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The Effect of Direct Current Discharge on the Adhesion Properties of Poly(ethylene terephthalate) Films

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Abstract—The adhesion properties of poly(ethylene terephthalate) (PET) films modified by direct current discharge at an anode and cathode have been studied. Adhesion interactions of these films with polypyromellitimide, ultrahigh molecular weight polyethylene, and polytetrafluoroethylene modified by plasma using various adhesives have been considered. It has been shown that direct current discharge on PET films significantly improves their adhesion characteristics due to the increase in the number of oxygen-containing groups on their surface and the formation of more developed rough surface.

Keywords: poly(ethylene terephthalate) films, polypyromellitimide films, ultrahigh molecular weight polyethylene films, polytetrafluoroethylene films, direct current discharge, treatment at an anode and cathode, T-test, Uran adhesive, Evathene UE654-04 adhesive

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We have previously studied the adhesion properties of polytetrafluoroethylene and polypyromellitimide films modified by low-temperature plasma using various adhesives according to the T-test by the ASTM 1876–01 international standard [1, 2]. It was shown that treatment of the polymers by direct current (DC) discharge results in a manifold increase in the peel strength and to a significant extent depends on the chemical nature of the adhesive.

Poly(ethylene terephthalate) (PET) is widely used in the electrotechnical and cable industry for insulation of wires and cables, as well as slot insulation of electrical engines in laminated material. In the latter case, adhesion characteristics of polymer should provide strength of the laminate produced for these purposes [3]. At present, laminated material based on PET is applied as a back panel in solar cell elements [4], which consist of two external layers of polyvinylidene fluoride and an internal layer of PET [5]. This material improves electrical insulation properties of the panel, decreases moisture penetration, and significantly decreases its cost.

PET is a polyester; however, the film surface does not possess hydrophilic properties and the contact angle by water (θ_w) is 70°–84° depending on the technology of its fabrication [6–8]. Recently, low-temperature plasma exposure is one of the most effective, technological, and ecologically safe methods for the improvement of contact properties of the surface of polymer materials [9–11]. For this reason, the study of adhesion properties of PET films modified by DC discharge at a cathode and anode using the T-test according to ASTM 1876–01 and various adhesives is of certain interest.

MATERIALS AND METHODS

The object of study is an industrial biaxial-oriented PET film of PETLAIN BT 1010 E brand (Superfilm Co., Turkey) with a thickness of 40 μm. The specimens were degreased before use with ethyl alcohol and dried under room conditions.

Modification by DC discharge was carried out according to the procedure and on the setup that we described in detail in [12]. PET film specimens were located at an anode or cathode and filtered atmosphere air represented a working gas, of which the pressure was ~20 Pa in the system, the discharge current was 50 mA, and the time of treatment was 10–180 s.

The contact properties of the surface were characterized by contact angles on deionized water, which were measured by an Easy Drop DSA100 instrument (KRUSS Co., Germany) and processed by the Drop Shape Analysis V. 1.90.0.14 program (error is ±1°).

The ASTM 1876–01 procedure was used for adhesion tests. The scheme of the preparation of the specimen to determine the peel strength of adhesive joints of the PET films by T-test was described by us previously in [1, 2]. Tests of the determination of the peel strength (A) were carried out on a Hounsfield H1K

Table 1. Peel strength (*A*)* of the PET films modified at the anode and cathode

Specimen	*A*, N/m		
	adhesive		
	ED-20	UP	EVA
Initial	10 ± 1	139 ± 14	197 ± 15
Anode-treated	231 ± 20	566 ± 45	533 ± 47
Cathode-treated	275 ± 25	422 ± 35	590 ± 49

* According to T-test of ASTM 1876–01 standard.
Contact pair is PET/PET films.

tensile machine at the rate of 100 mm/min. The result of measurements represents the average of ten tests.

Three adhesives were used. The first was an ED-20 modified epoxy resin (Russia), which was mixed with hardening agent, such as polyethylenepolyamine at a 4-to-1 weight ratio. The mixture was maintained in water bath at 70°C for 5 min and deposited onto one of the joined surfaces, while the second surface was attached from top. The joined specimens were connected using a laminator at room temperature. The joined specimens were maintained for 24 h until the end of hardening of the adhesive. The second adhesive (UP) was Uran adhesive, which is a solution of polyurethane rubber in acetone and ethylacetate. The adhesive was deposited onto both joined surfaces and dried for 5 min at 90°C; then the surfaces were connected and pressed under a pressure of 0.3 MPa and 100°C for 20 s. The third adhesive (EVA) was Evathene UE654–04 (USI Co., China), which is a copolymer of ethylene with vinyl acetate. The adhesive was deposited onto both joined surfaces, and the specimen for T-test was heated in the press also under a pressure of 0.3 MPa and 100°C for 20 s.

RESULTS AND DISCUSSION

The initial PET film is characterized by contact angle θ_w = 80° by water. The studies of the change of θ_w in the case of the films modified by DC discharge at the anode and cathode under the air pressure of ~20 Pa and the discharge current of 50 mA showed that a significant decrease in θ_w to 34° and to 26° is observed after 10 s of exposure, respectively. Further increase in the time of treatment by plasma results in a gradual reach of plateau of the θ_w parameter and after 50 s of exposure its value does not change and corresponds to 12° at the anode and 10° at the cathode [13]. Derived from the results, the PET film specimens modified at the anode and cathode under the pressure of ~20 Pa, discharge current of 50 mA, and the time of treatment of 50 s were prepared for adhesion measurements.

In Table 1, the experimental values of the peel strength (*A*) are given for the PET films (PET/PET contact pair) modified under the mentioned conditions. It is clear that treatment at both anode and cathode by plasma leads to a significant improvement of the adhesion characteristics of the polymer. A remarkable effect of the type of adhesive is also observed. One example is that, in the case of the adhesive based on ED-20 resin, the value of *A* increases after treatment of the film at the anode by the factor of ~23 and at the cathode by the factor of ~27, while in the case of UP adhesive it increases by around four and three times, respectively, and in the case of EVA adhesive this increase is ~2.7 and ~3 times, respectively.

The effect of discharge treatment on the adhesion properties of the PET film during its adhesion with the films of other polymers, which are widely used in various fields of engineering, such as polypyromellitimide (PI), ultrahigh molecular weight polyethylene (UHMWPE), and polytetrafluoroethylene (PTFE) is of certain interest. The PI (50 μm in thickness), UHMWPE (100 μm in thickness), and PTFE films (60 μm in thickness) were used in the experiments. The PI films were treated at the cathode and UHMWPE and PTFE films were treated at the anode under mentioned conditions. In Table 2, experimental peel strength (*A*) of three adhesives and PET/PI, PET/UHMWPE, and PET/PTFE contact pairs are given. A significant improvement of adhesion was obtained for PET/UHMWPE and PET/PTFE contact pairs, which include the polymers with initially very low adhesion characteristics, such as UHMWPE and PTFE. An increase in *A* by 10–20 times in the case of PET/UHMWPE pair and 16–28 times in the case of PET/PTFE pair was observed depending on the adhesive.

Our studies of contact properties of PET films modified by DC discharge showed that exposure to plasma results in a significant hydrophilization of surface, which retains for a long period [13]. Using X-ray photoelectron spectroscopy (XPS), it was determined that the number of oxygen-containing groups increases on the film surface treated by plasma at the anode and cathode. Using atomic force microscopy (AFM) and scanning electron microscopy (SEM), a change in the morphology of modified films was demonstrated, namely, a significant increase in their roughness values. Improvement of adhesion properties of PET films after treatment by plasma is presumably caused by the changes of chemical structure and morphology.

Previous Fourier IR spectroscopy studies (multiply attenuated total reflectance) of the chemical structure of the PI film surface modified by glow discharge in the air atmosphere showed that a significant increase in the content of NH and COOH groups on the film surface upon exposure to plasma due to ring opening in the polymer [14]. XPS method showed that a significant number of polar oxygen-containing groups, such as hydroxyl, carboxylic, and fluoroanhydride, are formed on the surface of PTFE [15] and UHMWPE

Table 2. The peel strength (*A*)* of the PET films modified at the cathode with various adhesives in contact with films of various polymers

Specimen		*A*, N/m		
		adhesive		
		ED-20	UP	EVA
PET/PI	Initial	10 ± 1	205 ± 16	142 ± 13
	Plasma-treated	261 ± 21	610 ± 51	550 ± 47
PET/UHMWPE	Initial	28 ± 3	20 ± 2	22 ± 2
	Plasma-treated	283 ± 25	461 ± 32	470 ± 30
PET/PTFE	Initial	10 ± 1	10 ± 1	10 ± 1
	Plasma-treated	160 ± 19	233 ± 16	280 ± 24

* According to T-test of ASTM 1876–01 standard.

films [16] upon exposure to low-temperature plasma in the working gas atmosphere represented by air. The AFM and SEM methods also showed a significant increase in the roughness of plasma-treated films.

Oxygen-containing groups, which are formed on the polymer surface, presumably interact with active groups of adhesives, which results in a significant increase in adhesion between plasma-modified PET, PI, PTFE, and UHMWPE film surfaces and adhesives. An increase in roughness also provides the growth of adhesion characteristics of modified films.

CONCLUSIONS

These results indicate that exposure of the PET films under DC discharge allows a significant improvement of the adhesion properties of polymer with respect to various adhesives, as well as in contact with PI, PTFE, and UHMWPE films modified by plasma.

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