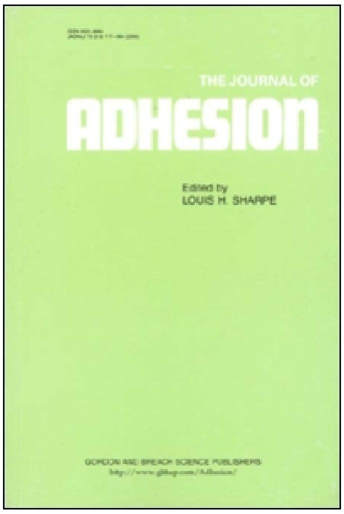

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Some Effects of Corona Discharge Treatment of Biaxially-Oriented Polypropylene Film

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The effects of the unit corona-treatment energy on the contact angle of various liquids, on the surface free energy, on the extent of oxidation of a surface layer, and on the adhesion of acrylic adhesive were studied using a biaxially-oriented polypropylene (BOPP) film. The surface free energy was determined with the van Oss-Chaudhury-Good (VCG) approach as well as with the wettability method. The extent of oxidation of the surface layer of the corona-treated BOPP film was evaluated with X-ray photoelectron spectroscopy. The adhesion strength of joints between the BOPP film and the acrylic adhesive was measured using the 180°-peel test.

In the range of the unit corona-treatment energy up to $1.2\,\mathrm{kJ/m^2}$, a rapid increase in the surface free energy with the treatment energy is observed. In the range above that value, the surface free energy rises relatively slowly. The extent of oxidation of the surface layer and the adhesion strength of joints between the BOPP film and the acrylic adhesive are approximately in direct proportion to the unit energy of the corona treatment. A five-fold growth of the adhesion strength of the studied joints within the examined range of the treatment energy was found.

Keywords: Contact angle; Surface free energy; Oxidation of surface layer; Adhesion; Corona-treated polypropylene film

INTRODUCTION

The value of the surface free energy is one of the quantities used for determination of physical properties of surface layers of polymers [1].

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It is utilized mainly when effects associated with wettability and adhesion are investigated. It enables one to estimate how well the surface layers of, *e.g.*, polymer films have been prepared for printing, adhesive bonding, laminating, metallizing, or decorating. Since most of such films are hydrophobic and are poorly wetted by paints and adhesives, their surface layers are being modified using different methods [2 – 5].

During the modification, many different physical and chemical processes proceed that are accompanied by an increase in the surface free energy. According to a commonly-accepted criterion, the surface of a polymer film becomes well wetted with printing paints and adhesives when the surface free energy of the film is higher by at least $10\,\mathrm{mJ/m^2}$ than that of the paints and glues [6]. Thus, both proper calculation of the surface free energy of modified polymer films and measuring the contact angle are important.

The surface free energy originates from unbalanced intermolecular interactions, mainly Lifshitz-van der Waals and polar interactions, occurring at an interface [7]. It can be calculated from the contact angle measured with use of a liquid (or liquids) of a known value of the surface free energy [8]. In the calculation, the Young's equation is applied [9 – 11]. This equation had been used to elaborate different methods for the determination of the surface free energy. The most recent of them is the van Oss-Chaudhury-Good (VCG) method [12 – 14], in which it has been assumed that long-range interactions (called the Lifshitz-van der Waals interactions) and acidic-basic interactions are the main interactions that affect the value of the surface free energy. Three measuring liquids, two of them being bipolar and one apolar, have to be used when applying this method.

In addition to the methods based on the measurements of the contact angle, a method for determination of wettability of polymer films with a modified surface layer is commonly used in industry [15, 16]. It consists in choosing a measuring liquid a thin layer of which separates into drops within 2 s when placed on a surface. It is assumed then that the surface free energies of both the examined film and the chosen liquid are the same.

In an ideal system, *i.e.*, one complying with Young's equation, a drop placed on the surface of an examined film is in thermodynamic equilibrium. The contact angle (Θ) is then called the equilibrium contact angle and it does not change with variation of the drop volume. In a real system, however, the drop is in a metastable state, and the measured contact angle may differ from the equilibrium value. Gradual increase in the drop volume leads initially to an increase in the contact angle only up to a limit called the advancing contact angle (Θ_A), without any change of the drop position. Then, a step-wise change of the drop position accompanied by a decrease in the contact angle occurs. On the other hand, gradual decrease in the drop volume causes initially a lowering of the contact angle only down to a limit called the receding contact angle (Θ_R). It is accepted in the literature that the surface free energy of polymer materials, including polymer films, should be calculated from the value of Θ_A [17 – 19].

In the present paper, there are presented the results of investigation of the influence of the unit energy of the corona treatment of a biaxially-oriented polypropylene (BOPP) film on its surface free energy, on the extent of oxidation of the surface layer, and on the adhesion. Calculations using the results of the contact angle measurements were made with the VCG method [12 – 14]. Then, the results of these calculations were compared with those of the measurements of the BOPP wettability, carried out using a method described elsewhere [15, 16]. The objective of this paper was to examine (i) the values of the surface free energy of the corona-treated BOPP film, determined by the VCG and wettability methods, and (ii) the relationship between this energy, the extent of oxidation, and the adhesion strength of joints including the studied film.

EXPERIMENTAL

Materials

A three-layer transparent film of BOPP (Bifol AG 4001), 40 μm thick, produced from a Malen PF 401 polypropylene (Petrochemia Płock S.A.) was investigated. Its main additives were antioxidants, antistatic lubricants, and antiblocking agents, introduced as small amounts during polymerization. This polypropylene was mostly isotactic (*ca.* 97 wt%), with a density of 0.91 g/cm^3, and a melt index of 2.6 ± 0.2 g/10 min (230°C, 21.2 N).

According to the literature [14], water, glycerol, and diiodomethane were used for the measurements of the contact angle. Two sets of mixtures: 2-methoxyethanol with formamide and formamide with water, with surface free energies within the ranges of 30–56 and 57–72 mJ/m^2, respectively, were applied as standard liquids for the measurements of the wettability [15, 16]. The surface free energies of the successive mixtures differed by 1 mJ/m^2.

Corona Treatment of the BOPP Film

The film samples were treated using a typical corona-discharge device with automated control of the discharge frequency in the range of 20–30 kHz. It provided the corona-discharge power up to 2 kW. The single-blade (knife-shaped) high-voltage electrode, 800-mm long, was made of aluminum, with no dielectric isolation. The ground metal roll covered with a 5-mm thick silicon rubber was another electrode. The discharge gap was equal 2 ± 0.2 mm, ambient temperature 23 ± 2°C, and relative humidity of the air $50 \pm 3\%$.

The unit energy (E_j) of the corona treatment was determined from the following relation:

$$E_j = P/l \cdot v \tag{1}$$

where P is the corona discharge power, l the length of the discharge electrode, and v the travel rate of the film in the gap. The E_j value was set by variation of both P and v.

Nine sets of samples treated with the unit energies shown in Table 1 were investigated. The upper limit of the unit energy of the corona treatment ($E_j = 5$ kJ/m^2, Table 1) is more than twice its average value applied during the corona treatment of a BOPP film in industrial processes.

Measurements of the Contact Angle and Wettability

The measurements of the contact angle were carried out with use of a G-11 goniometer (Krüss GmbH) directly after the corona-discharge

TABLE 1 Unit corona-treatment energies applied

Sample designation	p_1	p_2	p_3	p_4	p_5	p_6	p_7	p_8	p_9
E_j [kJ/m^2]	0	0.3	0.5	0.8	1.2	1.6	2.0	3.0	5.0

treatment was completed. The contact angle was measured as the advancing contact angle (Θ_A) and is designated by Θ_W, Θ_G, and Θ_D for water, glycerol, and diiodomethane, respectively. The volume of a drop placed with an A20 micropipette on the surface of an examined sample was 3 mm^3. The time that elapsed from the moment of the drop placement to the moment of the readout of the contact angle was within the range of 30–60 s in all of the measurements. Twelve measurements were performed for each sample; the lowest and the highest values were disregarded and the remaining ten were used to calculate an arithmetic mean, a variance, and a standard deviation, the latter being 3° or less.

The wettability was measured according to standard procedures [15, 16]. The measurements were performed directly after the corona treatment was completed. The ambient temperature was 23 ± 2°C and the relative humidity of the air $50 \pm 3\%$.

Calculation of the Surface Free Energy

According to the VCG approach [12–14], the surface free energy of a solid is equal to the sum of the following two components: γ_s^{LW}, connected with all long-range interactions (called the Lifshitz-van der Waals interactions), and γ_s^{AB}, connected with acidic-basic interactions. Thus,

$$\gamma_s = \gamma_s^{LW} + \gamma_s^{AB} \tag{2}$$

For a bipolar solid, the following formula is valid:

$$\gamma_s^{AB} = 2(\gamma_s^+ \gamma_s^-)^{0.5} \tag{3}$$

where γ_s^+ and γ_s^- are the components corresponding to the interactions of the Lewis acid and base, respectively. For an apolar or monopolar solid, the following relation is valid:

$$\gamma_s^{AB} = 0 \tag{4}$$

Hence, the calculation of γ_s consists in the determination of three unknown components: γ_s^{LW}, γ_s^+, and γ_s^-. It requires performing the measurements of the contact angle using three different liquids, two of them being bipolar and one apolar.

Then, the following general formula is obtained:

$$(\gamma_s^{LW}\gamma_{Li}^{LW})^{0.5} + (\gamma_s^{+}\gamma_{Li}^{-})^{0.5} + (\gamma_s^{-}\gamma_{Li}^{+})^{0.5} = \gamma_{Li}(1 + \cos\Theta_i)/2 \qquad (5)$$

with i = 1, 2, 3. This relation may be presented in the form of a system of equations as follows:

$$\begin{aligned}(\gamma_s^{LW})^{0.5} + g_1(\gamma_s^{+})^{0.5} + g_2(\gamma_s^{-})^{0.5} &= g_3(1 + \cos\Theta_1)\\ (\gamma_s^{LW})^{0.5} + g_4(\gamma_s^{+})^{0.5} + g_5(\gamma_s^{-})^{0.5} &= g_6(1 + \cos\Theta_2)\\ (\gamma_s^{LW})^{0.5} &= g_7(1 + \cos\Theta_3)\end{aligned} \qquad (6)$$

where Θ_1 and Θ_2 are the contact angles relating to the bipolar liquids and Θ_3 to the apolar liquid; g_i, with i = 1 – 7, are numerical coefficients.

According to the approach by Lee [20], verified experimentally for some polymer materials, it can be assumed that

$$\gamma^{+}/\gamma^{-} = 1.8 \qquad (7)$$

According to the VCG method, water, glycerol, and diiodomethane were used as the measuring liquids [14]. The values of the surface free energy and its components relevant to these liquids are shown in Table 2 [20, 21]. The values of the g_i coefficients appearing in the system of equations (6) were calculated using the data from Table 2. The calculation results for the applied measuring liquids are presented in Table 3.

TABLE 2 Surface free energy (γ_L) and its components relating to the interactions of Lifshitz-van der Waals (γ_L^{LW}), acid-base (γ_L^{AB}), Lewis acid (γ_L^{+}) and Lewis base (γ_L^{-}) for applied measuring liquids (in mJ/m^2) [20, 21]

Liquid	γ_L	γ_L^{LW}	γ_L^{AB}	γ_L^{+}	γ_L^{-}
water	72.8	21.8	51.0	34.2	19.0
glycerol	64.0	34.0	30.0	5.30	42.5
diiodomethane	50.8	50.8	≈0	0	0

TABLE 3 Values of the g_i coefficients used in Eq. (6)

g_1	g_2	g_3	g_4	g_5	g_6	g_7
0.93	1.25	7.80	1.12	0.39	5.49	3.56

Measurements of the Extent of Oxidation of the Surface Layer

The determination of the extent of oxidation of the surface layer of the corona-treated BOPP film was performed with X-ray photoelectron spectroscopy (XPS) technique using an Escalab-210 spectrophotometer (VG Scientific) provided with an aluminum anode emitting AlK_{α} radiation at a photon energy of 1486.6 eV. The pressure in the measuring chamber before the measurements was maintained at a level of 6×10^{-7} Pa. Since previous studies [22] have indicated that the X-ray radiation influences the state of the sample surface layer, the XPS measurements were performed here using a relatively low power (100 W) of the radiation source, *i.e.*, about 20% of the nominal power (the voltage of the X-ray tube was set to 10 kV and the discharge current to 10 mA). All the measurements were carried out using the same conditions for the particular spectrophotometer chambers. The irradiation time applied in the measuring chamber was equal to *ca.* 5 min.

The quantitative analysis of the surface layer composition enabled the determination of the extent of oxidation. It was performed with a special computer program (Eclipse Data System, Version 2.1), supplied along with the spectrophotometer. The results are an arithmetic mean of 5 measurements with a standard deviation of ± 1%.

Measurements of the Adhesion Strength

The determination of the adhesion strength of joints between the BOPP film and acrylic adhesive was carried out using the 180°-peel test, according to standard procedures [23, 24]. The examined joints consisted of an aluminum plate, a double-stick tape of the Z 2139 type obtained from Pyrmo, and the BOPP film. The tape was made of a foamed polyethylene copolymer, covered on both sides with a layer of acrylic adhesive. The measurements were performed with a Tiratest 2160 tensile testing machine and the velocity of the crosshead was 300 mm/min. The peel-off force was recorded as a function of the peel-off path. After a measurement, the surface of the peeled film was examined with an Amplival photometric microscope. It was found that an adhesion joint failure, *i.e.*, that occurring at the interface, appeared on *ca.* 95% of the surface area of the joints. Thirteen adhesion joints were investigated for each BOPP film of a given corona-treatment level; the lowest and the highest values were disregarded and the remaining eleven were subjected to a statistical analysis. The unit peel-off energy (W) was obtained from the following relation:

$$W = 2FL/sL = 2F/s \tag{8}$$

where F is the average peel-off force (of 11 measurements), s the width of the peeled film, and L the peel-off length.

RESULTS AND DISCUSSION

The results of the measurements of the contact angle are shown in Figure 1. Variation of this angle over the entire range of the unit energies of the corona treatment is similar for all the measuring liquids applied, the following inequality being obeyed:

$$\Theta_W > \Theta_G > \Theta_D \tag{9}$$

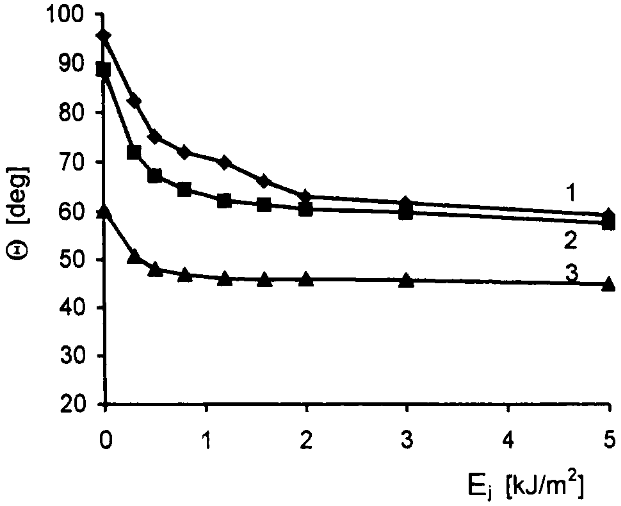

FIGURE 1 The contact angle (Θ) as a function of the unit energy of the corona treatment (E_j) for different measuring liquids: water (1), glycerol (2), and diiodomethane (3).

For $E_j \leq 0.5\,kJ/m^2$, the values of the contact angle decrease almost linearly, the rate of the decrease being the lowest for diiodomethane. Then, in the range of $0.5 < E_j \leq 2\,kJ/m^2$, this rate becomes significantly lower for all the liquids. In the range of $2 < E_j \leq 5\,kJ/m^2$, Θ_W and Θ_G diminish merely by *ca.* 3° while Θ_D is almost constant. The presented data indicate that an essential increase in the intermolecular interactions for diiodomethane occurs when the unit energy of the corona treatment is increased up to *ca.* $1\,kJ/m^2$ and, in the case of the remaining liquids, up to *ca.* $2\,kJ/m^2$. The growth of these interactions is caused by changes in the composition of the surface layer, occurring due to the corona treatment and connected mostly with the increase in the extent of oxidation [19]. The interactions originate from the surface layer of a thickness reaching *ca.* 1 nm [25].

The surface free energies (γ_s), determined by the VCG and wettability methods, are presented in Table 4 for different values of the unit energy of the corona treatment of the BOPP film.

The values of γ_s, calculated by the VCG method, are shown in Fig. 2 as functions of the unit energy of the corona treatment. It follows from this figure that the surface free energy increases, especially in the region of $E_j \leq 1.2\,kJ/m^2$. Beyond that region, the growth of γ_s is relatively little.

The values of γ_s^{LW} and γ_s^{AB}, being the components of γ_s calculated with the VCG method, are shown in Fig. 3 as functions of E_j. As seen, the contribution of the γ_s^{AB} component to the total value of γ_s is low and equals to 3.4 – 12.7%. These results confirm the conclusions presented by Dalet *et al.* [26] that the γ_s^{LW} component has a predominant effect on the wettability of polymer materials.

The results of the wettability measurements performed according to appropriate testing standards [16, 17] indicate that the values of γ_s are

TABLE 4 Surface free energies (γ_s [mJ/m^2]) determined with the methods of VCG and wettability (T) for the BOPP film treated with the corona discharge at different values of the unit energy (E_j)

Method	*E_j [kJ/m^2]*								
	0	0.3	0.5	0.8	1.2	1.6	2.0	3.0	5.0
OGC	29.5	35.9	38.3	39.7	40.9	41.0	41.0	41.3	42.5
T	33.0	45.0	51.0	54.0	58.0	59.0	60.0	62.0	65.0

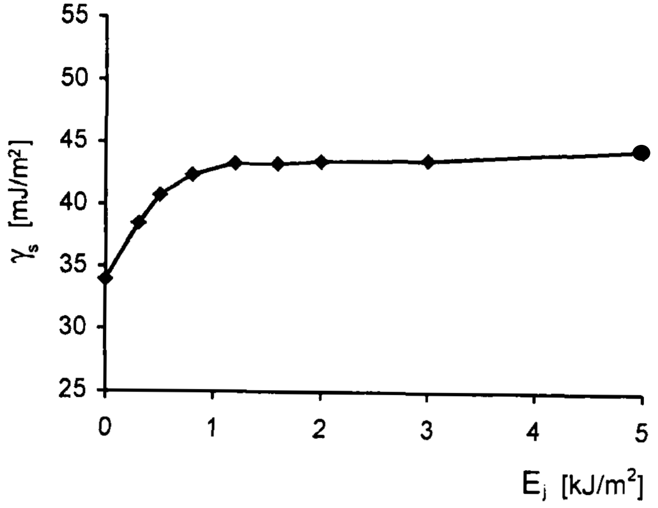

FIGURE 2 The surface free energy (γ_s) as a function of the unit energy of the corona treatment (E_j).

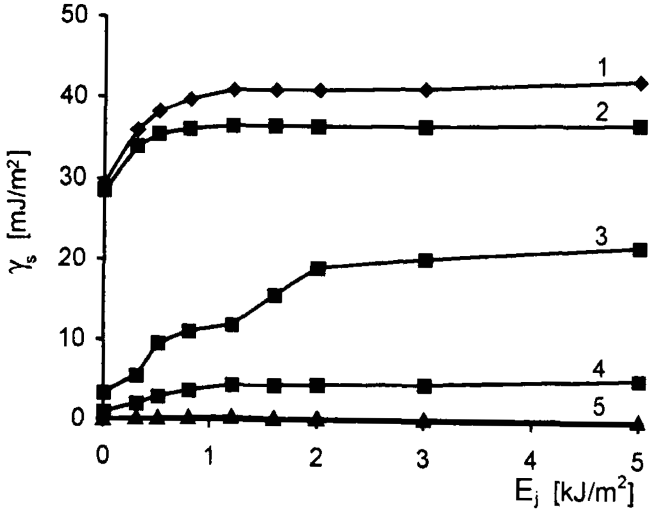

FIGURE 3 The surface free energy, γ_s (1), and its components, γ_s^{LW} (2), γ_s^- (3), γ_s^{AB} (4), and γ_s^+ (5), as functions of the unit energy of the corona treatment (E_j).

clearly higher when obtained with this method than those achieved with the VCG method (Fig. 4, Table 4). Two regions of E_j can be distinguished (Fig. 4), one in which γ_s increases rapidly and nonlinearly ($0 \leq E_j \leq 1.2\,kJ/m^2$), and the other in which a slow and almost linear increase of γ_s is observed ($1.2 < E_j \leq 5\,kJ/m^2$).

The XPS survey scans and spectra of the p_1 and p_9 samples are presented in Figs. 5a–5f. As follows from the figures, the nontreated

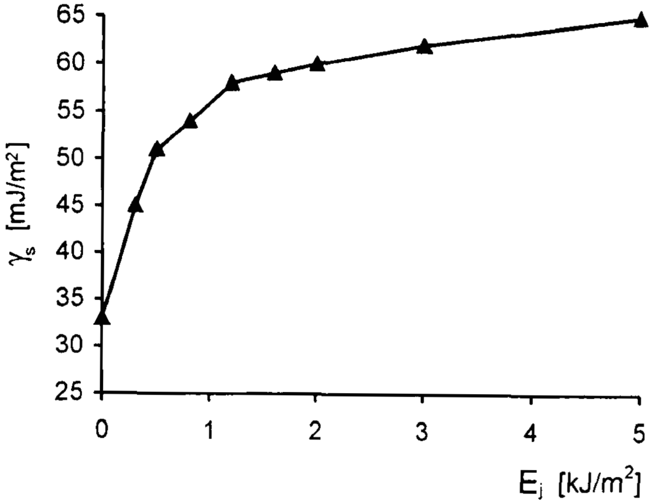

FIGURE 4 The surface free energy (γ_s) as a function of the unit energy of the corona treatment (E_j). Measurements were performed with the wettability method.

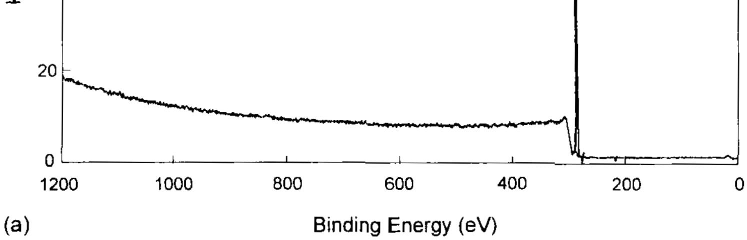

FIGURE 5 XPS survey scans and spectra of the p_1 and p_9 samples: a and b—survey scans for p_1 and p_9, respectively; c and d—C_{1S} peak spectra of p_1 and p_9, respectively; e and f—O_{1S} peak spectra of p_1 and p_9, respectively.

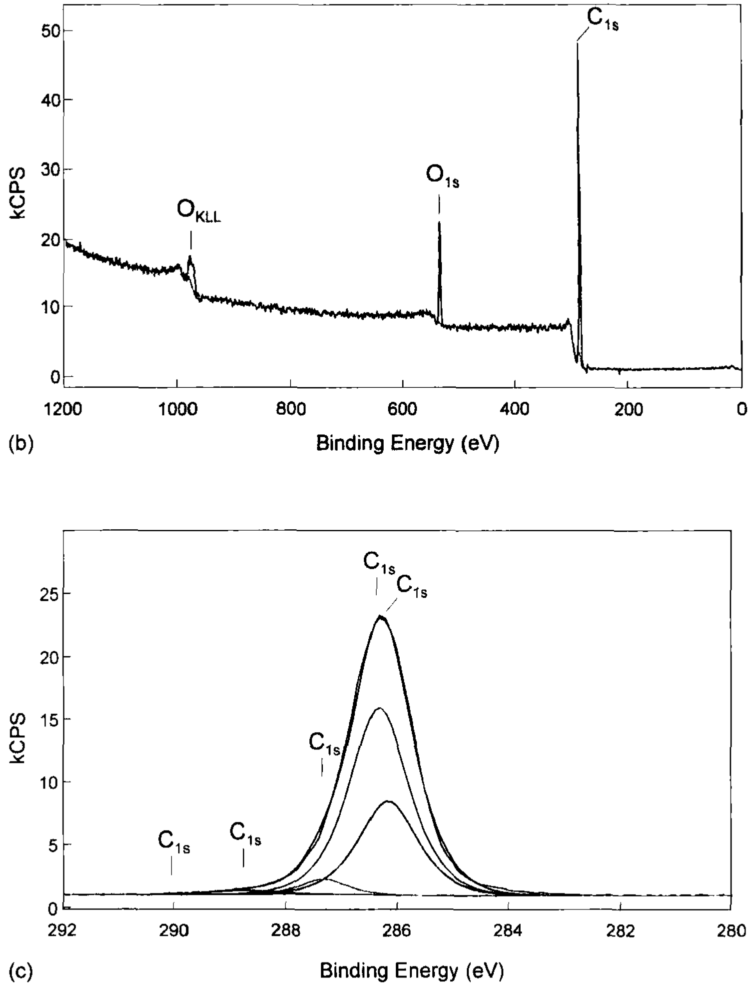

FIGURE 5 (Continued).

sample (p_1) contains a meaningless amount of oxygen which presumably comes from the ambient air and is adsorbed on the sample surface or originates as a residual product of oxidation of the surface layer of the film when the latter leaves an extrusion head. Instead, an intense peak indicating the presence of oxygen occurs in the spectrum of the sample treated at $E_j = 5\,kJ/m^2$ (p_9). A peak corresponding to Auger electrons accompanies this peak.

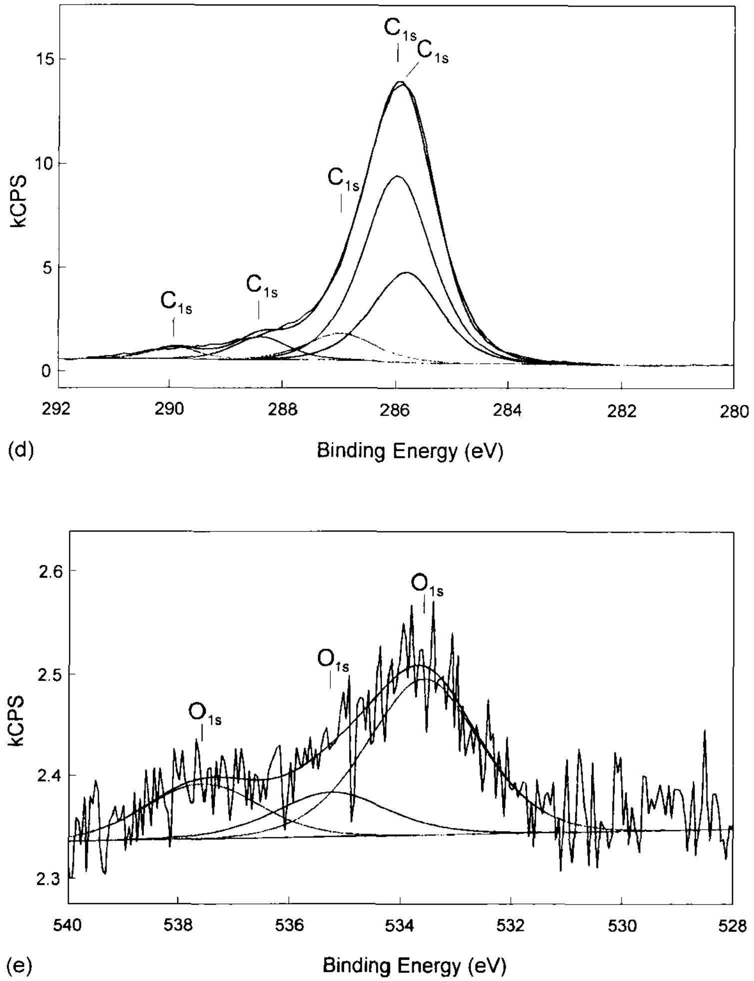

FIGURE 5 (Continued).

The extent of oxidation of individual samples, defined as the ratio of oxygen to carbon atoms (O/C), is shown in Fig. 6 as a function of the unit energy of the corona treatment. The plot can be divided into two parts, each being approximated with a line segment of a different slope. One of the segments is contained within the region of $0 \leq E_j \leq 1.2\,\mathrm{kJ/m^2}$ and the other, $1.2 < E_j \leq 5\,\mathrm{kJ/m^2}$. Such a course of oxidation results from the fact that it occurs initially in the outermost

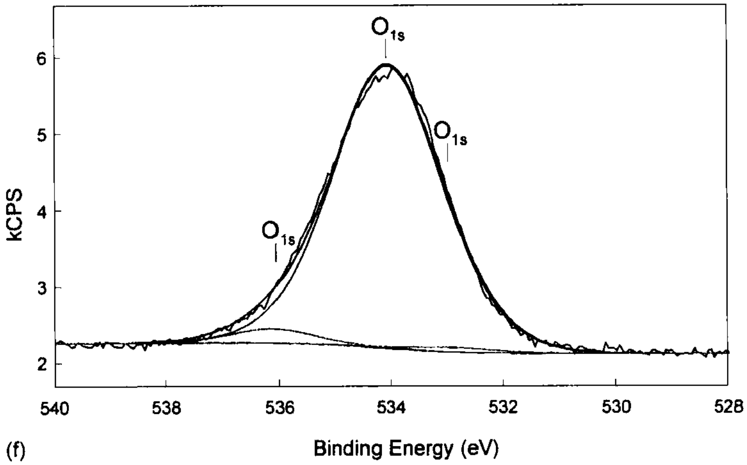

FIGURE 5 (Continued).

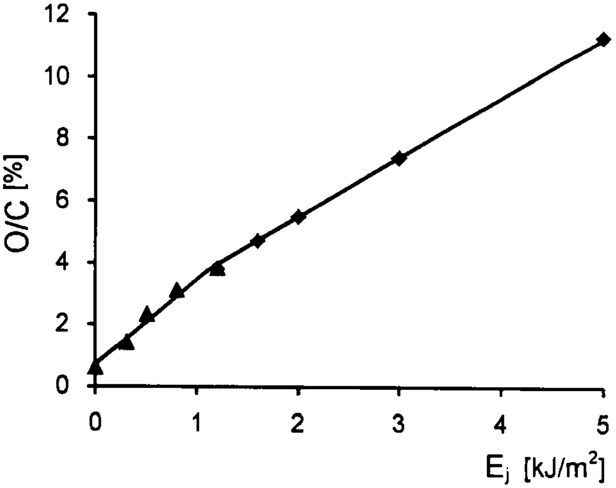

FIGURE 6 Extent of oxidation (O/C) in the surface layers of the samples $p_1 - p_9$ as a function of the unit energy of the corona treatment (E_j). The following relations are valid: $O/C = 2.744\,E_j + 0.7033$ for $0 \leq E_j \leq 1.2\,kJ/m^2$ and $O/C = 1.920\,E_j + 1.650$ for $1.2 < E_j \leq 5\,kJ/m^2$.

layer of the film. Then, deeper layers are oxidized, which are less easily accessible to oxygen; thus, the oxidation process proceeds more slowly. The oxygen content in the surface layer of the p_9 sample is almost 20 times higher than that of the p_1 sample.

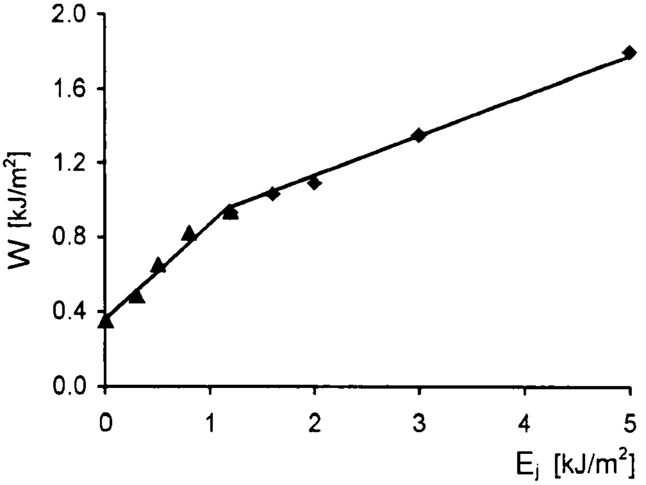

FIGURE 7 The unit peel-off energy (W) for the samples $p_1 - p_9$ as a function of the unit energy of the corona treatment (E_j). The following relations are valid: $W = 0.5073\,E_j + 0.3619$ for $0 \leq E_j \leq 1.2\,kJ/m^2$ and $W = 0.2151\,E_j + 0.700$ for $1.2 < E_j \leq 5\,kJ/m^2$.

The unit peel-off energy of the examined adhesion joints is almost in direct proportion to the unit energy of the corona treatment (Fig. 7), although the rise of W over the region of $0 \leq E_j \leq 1.2\,kJ/m^2$ is faster than that for $E_j > 1.2\,kJ/m^2$. The variation of the unit peel-off energy with E_j is, then, similar to that of the extent of oxidation in the surface layer (Fig. 6). From Fig. 7, it follows that W is equal to 0.35 and $1.80\,kJ/m^2$ for the p_1 sample ($E_j = 0$) and p_9 sample ($E_j = 5\,kJ/m^2$), respectively. Thus, the adhesion strength of the joints between the BOPP film and acrylic adhesive increases more than five times over the entire region of the unit energy of the corona treatment.

CONCLUSIONS

It follows from the presented results that growth of the unit energy of the corona treatment of the BOPP film is accompanied by a decrease in the contact angle of each of the measuring liquids as well as by an increase in the surface free energy, in the extent of oxidation of the surface layer and in the adhesion of acrylic adhesive to the film.

The surface free energy, the extent of oxidation of the surface layer, and the adhesion of the acrylic adhesive to the BOPP film vary in a similar way over the region of E_j up to 0.5 kJ/m^2. Above this value, only the extent of oxidation and the adhesion continue to change similarly to each other. The former feature indicates that, in the case of $E_j \leq 0.5$ kJ/m^2, a rapid increase in the oxidation of the outermost layer (1 nm thick) of the film occurs, which is the source of the growth of intermolecular interactions with the molecules of the measuring liquids. Further increase in the unit energy of the corona treatment leads to a progressive saturation with oxygen of this layer and to an increase in the extent of oxidation of deeper layers. The results of earlier investigations [27, 28], corresponding to a layer of thickness up to 4 nm, confirm this statement. Variations in the intermolecular interactions originating from the deeper layers do not reach the interfacial area. Thus, they do not affect changes in the contact angle and, consequently, in the surface free energy. In the range of the unit energy of the corona treatment of the BOPP film up to 5 kJ/m^2, a twenty-fold increase in the extent of oxidation of the surface layer and a five-fold increase of the adhesion of the acrylic adhesive to this film are observed.

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