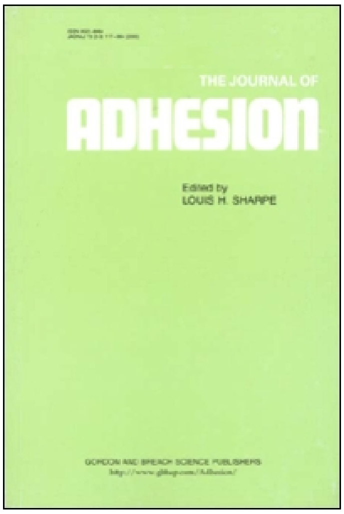

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Studies on Suspension and Emulsion

Part 4: Interaction between Polymer (Especially, SBR) and Filler Appreciated by Phase Separation

(Received Dec. 13, 1967)

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Abstract

In the previous paper¹⁾, it was reported that we could know the polymer-filler interaction from the localization of filler in the separated phases and from the amount of polymer absorbed onto the particle of fillers. In this paper, we used especially rubbers as polymer, and tried to obtain further the knowledge of the specific property of SBR observed in the previous paper. The results for α-Fe_2O_3 and various kinds of fillers were as follows. (a) Various kinds of fillers: (1) Two-rubber system; fillers remained much more in the phases of polymer with greater adsorption than in that with less adsorption. (2) The magnitudes of the polymer adsorption onto the particle of four kinds of filler (Titanium oxide, Calcium oxide, Crown clay, α-Fe_2O_3) decreased in the following order, namely, Hycar 1042>Aron rubber>High styrene>Natural rubber>SBR>Neoprene AC. (b) α-Fe_2O_3: (1) Rubber-polyvinylacetate (PVAc) system; α-Fe_2O_3 remained in the rubber phases with less adsorption. (2) As the roll-milling time of SBR was increased, the adsorption increased a little in the initial stage and soon it became constant. The roll-milling of SBR gave no effect on the localization of α-Fe_2O_3 in SBR phase of SBR-PVAc system. (3) The adsorption-desorption isotherm; the hysteresis curve was observed for SBR, but not for PVAc. (4) When SBR and α-Fe_2O_3 were roll-milled together, the mixture could not dissolve in benzene.

These results supported further the conclusion in the previous paper that SBR has the stronger interaction with α-Fe_2O_3.

J. THE ADHESION SOCIETY OF JAPAN, Vol. 4, No. 1, p. 8 (1968)

Effects of Impurities on Performance of Alkyl α-cyanoacrylate as Adhesive

(Received Dec. 12, 1967)

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Abstract

It was found that there were alkyl cyanoacetate, sulfur dioxide and water as impurities contained in alkyl α-cyanoacrylate, which had been prepared by our improved method described in the previous papers.

It was a surprising new fact that a small amount of water were contained in the monomer without catalytic effect for anionic polymerization of alkyl α-cyanacrylate; because it had been well known that the anionic polymerization of such monomer were catalized by water as well.

Consequently, it was possible that water existed in the monomer without polymerization, if water content was up to about 0.5% in methyl α-cyanoacrylate.

Further, effects of these three kinds of impurities on performance of alkyl α-cyanoacrylate as adhesive were studied.

The results were obtained as follows;

1) The impurities effected on performance of alkyl α-cyanoacrylate as adhesive.
2) Alkyl cyanoacetate in the monomer decreased joint strengths of the monomeric adhesive markedly.
3) Sulfur dioxide in the monomer retarded the set time.
4) The presence of a small amount of water shortened the shelf-life.

Thus, these effects were investigated quantitatively and discussed for those causes.

J. The Adhesion Society of Japan, Vol. 4, No. 2, p. 70 (1968)

Studies on Acrylic Adhesives

Part 2. Tensile Shear Strength of Polyester Film Bonded with Polyalkylacrylate

(Received Oct. 18, 1966)

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Abstract

Polyester film was bonded together with benzene solution of polyalkylacrylate. The relationships between the tensile shear strength of the laminate and the open time, and then the concentration of the adhesive solution were investigated. (1) In the case of the same open time and concentration, the tensile shear strength decreased in the following order: polyethyl->methyl-≧butyl->2-ethyl-hexyl-acrylate (2) The mode of failure was always cohesion for butyl and 2-ethylhexl, and the mixture of adhesion and cohesion for methyl and ethyl. (3) The tensile shear strength increased with longer open time. This is due to the evaporation of solvent from adhesive. (4) The tensile shear strength decreased with higher concentration of adhesive solution. This shows that the tensile shear strength decreases with thicker adhesive layer. (5) In the case of heat drying, the tensile shear strength decreased in the following order: polymethyl->ethyl-≧butyl->-2-ethylhexyl-acrylate. (6) If the adhesive, concentrated solution of polyalkylacrylate, is Newtonian and the shear failure phenomenon acts upon the Newton's flow law, the tensile shear strength (τ_S) should be given by

$$\tau_S = D\eta = \frac{dv}{dy} \cdot \eta \ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Where D is the slope of velosity, dv is the tensile velosity, dy is the thickness of adhesive, and η is the viscosity of adhesive. If dv and dy are constant, τ_S should be proportional to η. Here, the viscosity of polyalkylacrylate decreases with longer alkyl group. Therefore, the lower tensile shear strength with longer alkyl group is expected from the equation (1). Moreover, the higher tensile shear strength with longer open time would be due to the higher viscosity of adhesive with more evaporation of solvent. In addition, if dv and η are constant, τ_S should be in inverse proportion to dy. Therefore, the lower tensile shear strength with higher concentration of adhesive solution or thicker layer of adhesive is expected from the equation (1).

J. The Adhesion Society of Japan, Vol. 4, No. 3, p. 140 (1968)

Studies on Acrylic Adhesives

Part 3. Peel Strength of Polyester Film Laminate Bonded with Polyalkylacrylate

(Received Mar. 15, 1966)

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Abstract

The peel strength of polyester film laminate bonded with benzene solution of alkylacrylate homopolymers has been investigated. The results have been obtained: (1) The peel strength decreases according to the following order: polyethyl->methyl->butyl->2-ethylhexyl-acrylate. (2) For butyl and 2-ethylhexyl, the failures of both are perfectly cohesion. Their peel strengths are approximately constant (300-400g/25mm for butyl and 50g/25mm for 2-ethylhexyl) without regard to the open time of 10-60 minutes and the solid content of polyacrylate solution of 10-40%. (3) For methyl and ethyl, the failures of both are the mixed mode of adhesion and cohesion. Their peel strengths vary with the open time and the solid content (1-2 kg/25mm for methyl and 1-4 kg/25mm for ethyl). This variation of peel strength is based to the differences of the solvent content remained in adhesives and the thickness of adhesives on peel test. The peel strength increases or decreases with the content of remained solvent, and increases with the thickening of adhesive layer.

J. The Adhesion Society of Japan, Vol. 4, No. 3, p. 148 (1968)

Pressure Effect on Adhesion

On the Rate of Adhesion at Initial Stage

(Received April 30, 1968)

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Abstract

Under the conception of entanglement, the initial rates of adhesion are investigated. When the epoxy resin-amine systems are used as an adhesive, the relation of cure time *and initial, bond strength ratio are theoretically given as follows, in* $S(t)/S(t_0) = \alpha \cdot \Delta t + 1$, where, $S(t)$ and $S(t_0)$ are bond strength at cure time (t) and (t_0), respectively, α is the increasing coefficient of entanglement $\Delta t = t - t_0$. And the relation between cure pressure and bond strength ratio are theoretically given as follows, in $\ln S(\rho)/S(\rho_0) \ln \zeta + \beta \cdot \rho$, where $S(\rho)$ and $S(\rho_0)$ are bond strength under cure pressure (ρ) and (ρ_0) respectively, β and ζ are constant. The experimental results for the adhesion of copper with epoxy resin-amine systems justified the theoretical prediction mentioned above.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 191 (1968)

The Swelling of the Adhesive Layer by Water Adsorption and its Bonding Strength (I)

(Received Aug. 3, 1968)

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Abstract

This paper deals with the experimental research about the influence on the bonding strength between metal to metal when the bonded piece is dipped in water. The adhesive consisted of liquid epoxy resin and triethylenetetramine as curing agent. After the bonded specimens were dipped in water at room temperature, 50, 70, and 90°C for a given time, they were tested for tensile strength at 25°C. The bonding strength decreased in proportion as the dipping time increased. That is, the bonding strength of the specimens seemed to depend on the water adsorption of the adhesive layer, and therefore we introduced the following relation between the bonding strength F after dipping in water, and water adsorption of adhesive, M,

$$F = F_0 - k_1 M \cdot E \cdot (1 - k_2 \log C)$$

where F_0 is the bonding strength before dipping in water, E is the modulus of the adhesive, C is dipping temperature and k_1, k_2 are constants.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 196 (1968)

The Swelling of the Adhesive Layer by Water Adsorption and its Bonding Strength (II)

(Received Aug. 3, 1968)

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Abstract

In the previous report, the bonding strength after dipping in water was shown by the following equation,

$$F = F_0 - k_1 \cdot M \cdot E \cdot (1 - k_2 \log C)$$

As its strength was largely depended on the water adsorption of the adhesive layer, the Filler was added in epoxy adhesives, in order to decrease the water adsorption of it. The water adsorption of adhesive layers including the filler was smaller than without filler, and within the experimental conditions, the water adsorption of the resin in the adhesive with filler was the same value as one in the adhesive without filler, because the filler (SiO_2) did not adsorp water in it.

Therefore, the following equation can be derived for the bonding strength,

$$F = F_0 - k_1 \cdot M \cdot E \cdot \left(1 + k_2 \frac{W_2}{W_1}\right)(1 - k \cdot \log C)$$

where W_1, W_2, are respectively weight of resin and filler and k_1, k_2, k_3 are constants.

It is concluded that the bonding strength depends on the swelling of the adhesive layer, especially the resin in the adhesive by water adsorption.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 201 (1968)

Temperature Dependence of Peeling Strength of Solvent-type Paint Films

(Received Aug. 8, 1967)

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Abstract

The relation between glass transition temperature and the peeling strength (by the peeling test method) was examined for the solvent-type paint films casted on the glass plates, and the following relation was found, $Tp \overset{>}{\leqq} Tg$ where Tp is the temperature measured on a film, at which a max. peeling strength is given, and Tg is the glass transition temperature measured by the torsional pendulum method or the swinging beam method.

These results indicate that the peeling strength of the films is dependent on its complex visco elastic properties.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 206 (1968)

Mechanics of Peeling

5. Temperature Dependence of Peeling Strength of Polyisobutylene

(Received No. 2, 1967)

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Abstract

Experimental data for temperature dependence of peeling strength of polyisobutylene (PIB) and various adherend systems have been obtained as functions of peeling rate over a range of temperature far above the glass transition point of PIB. The data were analyzed in terms of the theory previously developed. It was found that the data are well described by using three essential parameters, P_0, the value of peeling strength extrapolated to zero peeling rate; $P\infty$, the peeling strength at infinite peeling rate; and τ, the relaxation time of three element visco-elastic model of adhesive interlayer.

The values of P_0 and $P\infty$ decreased with increase of temperature and the ratio $P\infty/P_0$ reached a maximum at a specified temperature.

The temperature dependence of τ was similar to that of the shift factor α_T, which might be derived from transient or dynamic visco-elastic measurements of bulk polymers. But the universal constants derived from the peeling data to fit the WLF equation differed from the original values of PIB. The τ for the system of PIB-Polyvinyl chloride (PVC) showed an abrupt change at a temperature in the vicinity of which the PVC undergoes glass transition. Similar phenomena were observed for PIB and Polytetrafluoroethylene (PTFE) system. It may be concluded that the mechanical behavior of adhesive interlayer are not determined by the bulk property of flexible members (PIB) but are determined by the composite mechanical properties of flexible members and adherend systems.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 212 (1968)

Relation Between Peel Strength and Thickness of Adhesive Layer

(Received July 6, 1967)

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Abstract

The relations between 180° peel strength and thickness of adhesive layer were investigated for polypropylene film-steel sheet laminate bonded with polyisobutylene or butyl rubber adhesive and polyvinylchloride sheet-steel sheet laminate bonded with polyethylacrylate adhesive.

It was found that the relation between peel strength (P) and thickness of adhesive layer (l) was approximately expressed by the following formula:

$$P/l = a - b \log l \tag{1}$$

$$\log P/l = a' - b' \log l \tag{2}$$

These relations correspond to the general formula on failure reported by Griffith, Gooding, Higuchi, and others.

Under a constant crosshead speed, with decreasing thickness of adhesive layer, true fracture speed may increase, the mode of fracture may change from fluid or ductile to brittle fracture, and the site of fracture may transfer from adhesive layer (cohesive failure) to the interface of adhesive and adherend (adhesive failure). Assuming that the true fracture speed is inversely proportional to the thickness of adhesive layer, the following may be derived from the equation (1),

$$P/l = \alpha - b \log t_b \qquad (3)$$

where t_b is breaking time. The equation (3) corresponds to the Bueche's equation on breaking time obtained from viscoelastic theory.

The shift of fracture site from adhesive layer to the interface of adhesive and adherend with decreasing thickness of adhesive layer was observed in the case of polyvinylchloride sheet-steel sheet laminate bonded with polyethylacrylate adhesive of polymerization degree of 1,800.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 223 (1968)

A New Method of Measuring Tackiness

(Received May 12, 1967)

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Abstract

For the measurement of the tackiness of viscous materials, we designed a testing apparatus attached to a tensile tester with constant speeds of elongation. Solutions in toluene of 10 ~ 15% by weight of high molecular weight polyisobutylene (PIB), $\overline{M}v = 1.32 \times 10^6$, blended with a low molecular weight PIB, $\overline{M}n = 1.0 \times 10^3$, or a hydrogenated rosin were used as samples.

Sample solutions were coated on a cellophane film and dried to remove solvent.

An unbonded strain gauge was fixed to a movable cross bar of the testing machine.

A flat surface of a cylinded brass connected to the sensitive part of the strain gauge was pushed upward onto the tacky surface of the sample for a definite constant time.

Then, the cross bar was lowered again, and a force necessary to separated the brass cylinder from the sample surface was recorded.

The maximum force f_b at the time of t_b of the force-time relation recorded on a chart was determined as a measure of the tackiness.

The relation between f_b and t_b satisfies the equation $f_b \propto t^{-}{}_{b}{}^{1/2}$, derived by D. D. Eley.

The f_b depends on the time (t_c) and the pressure at the contact, the temperature and the rate at the separation.

In this experiment, however, at constant value of p_c and t_c, r was varied from 1 to 20 mm/min, and the temperature from 10 to 50°C.

As a result, it was found that f_b increases as the increase of r and the rosin content.

The activation energy, obtained from the slope of plots of f_b against reciprocals of the absolute temperature shows the value of 2 ~ 6 Kcal/mol, which is similar to the activation energy of the viscous flow.

J. The Adhesion Society of Japan, Vol. 4, No. 4, p. 229 (1968)

Studies on Rheology of Pressure Sensitive Adhesive Tapes

IV. Viscosity of the Blends Composed of Natural Rubber and Tackifying Resin

(Received Sept. 4, 1967)

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Abstract

The mooney viscosities of blends composed of unvulcanized natural rubber and various tackifier resins were measured at various temperatures. Apparent activation energies of flow are analyzed by the following equation on adittivity

$$Evis = C_1\, Evis_{-1} + C_2\, Evis_{-2} + K\, C_1C_2$$

where $Evis_{-1}$, $Evis_{-2}$ are activation energies of flow, and C_1, C_2 are weight fractions of components.

The viscosity of blends at room temperature shows the minimum at a certain resin concentration, and it varies with resin type, of which reasons are discussed using Boyer's equation.

J. The Adhesion Society of Japan, Vol. 4, No. 5, p. 256 (1968)

Reaction of Alkyl Cyanoacetate with Formaldehyde

(Received Feb. 10, 1968)

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Abstract

When methyl cyanoacetate was reacted with formaldehyde in the presence of catalytic piperidine in the methanol solution, it was showed that the reaction was first order in the concentration of methyl cyanoacetate and formaldehyde respectively. But the reaction was not always first order in the concentration of catalyst in the methanol solution.

Concerning the catalyst, it was found that the over all reaction turned continuously from first order to ½-order. This change was due to the initial concentration ($= \alpha$ mole/l) of catalytic piperidine to a small quantity of water which was contained in the methanol solution of formaldehyde.

It was found that the reaction was first order under $\alpha = 4 \sim 6 \times 10^{-2}$, ½-order over its value.

J. The Adhesion Society of Japan, Vol. 4, No. 5, p. 260 (1968)

On the Stress-Strain Curves of Steel-Steel Lap Specimen Adhered with Alkyl α-Cyanoacrylates

(Received Aug. 8, 1967)

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Abstract

To study on the difference in the mechanical properties of the cured adhesive phase based on alkyl group for the homologous series of α-cyanoacrylate monomers, stress-strain curves of steel-steel lap specimen adhered with seven homlogs from methyl to i-amyl monomer respectively, in the case of tensile shear and compressive flexure, were measured.

Results were as follows.

(1) In these stress-strain curves no essential difference by alkyl group was observed, and their shapes were nearly similar figures.

(2) Therefore, failure strength or maximum force relates directly to failure energy, furthermore linearly to cohesive energy density of cured adhesive phase separating two series of n-homolog and i-homolog.

Thus, there is no possibility in which any essential problems by the difference of alkyl group is brought.

(3) Then, evaluating these homologs as the base of polymerization type adhesive, excellent monomer may be lower alkyl, especially methyl, ethyl, and i-propyl α-cyanoacrylate.

(4) Above conclusion agree with results of the previous report well.

J. The Adhesion Society of Japan, Vol. 4, No. 8, p. 478 (1968)