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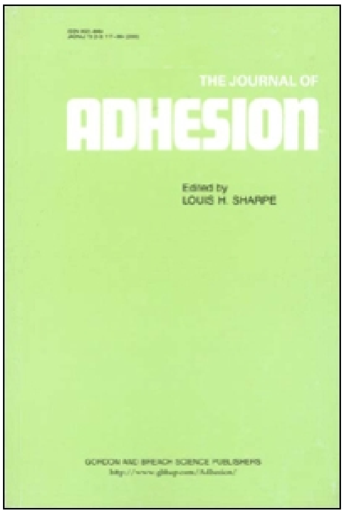

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Hydrogen Bonding and the Interfacial Component of Adhesion: Acid/Base Interactions of Corona-Treated Polypropylene*

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The effect of activation of the surface of polypropylene sheet, by a corona discharge, upon the contact angles of liquids and on the surface free energy parameters γ^{LW}, $\gamma^{\oplus}$ and $\gamma^{\ominus}$, was determined. Both advancing and retreating contact angles were measured. The "acid/base" theory of the components of surface free energy was employed.

The contact angles of water and glycerol were initially lower by as much as 30°, after treatment, and that of diiodomethane was lower by about 5°. With time, the advancing angles rose, and the $\gamma^{\oplus}$ and $\gamma^{\ominus}$ parameters fell, towards the values on the untreated solids, and attained more or less steady values after 5 to 10 days. The basic component, $\gamma^{\ominus}$, was the most strongly affected by the corona treatment; it rose, typically, from 2.2 to as high as $25\,mJ/m^2$. The acidic component, $\gamma^{\oplus}$, rose from zero to as high as $1.9\,mJ/m^2$. Its decay with time was only qualitatively the same as that of $\gamma^{\ominus}$,. The retreating angles, and the corresponding energy components, were changed in the same direction, and somewhat more strongly, than were the "advancing" data.

The well-known improvement in the property of forming strong joints or adherent coatings, after corona treatment, is no doubt due to the formation of sites or areas on the polymers where hydrogen bonds can be formed. The decay of the strength of adhesion with time is, no doubt, due to the decay of these sites or areas.

KEY WORDS: Surface activation; corona discharge; advancing and retreating contact angles; contact angle hysteresis; acid-base interactions; surface free energy; hydrogen bonding; oriented polyproylene; chemical composition of treated surfaces; XPS.

INTRODUCTION

Strong adhesion can exist between two phases, (*e.g.*, a substrate and an adhesive or coating) if strong forces, such as those due to hydrogen bonds or acid/base interactions act across the interface. Pretreatment of an apolar polymer such as polypropylene or polyethylene with a corona discharge, or exposure to a flame, can introduce polar

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groups that can enter into hydrogen bonding, either as basic (proton-acceptor) or acidic (proton-donor) groups. Pretreatment is particularly important for the adhesion of water-base coatings or adhesives[1,2].

If no pretreatment is employed, the surface behavior of an apolar polymer is characterized by a single term, γ_s^{LW}[3-7]. After pretreatment, two additional parameters are required: $\gamma_s^{\oplus}$ and $\gamma_s^{\ominus}$, the acidic and basic parameters, respectively. Together, they characterize the acid-base (AB) component of surface free energy[7-12]:

$$\gamma_i^{\mathrm{AB}} = 2\sqrt{\gamma_i^{\oplus}\gamma_i^{\ominus}} \tag{1}$$

The total surface free energy of substance i, and the interfacial free energy, γ_{ij}, are related to these parameters by:

$$\gamma_i = \gamma_i^{\mathrm{LW}} + 2\sqrt{\gamma_i^{\oplus}\gamma_i^{\ominus}} \tag{2}$$

$$= \gamma_i^{\mathrm{LW}} + \gamma_i^{\mathrm{AB}} \tag{3}$$

$$\gamma_{ij} = (\sqrt{\gamma_i^{\mathrm{LW}}} - \sqrt{\gamma_j^{\mathrm{LW}}})^2 + (\sqrt{\gamma_i^{\oplus}} - \sqrt{\gamma_j^{\oplus}})(\sqrt{\gamma_i^{\ominus}} - \sqrt{\gamma_j^{\ominus}}) \tag{4}$$

(Note that the formalism, $\gamma_i = \gamma_i^{\mathrm{LW}} + \gamma_i^p$, where γ^p is a single term that would characterize the "polar" properties of the surface, cannot be correct. It does not contain enough parameters; a minimum of three per substance are needed. See Refs.[10-11] for an extensive critique of the "γ^p" methodology.)

Many workers in adhesives and coatings have observed that the activation of a surface, such as polyethylene or polypropylene, decays with time after pretreatment. It is an interesting question whether the values of $\gamma^{\oplus}$ and $\gamma^{\ominus}$ of a solid, after pretreatment, decay at the same rate. The answer to this should cast some light on the practical bonding of adhesives or coatings to pretreated polymers.

The measurement of contact angles (θ) is the best available method for determining the parameters, γ^{LW}, $\gamma^{\oplus}$ and $\gamma^{\ominus}$ of a solid[7-12]. Three (or more) liquids are used, at least two of which must be polar. A purely apolar liquid (liquid l, for which $\gamma_l^{\mathrm{LW}} = \gamma_l$ and $\gamma_l^{\oplus} = \gamma_l^{\ominus} = 0$) may be used to determine γ_s^{LW} of the solid, using the equation,

$$\gamma_s^{\mathrm{LW}} = \gamma_l(1 + \cos\theta_l)^2/4 \tag{5}$$

When the contact angles of apolar liquid, 1, and polar liquids 2 and 3 have been measured, the set of three equations can be constructed:

$$\gamma_1(1 + \cos\theta_1) = 2\sqrt{\gamma_s^{\mathrm{LW}}\gamma_1^{\mathrm{LW}}} \tag{6a}$$

$$\gamma_2(1 + \cos\theta_2) = 2\sqrt{\gamma_s^{\mathrm{LW}}\gamma_2^{\mathrm{LW}}} + 2\sqrt{\gamma_s^{\oplus}\gamma_2^{\ominus}} + 2\sqrt{\gamma_s^{\ominus}\gamma_2^{\oplus}} \tag{6b}$$

$$\gamma_3(1 + \cos\theta_3) = 2\sqrt{\gamma_s^{\mathrm{LW}}\gamma_3^{\mathrm{LW}}} + 2\sqrt{\gamma_s^{\oplus}\gamma_3^{\ominus}} + 2\sqrt{\gamma_s^{\ominus}\gamma_3^{\oplus}} \tag{6c}$$

If liquid 1 is polar, *i.e.*, having $\gamma_1^{\oplus}$ or $\gamma_1^{\ominus} \neq 0$, Eq. (6a) will have two (or one) more terms, corresponding to the last two terms in Eqs. 6b and 6c.

The parameters, γ^{LW}, $\gamma^{\oplus}$ and $\gamma^{\ominus}$, for a number of useful liquids, have been determined (see Refs.[10–12]) and tabulated. With these data, Eqs. (6a, b, c) can be solved for the three parameters of the solid. See Ref.[11] for an algebraic method of treating the data. Table I shows the values of these parameters for the liquids that we used.

HYSTERESIS

It is commonly observed that the contact angle of a liquid on a solid exhibits hysteresis: an advancing angle (θ_a) that is quite appreciably greater than the retreating angle (θ_r).

$$H \equiv \theta_a - \theta_r \tag{7}$$

The advancing angle is probably more relevant to the process of forming an adhering system, and the retreating angle is probably more relevant to the mode of separation in adhesive failure.

Hysteresis is commonly much greater on a treated surface than on a virgin polymer. Probably the most interesting cause of hysteresis is chemical heterogeneity of a surface. Also, if a liquid swells a solid, or otherwise causes molecular rearrangement, that will (generally) cause hysteresis, and so will roughness[13–16]. If a solid surface contains two different kinds of sites, or of patches or strips, then the advancing angle will be characteristic of the lower-energy sites (or patches or strips) and the retreating angle will be characteristic of sites, etc., with the higher surface energy,[16]. We may call this concept the "dual surface" theory.

Advancing and retreating contact angles lead to two different sets of surface parameters, γ^{LW}, $\gamma^{\oplus}$ and $\gamma^{\ominus}$, for a solid, and the parameters based on θ_a data decay with time after treatment, at different rates from those based on θ_r values.

EXPERIMENT

Materials

Oriented polypropylene (OPP) films were employed. These were manufactured by the Mobil Chemical Co., Macedon, NY, and were used either without pretreatment,

TABLE I
Surface parameters of the test liquids (Ref. 10)

	γ	γ^{LW}	γ^{AB}	$\gamma^{\oplus}$	$\gamma^{\ominus}$
water	72.8	21.8	51.0	25.5*	25.5*
glycerol	64	34	30	3.92	54.7
formamide	58	39	19	2.28	39.6
diiodomethane	50.8	50.8	0	0	0
α-Br naphthalene	44.4	44.4	0	0	0

* Water was used as a reference liquid in determining $\gamma^{\oplus}$ and $\gamma^{\ominus}$. See Ref. 10.

or after pretreatment (by Mobil) with a corona discharge, in air. The film thickness was 0.001 inch. (0.0025 cm)

The polar test liquids were triple distilled water (pH = 6.8 to 7.4), and glycerol and formamide, which were purified by percolation through a column of activated alumina. The apolar liquids were diiodomethane (which was purified with activated carbon, and stored over pure copper shot, in a container that excluded light) and α-bromonaphthalene, which was also purified with activated carbon.

Equipment

A Ramé-Hart contact angle instrument was used. It was modified by means of a zoom lens, and a COHU solid state camera was employed, with a Javelin video monitor and a Mitsubishi video printer. The samples were placed on the stage of a Ramé-Hart environmental chamber, at 25°C, with a septum at the top so that saturation of the gas phase with the test liquid could be maintained. A Gilmont micrometer syringe was used to introduce the test liquid through the septum and to advance and retract the drop. Angles could be measured with the goniomenter in the optical system, or after photographing.

Measuring Method

The "recently-advanced" or "recently retreated" technique was used. For θ_a, liquid was slowly added to the drop until the drop front started to move. Addition was stopped, and the drop front was observed until apparent lateral motion ceased, and then a specified time was allowed to pass before angles were measured. For water, formamide, diiodomethane and α-bromonaphthalene, this was about 10 seconds; for glycerol, at least 30 seconds were allowed to pass. The needle of the syringe was left in the drop at all times, so the method can be described by the term, "captive drop". A similar procedure was used to measure retreating angles. Angles were measured at left and right sides, and averaged. At least 5 separate angle measurements were made in every case. The precision of the advancing angle measurements was generally better than ± 1° (90% confidence level) for any particular location on a solid. For retreating angle, the precision was better than ± 2°. All measurements were made at 25°C.

Some dependence of contact angle on drop size was noted. (The cause of the drop size dependence is not fully understood. See Ref. 17. It could not be a gravitational effect because gravity influences the drop shape only in regions of the drop that are well removed from the 3-phase line. A goniometer measurement employs the drop profile in the region very close to the point of intersection of the drop surface with the solid). The dependence was least with untreated polypropylene, and with the nonpolar liquids. With water on untreated OPP, there was no size dependence in the advancing angle for water between 2 and 12 mm drop diameter. With the retreating angle, on untreated OPP, θ_r was constant (*e.g.*, at 86°) above 6 mm diameter for water; and it decreased smoothly, to about 72°, for a 2 mm drop. On corona-treated samples, the advancing water angle rose and reached a "plateau" value by about 5 or 6 mm diameter; the retreating angle had a "plateau", usually between 3 and 6 mm, and it rose further, for larger drops. The size dependence for the retreating angle was smaller, when 30 seconds were allowed to pass, than for 10 seconds. Drops larger than 6 mm were used for θ_a measurements, and between 5 and 6 mm for θ_r.

Chemical Composition of Treated Surfaces

Some exploratory measurements were made in the laboratory of Professor Joseph A. Gardella (Dept. of Chemistry) using XPS.

RESULTS

Table II shows the contact angles on pure, untreated polypropylene, and the calculated surface parameters. On the basis of the parameters calculated from the advancing angles, the surface would be judged to be strictly apolar. The value of γ_s^{LW}, 32.6 mJ/m^2, corresponds qualitatively to the values of γ^d or to γ_c, the critical surface tension for wetting[19]. Both $\gamma_s^{\oplus}$ and $\gamma_s^{\ominus}$ were indistinguishable from zero, and γ_s^{AB} was zero.

On the basis of the retreating angles, however, a small degree of polarity was present. The values of $\gamma^{\oplus}$ and $\gamma^{\ominus}$ were, clearly, greater than zero, and $\gamma^{\oplus}$ was slightly larger than $\gamma^{\ominus}$. γ^{AB} was appreciable. γ_s^{LW} (retreating) was 32.6 mJ/m^2, and γ_s (retreating) was 39.2.

Table III shows the contact angles and surface free energy parameters on OPP that had been given corona treatment on both sides. All the contact angles in Table III are lower than the corresponding values in Table II, and the energy parameters, including γ^{LW}, are all larger, indicating higher polarity as well as higher surface free energy.

TABLE II
Contact angles and acid/base parameters of pure, untreated oriented polypropylene, OPP

	Angle, degrees	
	θ_a	θ_r
Water(W)	106	86
Glycerol(Gl)	88	64
Formamide(Fo)	77	53
Diiodomethane(D)	53	45
	Energy, mJ/m^2	
Energy parameter	from θ_a	from θ_r
$\gamma_s^{\oplus}$	0	1.3
$\gamma_s^{\ominus}$	0	0.9
γ_s^{LW}	32.6	37.0
γ_s^{AB}	0	2.2
γ_s	32.6	39.2

TABLE III
Contact angles and acid/base parameters of OPP, corona-treated both sides; 1 day after treatment

	Angle, degrees	
	θ_a	θ_r
Water	44	11
Glycerol	35	12
Formamide	17	(0)
CH_2I_2	35	25
α-Br naphthalene	22	11
	Energy, mJ/m^2	
Parameters	from θ_a	from θ_r
$\gamma_s^{\oplus}$	1.9	2.0
$\gamma_s^{\ominus}$	25.2	25.2
γ_s^{LW}	42.0	46.2
γ_s^{AB}	13.9	18.5
γ_s	55.8	64.7

Table IV shows the contact angles and surface free energy parameters on OPP that had been corona-treated on one side only. The three surface energy parameters can be calculated from data obtained with three liquids. When we used four or five liquids, we made calculations using each of the possible combinations, three at a time, and averaging the results. This gave very nearly the same values as those

TABLE IV
Contact angles and acid/base parameters of OPP, corona-treated one side only; 1 day after treatment

	Angle, degrees			
	Treated Side		Untreated Side	
	θ_a	θ_r	θ_a	θ_r
Water	68	42	88	58
Glycerine	52	32	70	53
Formamide	40	8	58	40
CH_2I_2	37	29	39	32
α-Br naphthalene	24	17	29	18
	Energy, mJ/m^2			
	Treated Side		Untreated Side	
	from θ_a	from θ_r	from θ_a	from θ_r
$\gamma_s^{\oplus}$	1.25	1.8	0.25	0.3
$\gamma_s^{\ominus}$	8.2	25.5	1.2	19.3
γ_s^{LW}	41.1	44.6	40.1	43.4
γ_s^{AB}	6.4	13.5	1.1	4.8
γ_s	47.6	58.2	41.2	48.2

obtained by using all four (or five) in an overdetermined set of linear equations, and minimizing the deviations due to inconsistency of the data. The changes in the contact angles for the treated side, from those in Table II, were smaller than those in Table III. The values for the "untreated" side were appreciably above the values of the polymer that had had no treatment at all. The values of θ_a and θ_r on the untreated side were independent of time.

Figure 1 to 5 show the changes of the contact angles of water, glycerine and diiodomethane, with time after treatment. The values of θ_a and θ_r follow a pattern of starting out below the value for the untreated side, and rising appreciably in the first seven days. The extent of this rise varied; for samples that had been given a single corona treatment, θ_a for water and glycerine rose to a level above that of the untreated side, and leveled off. For samples that had been given more than one treatment, the trends were more complex, but all appeared to attain an approximately steady value.

Figures 6 and 7 show the time trends of the $\gamma_s^{\oplus}$, $\gamma_s^{\ominus}$ and γ_s^{LW} parameters, for the treated samples. Consider first, Figure 6. The values of $\gamma_s^{\ominus}$ based on θ_a data for the samples treated two or more times decreased strongly for the first 5 days, and then approached a roughly constant value, *e.g.*, about 10 mJ/m^2 for 5 treatments.

$\gamma_s^{\oplus}$ also decreased, but attained much lower steady values than did $\gamma_s^{\ominus}$, relative to the initial values. The values of $\gamma_s^{\ominus}$ (retreating), in Figure 7, behaved rather like $\gamma_s^{\ominus}$ (advancing) except for the case of three corona treatments. The behavior of $\gamma_s^{\oplus}$

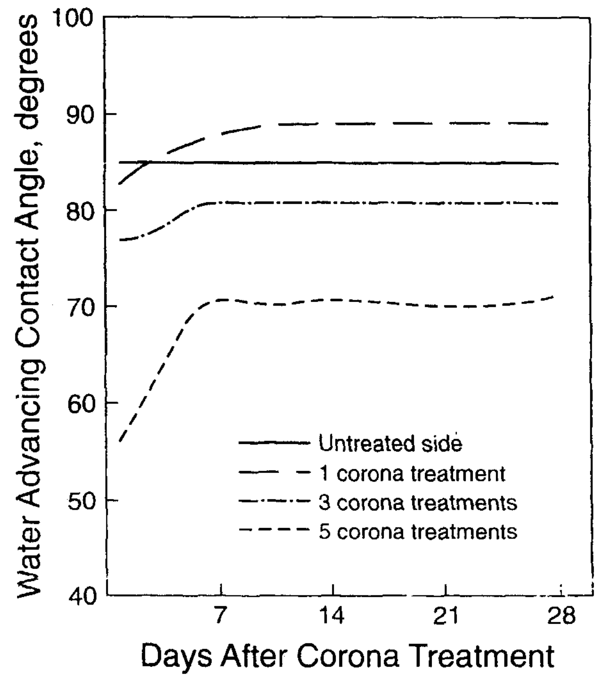

FIGURE 1 Advancing angle, θ_a, of water on OPP, corona treatment on one side only, *vs.*, time after treatment.

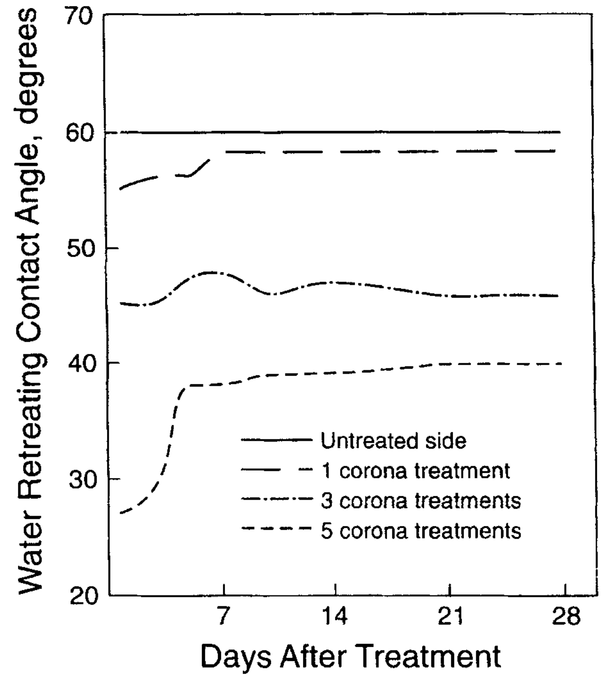

FIGURE 2 Retreating angle of water on OPP, corona treatment on one side only, *vs.* time after treatment.

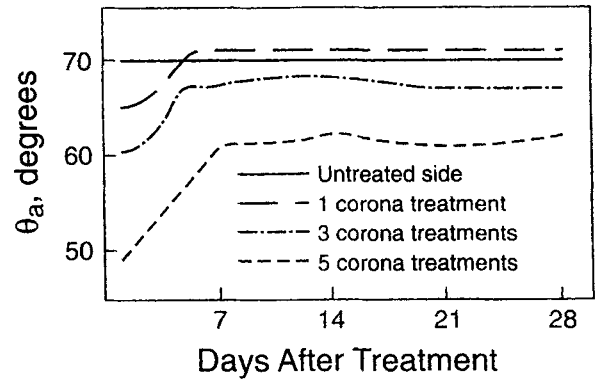

FIGURE 3 Advancing angle, θ_a, of glycerol on OPP, corona treatment on one side only, *vs.*, time after treatment.

(retreating) was quite different from $\gamma_s^{\oplus}$ (advancing); for 3, 4, and 5 treatments, $\gamma_s^{\oplus}$ increased slightly and then levelled off, while for 1 treatment, these was a decline. For 2 treatments, the value was close to that for 3 or more treatments, but slightly lower.

Figure 8 shows the effect of pH of the water used in the tests, on the contact angles. The acidic surface groups that are involved in the advancing angle appear to

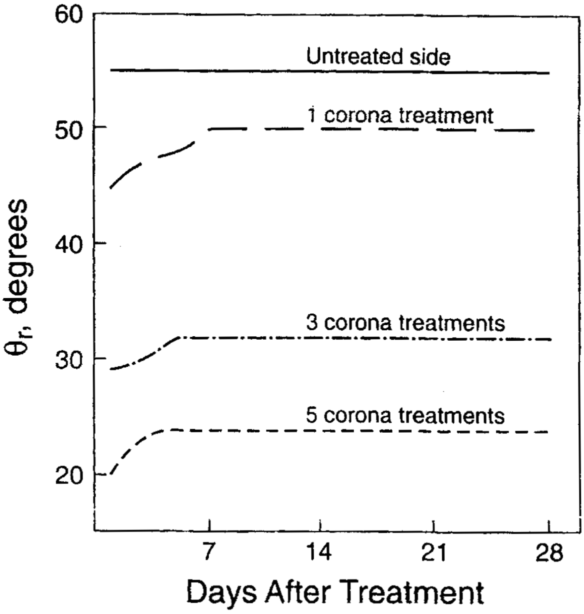

FIGURE 4 Retreating angle of glycerol on OPP, corona treatment on one side only, *vs.*, time after treatment.

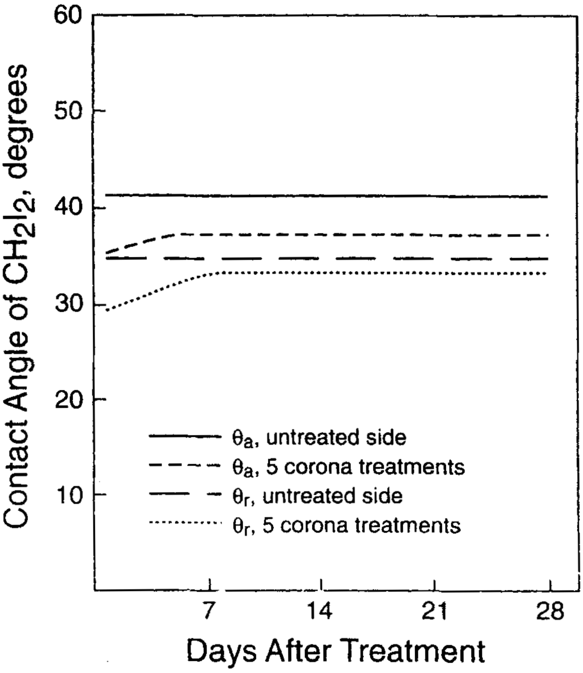

FIGURE 5 θ_a and θ_r of CH_2I_2 on OPP, corona treatment on one side only, *vs.* time after treatment.

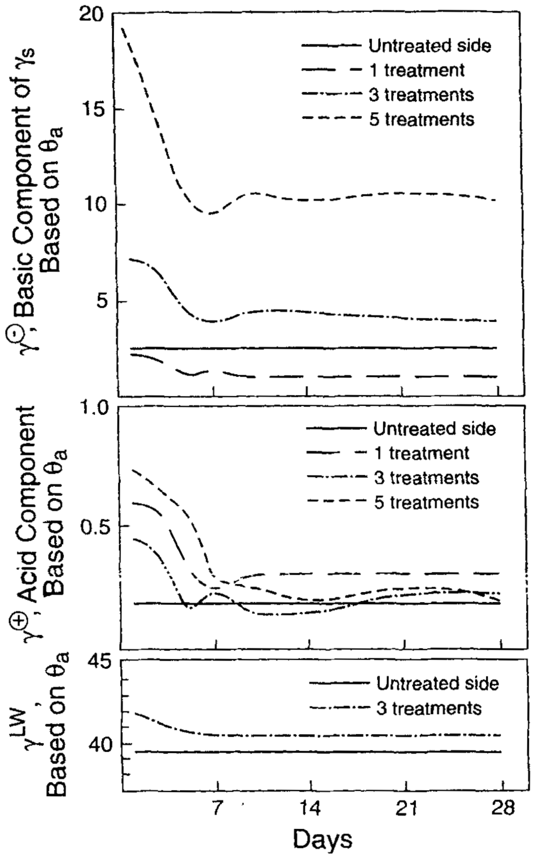

FIGURE 6 Components of γ_s based on θ_a data, *vs.* time after corona treatment.

be neutralized at pH 12, while those that are involved in the retreating angle are neutralized over the range, 10 to 12.

A set of exploratory XPS experiments was carried out for us on corona-treated films, by Professor Joseph A. Gardella and Mr. Robert Johnson, of the Chemistry Department of this University. Evidence was found for oxygenated groups: OH, C=0, COOH, epoxy, ether and ester. The carboxylic acid and alcohol groups could participate in hydrogen bonding, and contribute to the non-zero values of $\gamma_s^{\oplus}$. The carbonyl, epoxy, ether and ester groups could contribute to the $\gamma_s^{\ominus}$ functionality and to the proton-acceptor properties of the surface.

Discussion

The expected apolar character of untreated polypropylene was exhibited in the zero values of $\gamma_s^{\oplus}$ and $\gamma_s^{\ominus}$ in Table II, based on θ_a data. The higher values of all the energy parameters, in the last column of Table II (based on θ_r data) suggest the presence of

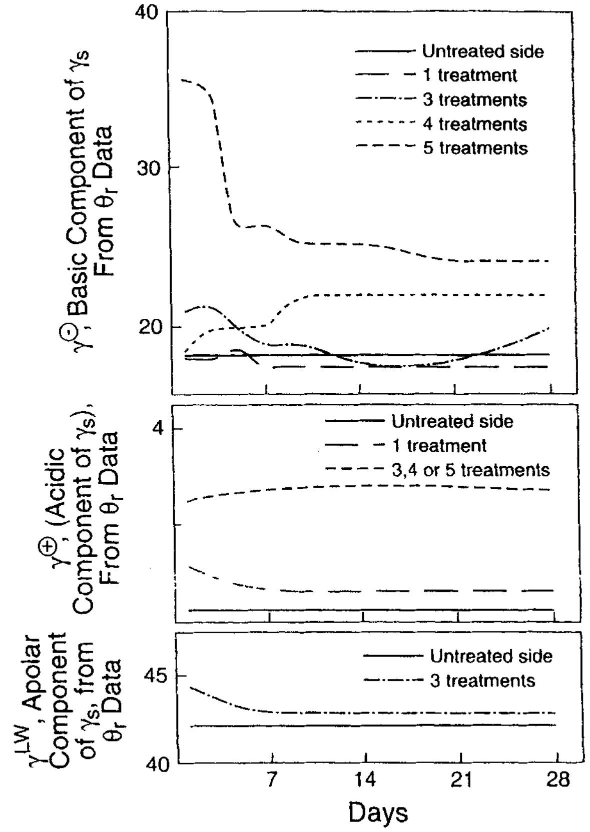

FIGURE 7 Components of γ_s based on θ_r data, *vs.* time after corona treatment.

impurities in the surface of the "as received" polymer. (The "dual surface" theory of hysteresis was mentioned above).

It is clear that the corona treatment increased the polarity of the polypropylene surface, as may be seen from Tables III and IV; also, the apolar component was increased. We may compare these resuts, based on θ_a values, with data on several oxygenated polymers. See Table V[10,11]. The values of γ_s^{LW} and $\gamma_s^{\ominus}$ in Table V are of the same general magnitude as those of the corresponding parameters in Tables III and IV. $\gamma_s^{\oplus}$, however, is between 1 and 2 mJ/m^2 for the corona-treated polymer, *vs.* nearly zero for the pure polymers listed in Table V.

The water contact angles were independent of pH for the pure olefin polymers, and this agrees with expectations based on the near-zero $\gamma_s^{\oplus}$ and $\gamma_s^{\ominus}$ values.

The time dependence of the contact angles themselves has already been described. We cannot at present go beyond simply stating that the trends of the three surface parameters (Figs. 6 and 7) "explain" the contact angle trends. The active surface groups may be reoriented, with time, so that they do not "face toward" the air side of the interface. Or they may diffuse into locations where they form hydrogen bonds within the surface of the solid. Or they may react with chemical species from the air. These are, of course, speculations that need independent support.

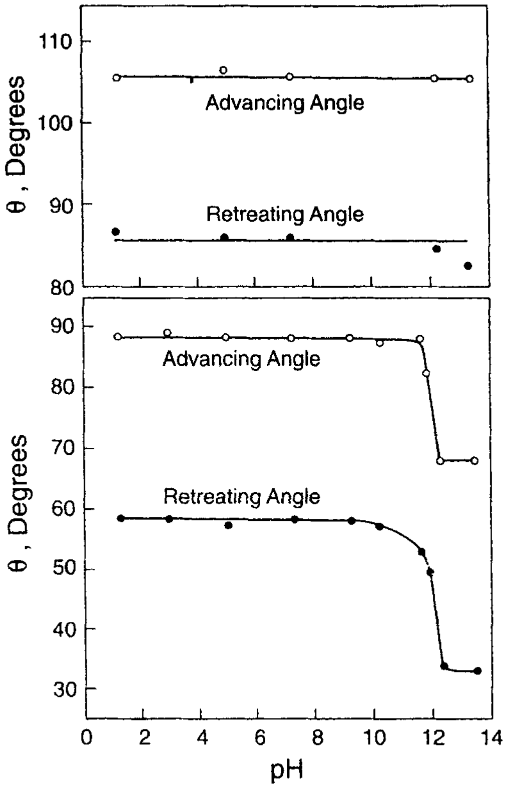

FIGURE 8 Water contact angles on pure OPP *vs.* pH; upper graph: untreated; lower graph: once-treated, one side.

TABLE V
Surface energy parameters of certain oxygenated polymers. Data from Refs. 9–11

	Surface energy parameter, mJ/m^2		
Solid	$\gamma_s^{\oplus}$	$\gamma_s^{\ominus}$	γ_s^{LW}
Poly(methylmethacrylate)	0	9.5 to 22.4	39 to 43
Cellulose acetate	0.3	22.7	35
Agarose	0.1	24	41
Poly(oxyethylene), PEG 6000*	0	66	45

* Union Carbide.

The behavior of $\gamma_s^{\oplus}$ derived from retreating angle data, for 3, 4 or 5 treatments (Fig. 7) is unlike that for $\gamma^{\oplus}$ derived from advancing angle data, Figure 6. This observation lead to the conclusion that different groups or molecular species are responsible for $\gamma^{\oplus}$ in the θ_a and in the θ_r measurements. As already noted, the three surface energy parameters indicate chemical changes in the solid, even when the surface was on the side opposite to where the electrode was. There may have been a bulk effect, penetrating right through the film, and directly causing the surface chemical changes. Or a charge may have been induced that caused ionization in the gas phase, and bombardment of the surface with reactive species.

Conclusion

An obvious application of this technique of studying activation of low-energy surfaces is that, for high adhesion, the surface composition of the adherends should be complementary, in the acid/base sense. Now, contact angle data can be employed for establishing this complementarity.

Most of these results were reported in the theses of Dr. L. K. Shu, 1991, and Mr. H.C. Chiu, 1991.

References

1. R. F. Grossman and W. A. Beasley, *J. Appl. Polymer Sci.*, **2**, 163(1959).
2. C. Y. Kim and A. D. Goring, *J. Appl. Polymer. Sci.*, **15**, 1365(1971).
3. L. A. Girifalco and R. J. Good, *J. Phys. Chem.*, **61**, 904(1957).
4. R. J. Good, and L. A. Girifalco *J. Phys. Chem.*, **69**, 561(1960).
5. F. M. Fowkes, *Ind. Eng. Chem.*, **56**, 40(1964).
6. F. M. Fowkes and M. A. Mustafa, *Ind. Eng. Chem. Prod. Res. & Dev.*, **17**, 3(1978).
7. C. J. van Oss, M. K. Chaudhury and R. J. Good, *Advan. Colloid Interface Sci.*, **28**, 35(1987).
8. C. J. van Oss, M. K. Chaudhury and R. J. Good, *Sep. Sci. and Technology*, **22**, 1(1987).
9. C. J. van Oss, M. K. Chaudhury and R. J. Good, *Chem. Rev.*, **88**, 927(1988).
10. R. J. Good, M. K. Chaudhury and C. J. van Oss, in *Fundamentals of Adhesion*, L. H. Lee, Ed. (Plennum Press, NY, 1991), p. 153.
11. R. J. Good, *J. Adhesion Sci. & Technology*, **6**, 1269(1992).
12. C. J. van Oss, R. J. Good and H. J. Busscher, *J. Dispersion Sci. and Technology*, **11**, 75(1990).
13. R. J. Good, *Am. J. Chem. Soc.*, **74**, 5041(1953).
14. R. E. Johnson, Jr. and Robert H. Dettre, in *Contact Angle, Wettability and Adhesion*, Advan. Chem. Series, no. **43**, F. M. Fowkes, Ed. (Am. Chem. Soc., Washington, DC, 1964), p. 112.
15. R. H. Dettre and R. E. Johnson, jr., *ibid*, p. 136.
16. R. J. Good, in *Colloid and Surface Science*, no. **11**, R. J. Good, and R. R. Stromberg, Eds. (Plenum, NY, 1979), pp. 1–29.
17. R. J. Good, and M. N. Koo, *J. Colloid Interface Sci.*, **71**, 283(1979).
18. R. H. Cramm and D. V. Bebee, *TAPPI Press*, **65**, no. 8, 75(1982).
19. R. H. Fox and W. A. Zisman, *J. Colloid Interfce Sci.*, **7**, 428(1952).