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Investigations have shown an excellent adhesion of particularly polyurethane adhesives to Corona-Discharge-treated current automotive topcoats, independent of the type and amount of many top-coat additives. Adhesion experiments and the determination of surface energy have shown an insensitive resulting performance towards changing process parameters.

Scientific examinations with IR and ESCA analysis as well as electron microscopy provide valuable explanations for the efficiency of the *Corona Discharge* treatment.

The Problem

In the Automotive Industry many coated materials are bonded frequently and in great extent together with glass, plastics, and textiles today. Examples are: direct glazing, fastening of plastic parts, spoilers and friezes, bonding of SMC panels to the load bearing frame.

All paints and coatings contain additives, which ease the compounding or the application or even provide some finishing

properties to the paint surface. The most important properties amongst those are as follows: dispersion, self leveling, antifoaming, pigment wetting, conductivity, brilliance, and dust repulsion.

These additives are non-volatile surface-active agents, which also migrate to the surface of the finished paint coating. Additives at the surface of electrocoats, surfacer, primer, and base coats become dis-

solved in the next wet system applied and migrate to the surface of the top coats, *i.e.* the clear coats. Most additives are derivatives of silicone oils containing methyl and phenyl groups. They create a hydrophobic layer on the surface and reduce the surface energy to less than 30 mN/m (*Table 1*). As a result, they have poor wettability characteristics when used with adhesives possessing higher surface tensions. In contrast to top coats, these surface-active agents contain no functional groups for powerful interaction or chemical reaction with conventional adhesives (*Fig. 3*).

Table 1

Surface Energy [mN/m]			
Glass	73	Water	73
Phosphatised Steel	43–46	Glycerine	63
Cathodic E-Coat	32–40	Ethylene glycol	48
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Polypropylene	28	Butanol	23
Polytetrafluoroethylene	19	Xylene	29
Alkyd Resin	33–60	Butylacetate	25
Alkyd Melamine		Diisobutylketone	22
Top Coat	30	Solvent	
Polyvinylchloride	39–42	Naphtha	18–22
Polymethacrylate	32–41		
Epoxy Resin	45–60		
2 pt Epoxy Adhesive	39–44		
2 pt PUR Adhesive	44		
Polyamide	46		
Polydimethylsiloxane	22		
Togocoll FH 500	33		
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Effect

In many cases, as a result of poor wetting, insufficient compatibility, and lack of functional or polar groups, adhesion of the adhesive is unsatisfactory, and the bond strength is lower than specified. Additionally, in the case of trim moldings, the molding may separate itself from the paint. To cope with such problems, it is necessary to mask the adhesion zone before applying the top coat or to utilize a paint primer in the adhesion zone. Both alternatives are expensive, labor intensive, and create another error parameter in the process.

To obtain improved reliability, higher bond strengths and lower production costs for windshield and backlight assembly operations in the world-wide automotive industry, the *Corona Discharge* process will be introduced.

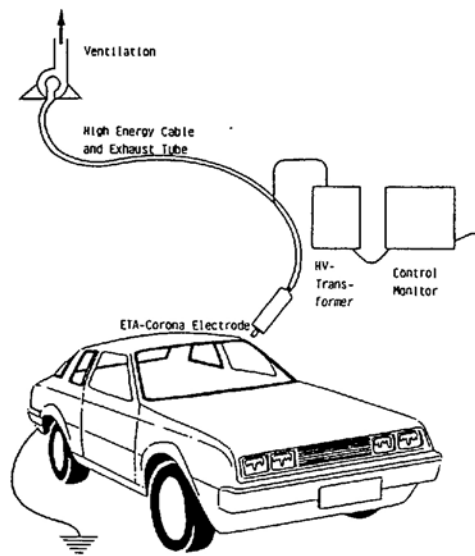


Fig. 1. Corona discharge treatment

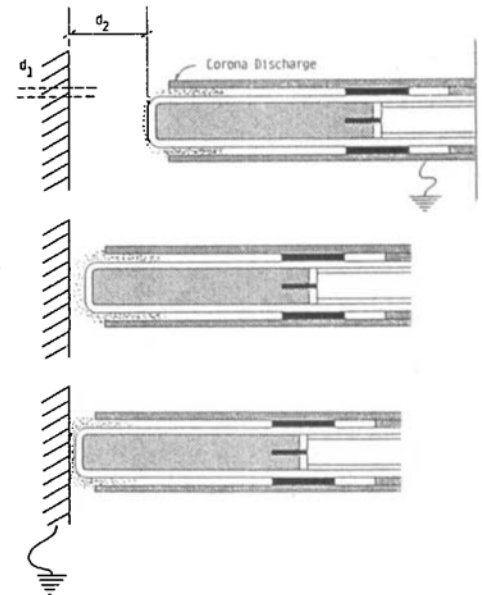


Fig. 2. Corona build-up between electrode and work piece

Process

The main feature of the process is the use of high-frequency corona-discharge technique, already widely used in the olefin film industry to improve bonding characteristics for adhesives and printing inks. During the process, the paint surface in the adhesive zone is exposed to corona discharge by an electrode, which is robotically controlled. The discharge creates molecular alterations in the surface of the paint (Fig. 2).

A high-frequency current is used to develop a discharge between the high-tension electrode and the painted panel surface. The gap between the electrode and the surface is ionized, producing reactive radicals and ozone. Besides oxygen and nitrogen radicals and ions, nitrogen oxides (NO_x) are formed. Oxygen and NO_x are deposited on the treated surface with the resultant formation of polar groups in the boundary layer molecules. Typically, carboxyl, carbonyl, hydroxyl, nitro, and ether groups are present in the treated surface. Additional impacts of accelerated electrons and

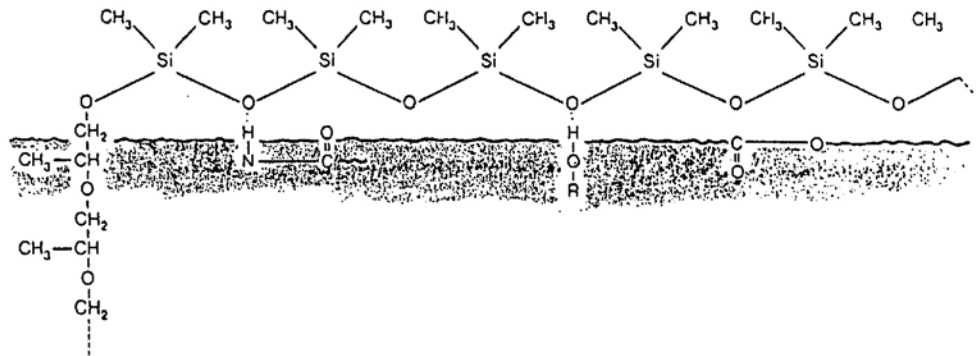


Fig. 3. Paint additive on the paint surface

ions lead to a partial cleavage of C-C bonds and secondary reactions at the exposed surface.

Significantly higher 'wettability' and substantially improved compatibility and reactivity to adhesives is the result of the modified molecular layers in the painted surface.

The electrode has a diameter of 3 to 5 cm and a length of 30 cm. Within the active

area of ca. 10 cm² corona discharge takes place. All exposed surfaces of the equipment are grounded (Fig. 2). NO_x and O₃ generated by the corona discharge are evacuated through the gap between the electrode and the jacket by vacuum. Subsequently, these gases are passed through a catalyst system where they are converted to harmless products. For the corona treatment, the moving electrode is spaced

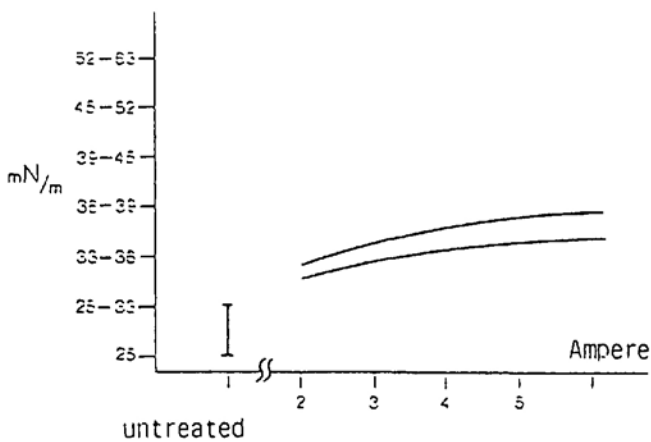
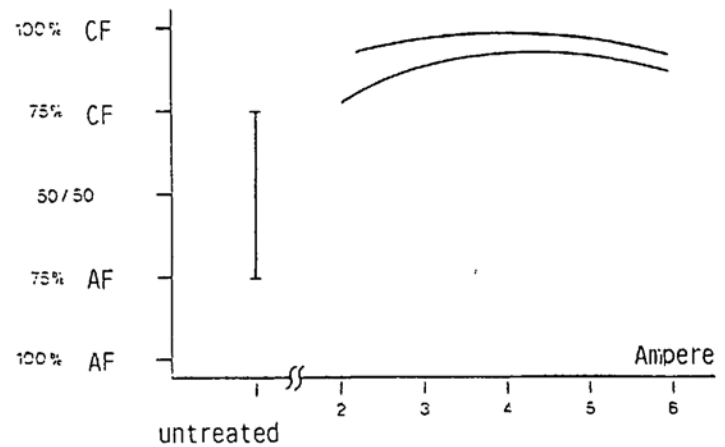


Fig. 4. Surface tension and peel adhesion vs. effect of current



AF = Adhesive Failure
CF = Cohesive Failure

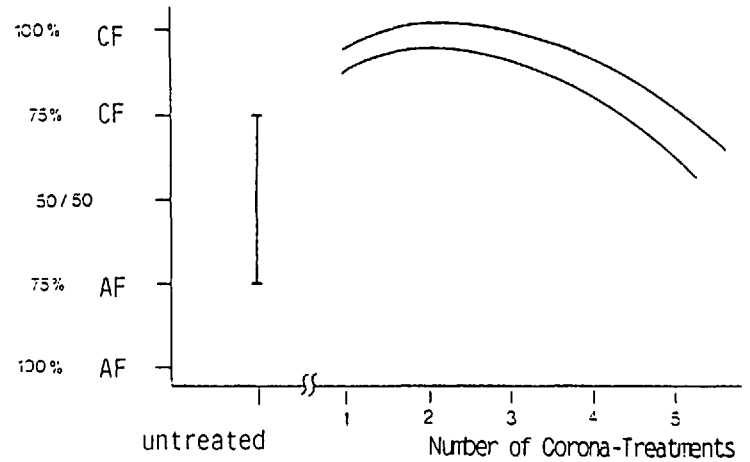
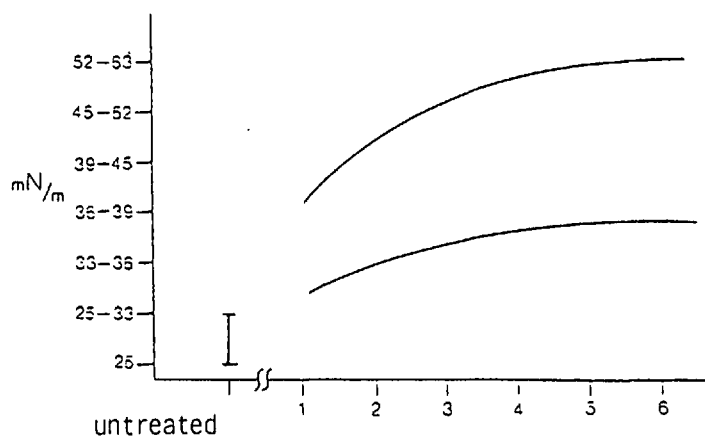


Fig. 5. Surface tension and peel adhesion vs. effect of number of treatments

AF = Adhesive Failure
CF = Cohesive Failure

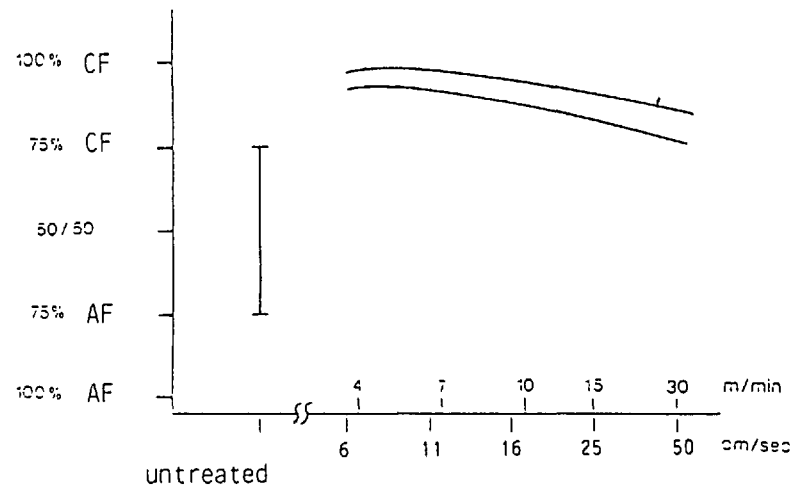
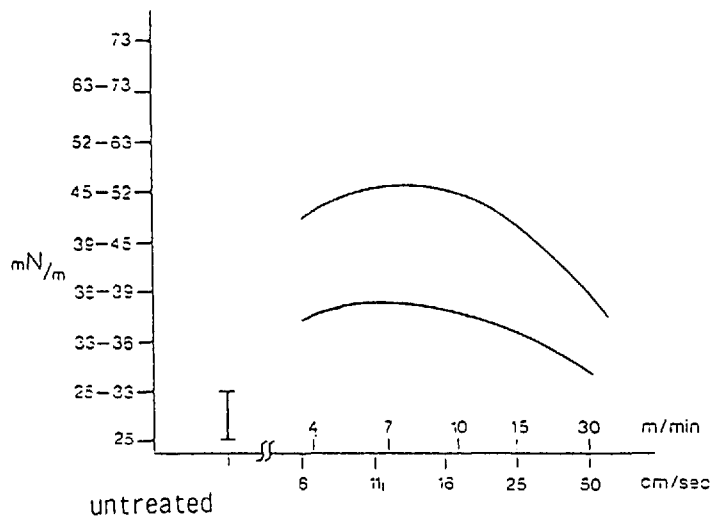


Fig. 6. Surface tension and peel adhesion vs. effect of traverse speed

Traverse Speed

AF = Adhesive Failure
CF = Cohesive Failure

equidistantly of 2-4 mm to the surface by a free arm robot, and the surfaces are normally treated at a linear speed of 10 to 30 m/min. With a fixed arrangement, the electrodes are positioned in a row configuration, a few millimeters from the surface, during 0.3 to 3 s.

In contrast to corona treated plastic films, which in time, lose the molecularly changed surface structure, corona-treated painted surfaces retain the molecularly changed layer on the surface. Therefore, treated surfaces may be bonded immediately, or several days later, if contamination with dirt and oil is prevented.

Equipment, Parameters, Results

The equipment for corona-discharge treatment of paint surfaces consists of a high-voltage transformer, a generator and an electrode, whose particular property is its inherent safety, since no electrical potential is developed between the electrode and ground for an operating or unused electrode. The ionized air surrounding the surface of the electrode tip has a high con-

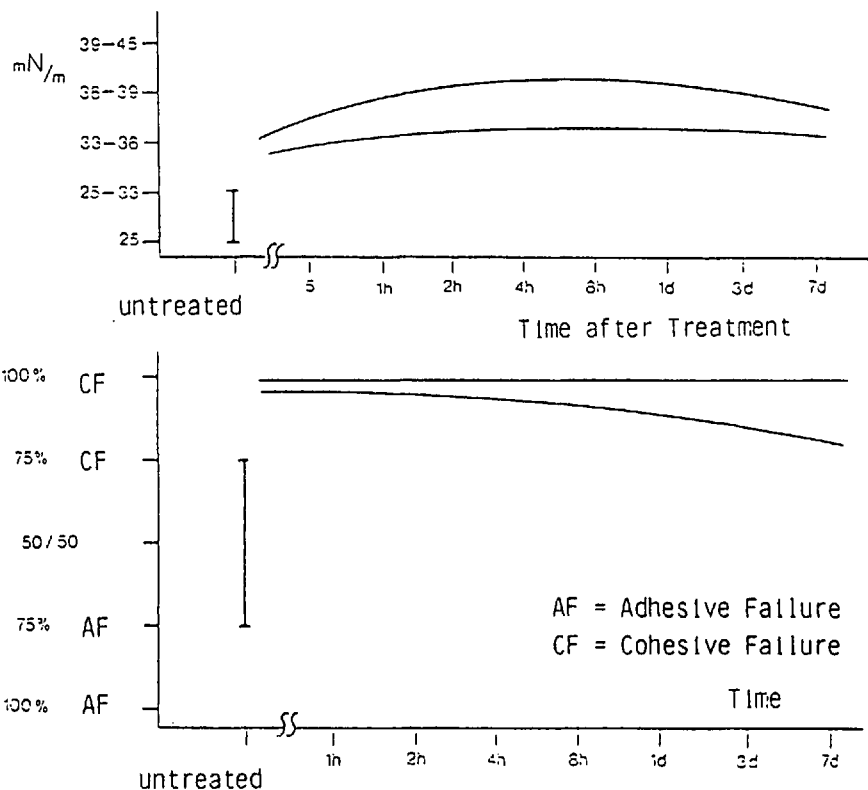


Fig. 7. Surface tension vs. time after treatment and peel adhesion vs. effect of time interval between treatment and application of adhesive

ductivity and is in contact with the grounded jacket. Even by touching the electrode accidentally during operation, no electrical shock must be feared. Approaching a painted surface with the electrode, the corona discharge is preferentially formed between the electrode and the painted surface. In the gap between the electrode and the jacket, air may be evacuated to remove free O₃ and NO_x products by ventilation or in a solid-state catalyst.

Testing the Efficiency of the Corona-Discharge Treatment

Surface energies of the paint surfaces before and after the corona treatment have been determined to be between 22 and 70 mN/m by using test inks of known surface energies. Adhesion properties have been measured on bonds with a direct glazing adhesive on the basis of a one-part moisture cure polyurethane. Changes taking place in the treated surface were recorded by IR analysis, ESCA (XPS) method and electronmicroscopy. The efficiency was tested on numerous European and USA serial and model top coats (Table 2). A two-component polyurethane top coat, in commercial use at a German automobile producer, was used as a base to prepare samples with varying additive formulations. Amounts, type, and composition of the used additives are listed in Table 3.

Results

The following figures illustrate the dependence of adhesion and surface tension on varied process-treatment parameters for the range of European and USA paint systems. The evaluation of the peel fracture surface of adhesive bonds suits well to judge the quality of adhesion. The adhesion ranges from 100% adhesive fracture (af, complete loss of adhesion), to 100% cohesive fracture in the adhesive (cf). Fig. 4 illustrates that peel adhesion and surface energy is relatively unaffected by increasing current levels. Variations may occur in the generator and by variable distances of the electrode to the material surface.

Table 2. Production Top Coats

European Manufacturers	
Paint No.	
16	Daimler-Benz
17	BMW
18	Citroen
19	Ford
20	GM I
21	GM II
22	Opel
23	Peugeot
24	VW
USA Manufacturers	
Paint No.	
25	GM 2-layer base coat/clear coat
26	Ford straight shade (Inmont)
27	Ford 2-layer top coat
28	Nissan straight shade
29	Nissan 2-layer base coat/clear coat

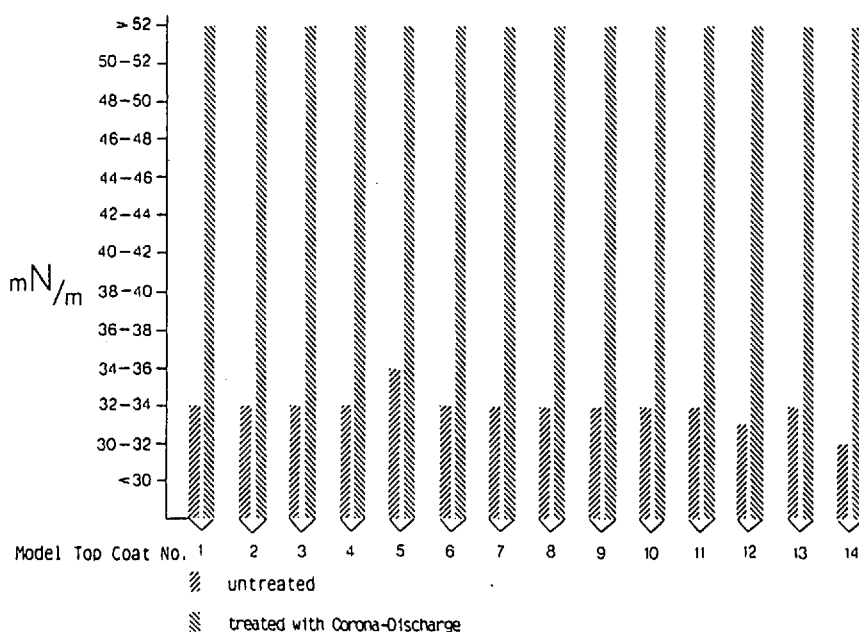


Fig. 8. Surface energy of Herberts model top coats

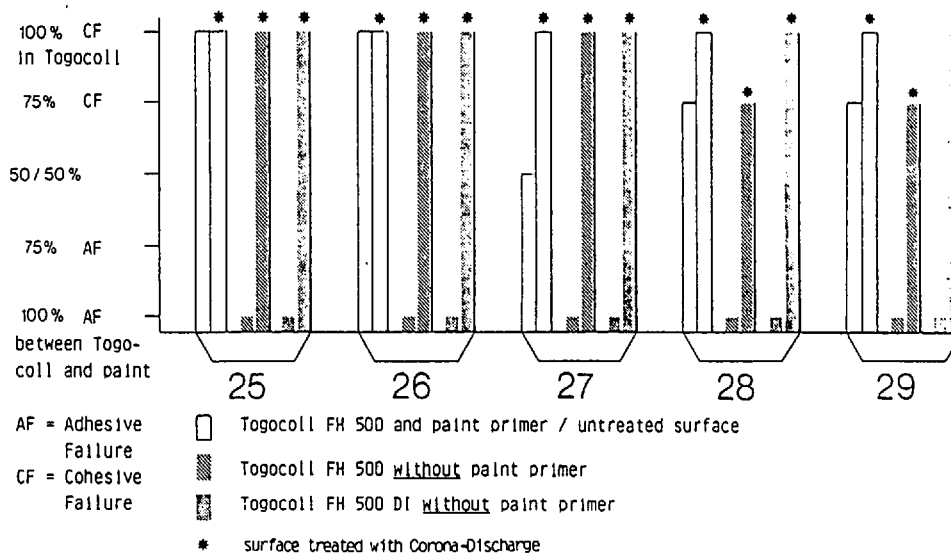


Fig. 9. Peel adhesion tests of USA top coats

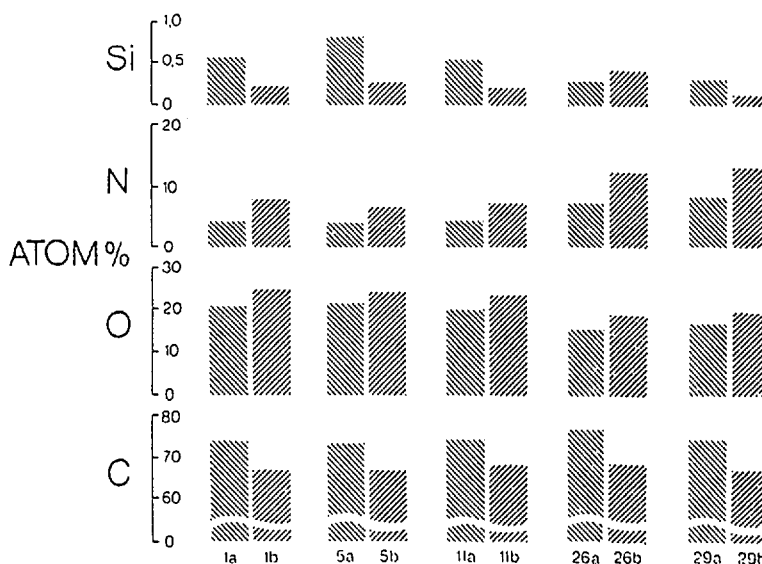


Fig. 10. Composition change of paint surfaces by corona treatment. a) Untreated, b) after treatment.

Table 3. Model Top Coats for Corona Treatment

Adhesion Trials	Composition
1. Original 2-component high solids PUR top coat	Hydroxy-functional polyesters and polyacrylates. Aliphatic polyisocyanate
2. Original + 0.2% Dimethylaminol	Diethanolamine
3. Original + 0.2% Byk P 104 S	Salt of an unsaturated polycarbonic acid
4. Original + 0.5% Duomeen TDO-Lsg. 15%	cationic stearylamine derivative
5. Original + 0.2% Byk 354/50	Clear polyacrylate
6. Original + 0.3% Disperbyk 160/30	Alkanolamine salt of high molecular weight, Polyacrylic acid copolymers
7. Original + 1.0% Silicon Oil OL 1% (Bayer)	Methylphenylpolysiloxane, polyethermodified
8. Original + 0.1% Silicon Oil OL 10% (Bayer)	Methylphenylpolysiloxane, polyethermodified
9. Original + 0.1% Silicon Oil AMRA (Langer)	Silicon oil
10. Original + 1.0% Silicon Oil PL (Bayer)	Low-molecular-weight methylphenylpolysiloxane
11. Original + 1.0% Silicon Oil AK 150	Middle molecular weight polydimethylsiloxane
12. Original + 1.0% Tego-Glide 100	Polyethermodified silicon oil
13. Original + 1.0% Silicon Oil MA (Bayer)	Low-molecular-weight polydimethylsiloxane with highly volatile portion
14. Original + 0.1% Silicon Oil L7500	Silicon oil with polyether side chains
15. High-molecular-weight cross-linked polydimethylsiloxane	

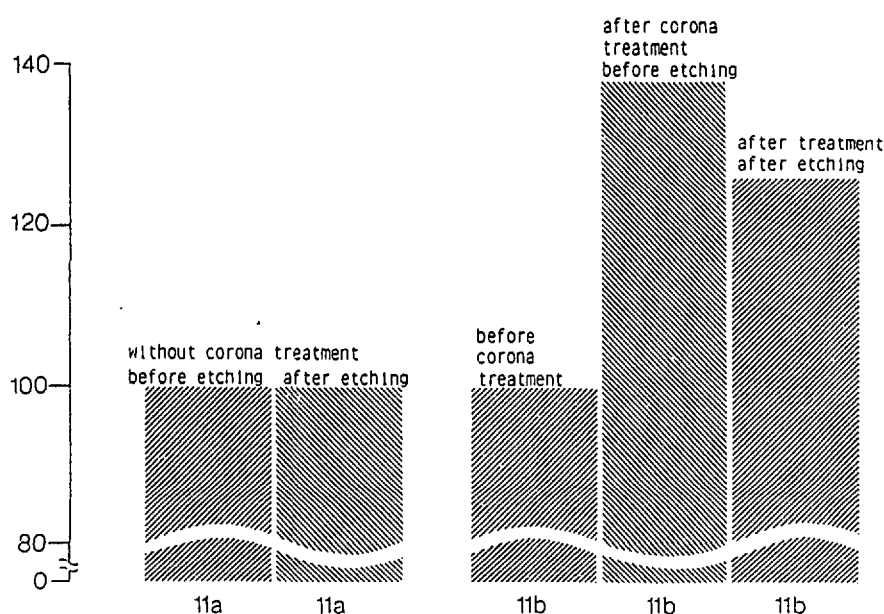


Fig. 11. Relative nitrogen content of the surface before/after Ar ion etching

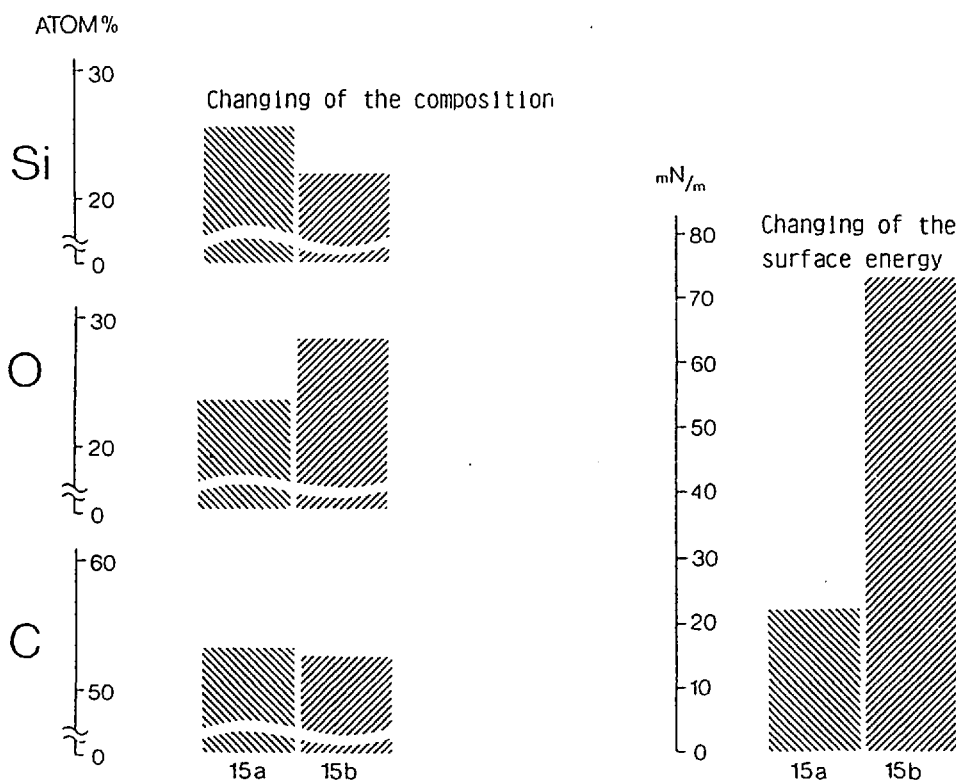


Fig. 12. The effect of corona treatment of the surface of polydimethylsiloxane

Fig. 5 illustrates that two or three-fold treatment, i.e. in repair, is barely altering the positive result. However, after multiple applications or long-lasting treatment, peel adhesion may impair as a result of damage to the surface molecules due to further bond scission or oxidative degradation from continued application of the corona treatment. The surface tension is monotonically increasing with the number of treatment, demonstrating that the improvement in wetting is not necessarily parallel to the peel adhesion, since a treated, weak boundary layer may still increase its wetting properties, while a strength decrease occurs. Fig. 6 shows that with a constant level of current, the speed can be varied between 3 and 30 m/min without affecting the strength performance, since the minimum requirement of surface energy for good wetting is always fulfilled.

Contrary to the corona treatment of polyolefins, the increase in adhesion is not dependent on the time interval between the corona treatment and the application of the adhesive or assembly of the treated parts. It must, however, be ensured that there is no contamination of the treated surface between treatment and bonding. Fig. 7 shows that, even after an interval of 7 d, there is practically no loss of the adhesive effect.

The exceptional effectiveness of the corona treatment and independence of the composition of the paint system is illustrated in Fig. 8, which shows that the surface tension of the paint samples increases critical levels of about 30 mN/m before treatment to over 50 mN/m after the treatment, independent of the quantity and kind of additive. Such surfaces can also be perfectly wetted with very polar bonding agents, even with aqueous systems. Although the adhesion to the model top coats is sufficient with polyurethane adhesives without pretreatment, the potential of this method may be clearly demonstrated.

The results of improved adhesion achieved by the corona process on 5 USA high-solid top coats are quoted as representative for a large number of serial top-coats in Fig. 9. The left and right columns of each pair of columns describe the results before and after being treated with the corona process. The left-hand pair of columns illustrates the adhesion of the TOGOCOLL FH 500 direct glazing adhesive in general use combined with the use of a priming agent, the middle pair of columns shows the primerless adhesion the same material before and after being treated and the right-hand pair of columns illustrates the adhesion of a new generation direct glazing adhesive containing additional adhesion additives (TOGOCOLL FH 500 DI).

Although neither of the two adhesives show sufficient adhesion to these top coats without priming before treatment, adhesion values higher than those required by industry are received after the corona

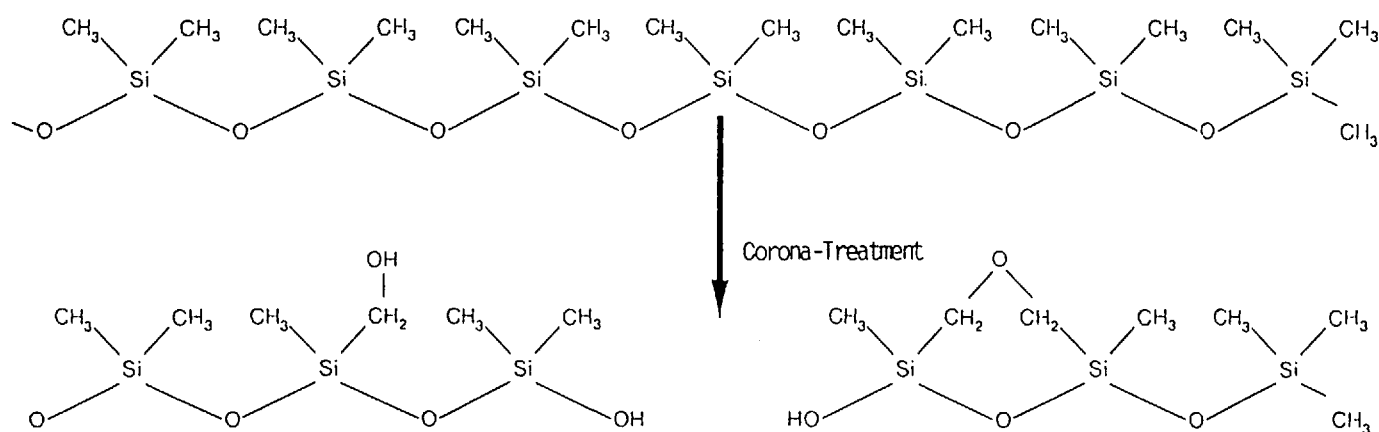


Fig. 13. Effect of the corona treatment on the structure of polydimethylsiloxane

treatment. The fact that, on certain untreated high-solid top coats, perfect adhesion cannot even be achieved, when using primers, is also very illustrative. This problem led to certain American manufacturers covering the bonding areas of the window flanges with masking tape after the application of the electrocoat and later bonding to electrocoat layers which do not have the adhesive problems of high-solid top coats.

Explanations

Our examinations of the paint surfaces before and after the ETA treatment by means of IR spectrometry, electron microscopy, and ESCA-analysis have shown chemical changes in a few molecular surface layers of the top coat to be responsible for the adhesive effect. As expected, the oxygen content of the layers increased through the corona treatment, but the even bigger increase in nitrogen due to surface

nitration by formation of nitric oxides was unexpected and astonishing. In Fig. 10 the changing amounts of silicon, nitrogen, oxygen, and carbon before and after the corona treatment are displayed. The oxygen increase due to the corona treatment is between 10 and 20%, whereas the nitrogen content increases by 40 to 100% depending on the paint system. To get a better picture an Ar ion etching on the corona treated surface has been carried out (Fig. 11). This etching process is able to level off the very top nanometers of the surface and thus may reveal the order of the nitrated layers. By the Ar etch in this case, an approximate layer of 20 nanometers was carried away. Therefore we can estimate the modified surface layer thickness to be of the order of 50–60 nanometers.

It is also remarkable to note that, through the corona treatment, silicon additives on the surface are partly changed to adhesion-compatible oxidised products and partly degraded to volatile fragments.

In Fig. 12, the effect of corona treatment on the surface of polydimethylsiloxane is illustrated by the changing amount of silicon, oxygen, and carbon in the very top surface layers measured by IR spectroscopy. The change of the molecular structure of the polydimethylsiloxane into hydroxymethyl-, hydroxy- and ether-group-containing siloxanes was determined by ESCA analysis and illustrated in Fig. 13.

Summary

Through the corona treatment, adhesion resistant top coats become adhesion-compatible substrates for polyurethane adhesives. This reliable, purely physical process which can be automated, replaces the expensive, manual, wet pretreatment using body primers containing solvents which are inflammable.

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Recent Developments in the Field of Powder Coatings

Jean-Marie Loutz*

1. Introduction

In 1960, the first electrostatic spray experiments were carried out allowing the application of powder paint to cold and intricately shaped metallic objects. With this process, it is possible to apply films, with a thickness varying from 40 to 300 μm , in one single coat. At that date powders based on epoxy resins and dicyandiamide hardener were used in the functional

coating field, e.g. for pipe protection. Due to photodegradation of bisphenol A based epoxies, leading to chalking and loss of gloss, there was a need to develop alternative mixtures of resins and cross-linkers for the decorative coating market.

Following the emergence of several possibilities to overcome the drawback, associated with bad weatherability, various directions were taken on the different continents. The USA and Canada chose the so-

called polyurethane powders (in fact the urethane is made *in situ* from blocked isocyanate and hydroxylated polyester), because the dominant position of the automotive industry for which the flow of the paint is of prime importance. Japan opted for acrylic copolymers, recognized for their resistance to UV-induced degradation, containing glycidyl methacrylate which are cross-linked by means of long chain aliphatic diacids. Trials conducted with these systems failed in Europe either due to the volatile emission of caprolactam during baking or to poor mechanical proper-

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