times the mean intensity, while the depth of volume charge "penetration" was between 0.43 and 16 μm .⁷

2. The TSD charge drop after the maximum can, probably, be explained by the fact that the volume charge in the specimen grows with increasing filler content. Therefore, the charge carriers found in a strong field of the volume charge can leave the specimen at an increasingly lower speed during depolarization, $i(t) \rightarrow 0$.¹ In addition, TSD charge drops owing to the fact that, with greater filler concentrations, the polymer material volume which undergoes polarization/depolarization decreases.

Higher strengths of the microspecimens from OF- or TF-fibers treated in electric field and in corona discharge field are associated both with stronger adhesion interaction of the binder and the filler, as in the previous case, and with higher crosslinking in the oligomer (BL). This was supported by the analysis of the exothermal peaks found on the curves of the differential thermal analysis (DTA).

The degree of solidification was determined from the peak areas' ratio of the electret to the reference specimens (Figure 2). The temperature region of the exothermal peaks is found in the range of 132–200°C and corresponds to the temperature region in which phenol formaldehyde resin undergoes solidification.

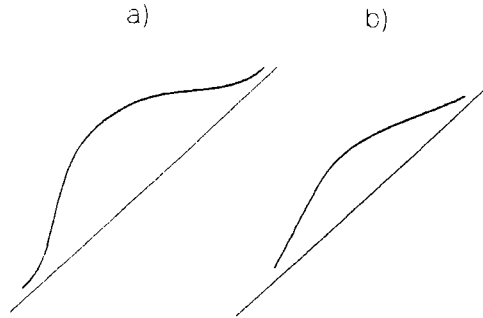


FIGURE 2 Exothermal peaks on the DTA curve for specimens made from BL: a) polarized specimens, b) unpolarized specimens.

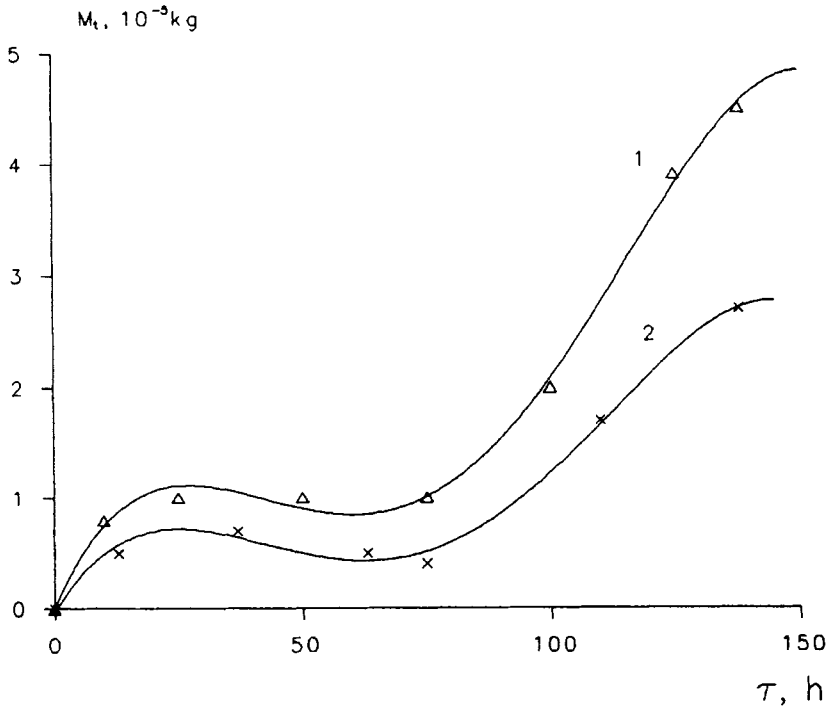


FIGURE 3 Kinetics of water absorption in case of PA-based glass-fiber-reinforced plastics: 1, reference specimens; 2, specimens in electret state.

The experimental results showed that the specimen's crosslinking in the electret state increased by 35 to 40%. This is indicative of the fact that the binder's crosslinking proceeds more fully in case of polarizable specimens than in case of the control, and leads to a higher concentration of the transverse bonds and a higher strength of the composite material.

The lower loss of strength of the glass-fiber-reinforced specimens possessing electret charge, after they have been exposed in water, can be explained by the fact that they do not absorb moisture intensely, as can be seen from the water-absorption curves, Figure 3.

Deterioration of the physical-and-mechanical characteristics in the humidified specimens results from physical and chemical processes such as capillary phenomena, diffusion, hydration, etc., taking place in the materials under consideration. Water molecules (the diameter being 2.76×10^{-10} m, the dipole moment 6.2×10^{-30} C·m) possess high penetrating power. Their high polarity contributes in weakening of the adhesional bonding in the material, violation of cooperative work of the glass-fiber fillers and the polymeric matrix, as well as breaking of weak van der Waals bonds. The decreased strength of the glass-fiber-reinforced plastics accelerates swelling of the polyamide matrix in the course of water molecules' sorption which causes desorption effects if a mechanical load is applied.¹⁷

3. CONCLUSION

The results presented here show that the composite polymeric materials whose components possess electret charge have increased strength and possess good resistance towards corrosive media. These qualities permit the utilization of polymeric electrets as structural materials in mechanical engineering.

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