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Polymer-Polymer Adhesion in the Presence of Compatibilizer*

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The influence of compatibilizers in three types of joints-natural rubber (NR)-polyethylene (PE), silicone rubber (Si)-ethylene propylene diene rubber (EPDM) and hydrogenated nitrile rubber (HNBR)-nylon has been demonstrated. The compatibilizers increase the peel strength in every case. The results are explained with the help of new approaches based on (1) strain energy density, (2) reptation scaling theory. It is observed that the diameter of the strained tip for NR-PE and Si-EPDM systems increases in the presence of compatibilizers. Also, the peel strength of the HNBR-nylon joint has a first power dependence on the interfacial thickness.

KEY WORDS: Adhesion; adhesive joints; compatibilizer; rubber; polymer blend; reptation theory; peel strength.

INTRODUCTION

The ability of two polymers to form a strong joint and to resist separation against various forces is important for designing polymer blends and composites with improved properties. When a polymer plate (A) is adhered to another similar or dissimilar plate (B), the following (and other) phenomena can occur:

- (i) dispersion and/or polar forces may operate at the interface.
- (ii) diffusion of A or B or both may take place, and
- (iii) there may be interfacial crosslinking.

The types of polymers and their ingredients, temperature and rate of contact/testing, etc., determine the type of interface and its strength.

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Polymer-polymer adhesion has been studied by many workers, including the present authors¹⁻¹¹. These studies can be divided into two broad aspects: (i) investigation of thermodynamics, wetting, and interfacial thickness, and (ii) formation and behaviour of the adhesive bond/interface under a variety of conditions. There is rarely any systematic study correlating these two aspects, although attempts have been made. The situation is further complicated when a compatibilizer is added to one (or both) of the polymers. The compatibilizer renders a mixture of two or more polymeric materials homogeneous so as to prevent separation or stratification of the components during the lifetime of the product and to improve ultimate properties by making the polymers in a blend less incompatible. Coran¹² discussed the usefulness of technological compatibilization. A large number of blends has also been made by using compatibilizers. However, their effect on interfacial thickness and adhesion behaviour is not well understood.

The objective of the present study is to understand the factors responsible for and the mechanism of polymer-to-polymer adhesion in the presence of compatibilizer. For this purpose, three types of joints, natural rubber (NR)-polyethylene (PE), silicone rubber (Si)-ethylene propylene diene rubber (EPDM) and hydrogenated nitrile rubber (HNBR)-nylon, using a number of physical and chemical compatibilizers as discussed in the text, have been chosen. These represent joints where the glass transition temperature of both of the components is below room temperature and joints where one of the components has a T_a below room temperature.

EXPERIMENTAL

Materials

System-I

Natural Rubber (NR): ISNR-5 was supplied by the Rubber Board, India. Molecular weight = 780000, intrinsic viscosity in benzene at $30^{\circ}C = 4.45 \text{ dl/g}$.

Polyethylene (PE): Indothene 16 MA 400 was supplied by IPCL, India. Density = 916 kg/m³, melt flow index = 40 g/10min.

The compatibilizers used for NR/PE joints were:

Chlorinated Polyethylene (CPE): CPE with 36% chlorine was supplied by Dow Chemical, USA; specific gravity = 1.16, Mooney viscosity, $ML_{(1+4)}$ at $121^{\circ}C = 80$.

Epoxidized Natural Rubber (ENR): ENR25 was supplied by MRPRA, U.K. Density = 970 kg/m³, epoxidation level = 25 mol %, Mooney viscosity $ML_{(1+4)}$ at $100^{\circ}C = 110$.

Modified Polythylene (PEm): The modification of polyethylene was carried out by melt mixing PE with 5 parts maleic anhydride (MAH) at 150°C in a Brabender Plasticorder at 60 rpm. After they were well mixed, 0.82 part of benzoyl peroxide was added and the mixing continued until the decreasing mixing torque levelled off. The MAH content was 25%⁷. Details of the procedure for preparation and characterization were reported in earlier papers^{7, 13}.

System-II

Silicone Rubber: JSR Silicone EH5270, heat cured type silicone rubber, specific gravity = 1.31, was supplied by Japan Synthetic Rubber Co. Ltd., Japan.

EPDM: Kelton 520, ethylene content = 55 mol %, diene (DCPD) content = 4.5 mol %, density = 860 kg/m^3 was supplied by DSM, Holland.

The compatibilizers used for System-II were:

Ethylene Methylacrylate Copolymer (EMA): Methyl acrylate content 21%, density = 940 kg/m³, melting point = 81°C, crystallization temperature = 67° C, was supplied by Exxon Chemical, USA.

Sulfonated EPDM (S-EPDM): Ionomer 2590 was supplied by Uniroyal Chemical Co., USA, density = 1120 kg/m³, Mooney viscosity $ML_{(1+4)}$ at 100°C = 45, average number of SO₃⁼ groups/molecule = 13, ionic groups = 2.7wt%.

Maleated EPDM (MA-EP): Royaltuf 465, specific gravity = 890 kg/m³, total maleic anhydride/acid = 1%, Mooney viscosity $ML_{(1+4)}$ at $125^{\circ}C = 60$, iodine value = 17, was supplied by Uniroyal Chemical Co., USA.

Acrylamide-grafted Silicone Rubber (Am-g-Si): 100 g silicone rubber, 9.0 g acrylamide and 0.25 g ammonium persulfate were reacted in a Brabender Plasticorder, PLE-330, at 80°C and 100 rpm speed for 15min. The grafting level was 26 m.mol per mol of siloxane, as determined from an IR study¹⁴.

System-Ili

Hydrogenated Nitrile Rubber (HNBR): Zetpol 1020, $T_g = 20^{\circ}$ C, iodine value = 25, $M_n = 105000$, was supplied by Nippon Zeon Co. Ltd., Japan.

Nylon (MXD6): $T_g = 90^{\circ}$ C, $Tm = 235^{\circ}$ C, $M_n = 25300$, was supplied by Mitsubishi Gas Chemical Co., Japan.

2,5-dimethyl (t-butyl peroxy) Hexane: It was used as crosslinker.

The compatibilizer used in this study was:

Liquid Carboxy-modified Nitrile Rubber (LXNBR): 20.0 wt.% ACN, 0.08 equivalent phr carboxyl content, viscosity at $50^{\circ}C = 20,000$ cps, was supplied by Nippon Zeon Co. Ltd., Japan.

Preparation of Samples and Measurement of Peel Strength

For System-I, CPE, ENR and PEm were mixed with natural rubber or polyethylene, as the case might be, in a Brabender Plasticorder. Fabric-backed rubber sheets (2mm thick) and plastic sheets (1mm thick) were prepared in a hydraulic press at 100° C and 150° C, respectively. Then, the plastic sheet was put over the fabricbacked rubber sheets in between which a cellophane paper was partially introduced to obtain the demarcation line. The assembly was heated for 10 min at 100° C followed by 15min at 150°C and was subsequently cooled. For System-II, the fabric-backed rubber sheets (1.5 mm thick) containing appropriate compatibilizers were joined together as given above. The assembly was then heated at 150°C for 10min under mold pressure.

For System-III, the compatibilizer and the peroxide were mixed with HNBR on an open roll mill and smooth sheets of 1.5mm thickness were made at 100° C in a hydraulic press. Nylon sheets (1mm thick) were made at 250°C under pressure. These were then joined under mold pressure at 100° C for 5 min and subsequently at 250° C for 5 min. The joints were cooled under pressure.

The test pieces were punched out from the molded sheet at a width of 20 mm by a hollow punch. Peel testing, Figure 1a, at 180° was done in a Zwick UTM (1445 model) at different rates and temperatures. The peel strength, G_a , was calculated using the following relation,

$$G_a = \frac{2F}{\omega} \tag{1}$$

where F is the force required to separate the layers and ω is the width of the specimen.

Preparation of Samples and Testing of Lap Shear Strength

Silicone rubber or EPDM in System-II was mixed with the desired amount of the compatibilizer in the Brabender Plasticorder for 5 min at 10 rpm and 120°C. The stocks were molded for 5 min at 120°C. 75 mm \times 25 mm strips of EPDM were cut from these specimens. Silicone rubber was cut as 25mm squares. The lap shear



FIGURE 1 Adhesion tests used in this study. (a) T-peel test; (b) Single lap shear test.

specimens were prepared by sandwiching the silicone strip between two EPDM strips initially by hand. Finally, the specimens were aluminium backed and hot processed in a press for 10 min at 150°C for systems containing Am-g-Si, MA-g-EP and s-EPDM and 180°C for systems with EMA (Fig. 1b). The samples were tested at 25°C at a crosshead speed of 25 mm/min after conditioning for 24 h. The force recorded was divided by the area of joining to obtain the value of lap shear strength.

Interfacial Parameters

Interfacial thickness between nylon and HNBR in System-III was measured by an automated Ellipsometer (EL-8, Optec Co., light of 632.8 nm wavelength applied at an angle of 70°). Nylon was first melt pressed to form a flat substrate (0.5 mm thick). Clean HNBR solution in monochlorobenzene (6% by weight) was then spin coated onto the nylon at room temperature. The bilayer specimen was then inserted into a hot chamber at 250°C in nitrogen atmosphere to measure the retardation, Δ , and the reflection ratio, ψ . For data analysis, a four-layer model and the well-known Drude Equation, as described earlier, ^{5,10,11} were used. Numerical calculation was carried out by a Hitachi Computer, HITAC M660K.

Measurement of Strain Energy Density

Dumbell-shaped specimens (BS-E type) were cut with a hollow punch from the test slabs. Tensile tests were performed at room temperature in the Zwick UTM at different crosshead speeds. The strain energy density was calculated from the area under the stress-strain curve.

RESULTS AND DISCUSSION

(A) Natural Rubber-Polyethylene Joints (System-I)

The values of peel strength between NR and PE with chlorinated polyethylene (CPE) as a physical compatibilizer and epoxidized natural rubber (ENR)/modified PE (PEm) as a chemical compatibilizer, at different rates and temperatures, are shown in Table I. For comparison, the results of NR/ENR//PE and NR//PEm/PE at one temperature and rate are included. The peel strength between NR and PE is 140 J/m². The incorporation of chlorinated polyethylene increases the peel strength due to its structural similarity with polyethylene and its rubbery characteristics. As a result, it can interact physically with both the components. There is a further increase in peel strength in the case of NR/ENR//PEm/PE. This is certainly due to the interfacial chemical reaction between the components through modifiers as shown below, Scheme $1a^{4,7}$.

The interaction also changes the failure behaviour. For example, the modified systems show wider variation of force with continuous failure initiation and arrest than does the control system NR//PE. The strength values are also a function of the

	Temperature				_		
System	Rate	25°	50°	75°C	Observations		
NR//PE	50	100	80	45	Sharp interface at all temperatures & rates		
	200	140	100	50			
	500	140	110	55			
	750	150	110	60			
NR/CPE//PE	50	145	75		Stick-slip failure at low rates and		
	200	155	80		high temperature		
	500	200	100	160			
	750	250	100	140			
NR/ENR//PE	200	160	_		Smooth failure		
NR//PEm/PE	200	175			Smooth failure		
NR/ENR//PEm/PE	50	350	250	200	Stick-slip failure at low rates and		
	200	430	265	230	high temperature		
	500	500	280	250			
	750	520	300	260			

TABLE I	
Values of peel strength in J/m^2	of various joints ⁴

test rates and temperatures as shown in Table I and discussed in an earlier communication⁴.

The peel strength increases with an increase in the test rates (Table. I). As the temperature is increased, the joint strength decreases for almost all the systems. At very high temperature (100°C), the plastics start to soften and undergo rupture. In general, NR//PE shows "smooth" failure at all rates, whereas NR/ENR//PEm/PE shows "stick slip" failure at low rates and high temperatures. Such transition from "smooth" to "stick-slip" failure is due to change from an elastic response (interfacial failure) to a viscous flow response (cohesive failure). However, the changes in joint strength due to changes in rates and/or temperatures could be accounted for by the viscoelastic properties of the interfacial region.

(B) Lap Shear Strength Between Silicone Rubber and EPDM and the Effect of the Compatibilizer (System-II)

The values of lap shear strength between silicone rubber and EPDM in the presence of various compatibilizers are shown in Table II. 1, 3 and 5 parts of AM-g-Si were mixed with silicone rubber. Similarly, 1, 3 and 5 parts of sulfonated EPDM and MA-g-EP blended with EPDM were used. The lap shear strength of the control is 100 MPa. With the addition of the compatibilizer, either S-EPDM or MA-g-EP, in EPDM, the lap shear strength increases. The higher the concentration of the compatibilizer, the higher is the lap shear strength. For example, 1phr of S-EPDM doubles its value. With the same amount of MA-g-EP, the strength is 2.6 times that of the control joint. With 5phr of MA-g-EP, the joint strength registers a value of 416MPa. The higher strength may be ascribed to the dipole-dipole interaction or chemical reaction at the interface and a higher level of interaction leads to higher lap shear strength. In fact, higher chemical interaction is further supported by the low values of the joint strength obtained when lower molding temperature $(35^{\circ}C \text{ or})$



SCHEME 1a Interaction between natural rubber and polyethylene in presence of compatibilizer.

	Substrate with wh the compatibilized mixed and its dos	tich t was age	Temperature of hot processing,	Lap shear strength, MPa $\times 10^{-2}$
Compatibilizer	Silicone	EPDM	°C	
None				1.0
Acrylamide-g-silicone	1	1	150	1.99
in silicone rubber and	3	3	150	3.13
sulfonated-EPDM in EPDM	5	5	150	3.18
Acrylamide-g-silicone	1	1	150	2.68
in silicone rubber	3*	3*	150	3.56
EDM rubber	5	5	150	4.16
Er Divi Tubbei Ethylene Methylacrylate	25		180	2.2
Emplene Weinylaer ylate	5	_	180	3.8
	10		180	4.8
	15		180	5.3

TABLE II	
Lap shear strength between silicone and EPD!	M in the presence of compatibilizer

*The values of lap shear strength of 1.77×10^2 and 1.18×10^2 MPa were obtained when the molding temperature was reduced to 70° and 35°C, respectively.

 70° C) is used. The possible interaction between the components may be as illustrated in Scheme 1b&1c.

Similarly, a marked improvement in lap shear strength is found when EMA is added into silicone rubber (Table II). The lap shear strength increases also with the increase in EMA concentration. The compatibilizer plays the role of strengthening the silicone phase (90% increase in tensile strength by 10 parts of EMA) and also increases the surface energy (the work of adhesion increases from 85 to 109 mJ/m²)¹⁴. Further, the compatibilizer can react with EPDM (Scheme 1d).

(C) A New Approach to the Understanding of the Adhesive Strength

Though the results of the joints of Systems-I and -II can be explained with the help of physical or chemical interaction, it is difficult to predict the joint strength from these factors alone. This is partly because of the viscoelastic nature of the interfacial region. This is further complicated by the nature of failure of the joints. In some cases, it is "smooth"; in others it is "stick-slip", as described in earlier sections. In order to understand the behaviour, a fracture mechanics approach is adopted here.

In the case of an edge crack in highly extensible polymers, such as rubbers deformed in simple extension and shear, Thomas¹⁵ deduced a general theoretical relationship between the strain energy around the tip of the crack and the value of the tearing energy, G_c , calculated from the overall applied forces or strains on the test piece. For the case of an incision terminated by a semi-circle of diameter, d, the

For SEP-AM-g-Silicone system



SCHEME 1b Dipole-dipole interaction in modified silicone rubber and S-EPDM.

relation is:

$$G_{c} = \frac{d}{2} \int_{-\pi/2}^{\pi/2} W_{\theta} \cos\theta \, d\theta \tag{2}$$

where W_{θ} is the strain energy density at the tip at an angle to the pole (Fig. 2a). For most purposes, the equation can be adequately approximated as

$$G_c \approx dW_t$$
 (3)

where W_t is the maximum strain energy density at the tip. If the test piece is pulled until rupture occurs, W_t becomes the work to break per unit volume of rubber. The relation has been found to hold good for large strains and non-linear stress-strain relations. A large number of experimental investigations by Thomas and his coworkers confirm the approximate validity of the equation¹⁶.

If the same relation is assumed to hold in the interfacial region, the peel strength, G_a , will be given by,

$$G_a \approx d W_t \tag{4}$$

However, d is here the effective diameter of the tip of the strained interfacial region (Fig. 2b) and W_t is the work to break per unit volume of the interfacial region. Since many bonds/chains at the interface must be deformed before chain pull-out or chain fracture, G_a is a function of d and W_t . Measurement of G_a and W_t can give an idea

For MEP-AM-g-Silicone system

1. $H_3C-Si-CH-CH_2CH_2-CH H = \begin{pmatrix} 0 & 0 & NH_2 \\ H & C & (+VMQ-CH_2-CH) \\ H & C & (+VMQ-CH_2-CH) \\ 0 & 0 & NH_2 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & C-CH_2 & CH_2 \\ 0 & C-CH_2 \\ 0 & C-CH_2 & CH_2 \\ 0 & C-CH_2 \\ 0 & C-CH_2$

SCHEME 1c Chemical reaction between amide-grafted silicone rubber and maleated EPDM.



SCHEME 1d Interaction of silicone rubber and EPDM through EMA.

about the diameter of the tip of the strained interface and hence, the area of the strained interface. Such calculations have been done using Equation (4) to estimate the value of d. The values are reported in Table III. It is demonstrated that the diameter of the strained interface increases from 14 µm for NR//PE system to 34.4 µm for the NR/ENR//PEm/PE system, a 2.5 fold increase. Hence, a larger area of the interface will be strained in the latter case, causing the peel strength to increase. Similarly, there is a 270% increase from $48\mu m$ to $178\mu m$ of effective diameter, d, when AM-g-Si and MA-g-EP are used as compatibilizers. In System-I, the strain energy density has been measured on 70:20:3:27 of NR/ENR/PEm/PE and 70:20:30 of NR/CPE/PE, assuming that a greater amount of rubber will be deformed when fracturing such a joint. Similar calculations have been done using W_t on 50:50 mixtures and are shown in the same Table. The same trend is obtained although the d values are increased. In the case of System-II, 50:50 mixtures have been used for the measurement of W_{i} , as both the phases are rubbery in nature. The estimation of d can become accurate, provided we know the exact composition of the interface. However, the relative trend could be explained by using the above approach and theoretical predictions are in line with the experimental observations.



FIGURE 2 (a) Figure showing an incision terminated by a semi-circle of diameter, *d.* (b) Stretched interface before rupture. (c) Interdiffusion process at a polymer-polymer interface and reptation of chains. (only one side is shown for convenience).

System	$\overline{G_a}, J/m^2$	$W^*, MN/m^2$	<i>d</i> , µm
NR//PE	140	10.0	14.0
NR/CPE//PE	155	11.0	14.1
NR/ENR//PE	160	11.5	13.9
NR/PEm//PE	175	9.5	18.4
NR/ENR//PEm//PE	430	12.5	34.4
Silicone//EPDM	12	0.25	48.0
1AM-g-Silicone/Silicone/1MA-g- EP/EPDM	16	0.24	67.0
3AM-g-Silicone/Silicone// 3MA-g-EP/EPDM	48	0.27	178.0
3AM-g-Silicone/Silicone// 3-S-EP/ EPDM	20	0.19	105.0

 TABLE III

 Values of diameter of the deformed tip at the interface following Equation (4) at 25°C

*W values were measured on 50:50 blends in the case of silicone: EPDM system. The numerical digits indicate the amount (in grams) of the compatibilizer present in 100 g rubber. In the case of NR/PE systems (rubber/plastic blends) W values were determined on a 70:30 blend, assuming a greater amount of rubber would be deformed. W values measured on 50:50 blends generate a higher value of d. For example, $d_{\text{NR/PE}} = 41 \,\mu\text{m}$ and $d_{\text{NR/CEE/PE}} = 63 \,\mu\text{m}$.

(D) Adhesion Between Nylon and HNBR in the Presence of the Compatibilizers

The interfacial thickness between nylon and HNBR is plotted against time in Figure 3. The thickness increases rapidly with time in the initial stages and then levels off to a value of 48nm. Addition of LXNBR compatibilizer increases the thickness further. At 3 parts of LXNBR mixed with 100 parts of HNBR, the interfacial thickness becomes 79 ± 2 nm. The increased interfacial thickness between nylon and HNBR is due to increased wetting and mixing of the components at the interface with increased time and due to reduced interfacial tension. It is also due to increased interaction between nylon and HNBR through LXNBR as the concentration of LXNBR, is increased to 3 parts. Our earlier paper reported these observations on several compatibilizers⁵. The specific interaction may be depicted in Scheme 1e.

The peeling energy, G_a , between nylon and HNBR containing various amounts of LXNBR was determined. A typical force chart of the control HNBR/nylon joint is shown in Figure 4. This reflects the stick-slip behaviour of the joint. An average value of G_a was then calculated using Equation (1). The results indicate that the peeling energy increases with the concentration of LXNBR up to 3 parts. These values are in line with the interfacial thickness. As shown in Figure 5, G_a increases with the increase in the interfacial thickness, λ . When G_a values are plotted against the corresponding value of λ , using logarithmic scales for both axes, the results are found to be consistent with a linear relationship. A most important observation is that this best fit line has a slope of 1. Equation (4) predicts that the peel strength has a first power dependence on the effective diameter of the tip of the strained interface. From a geometric argument (shown in Fig. 2b), we can assume that d is directly proportional to the interfacial thickness. The higher the value of λ , the higher will be the magnitude of d. Hence, the observation in Figure 5 could be predicted from Equation (4). Such a dependence could also be derived from the reptation scaling theory as follows.



FIGURE 3 Time variation of interfacial thickness at 250° C. \bullet , bilayer specimen of nylon and HNBR premixed with 0.9 phr crosslinker. \Box , bilayer specimen of nylon and HNBR premixed with 0.9 phr crosslinker and 1 phr LXNBR. \blacksquare , bilayer specimen of nylon and HNBR premixed with 0.9 phr crosslinker and 3 phr LXNBR.



SCHEME 1e Interaction between HNBR and nylon in presence of compatibilizer.

As shown in Figure 2c, when two polymer plates are joined together, molecular contact is achieved at the interface and the chains begin to reptate via their chain ends. As time progresses, the minor chain escaping from the original tube interdiffuses at the interface. At $t \rightarrow t_{\infty}$, complete interpenetration and re-entanglement is achieved. From this model, Wool⁹ has discussed that for a polymer-polymer joint

$$G_a \sim t^{1/2} M^{-1/2} \tag{5}$$

where t is the contact time and M is the molecular weight. Since D_r the diffusion coefficient for curvilinear diffusion of the chain along the tube, is inversely proportional to M and the mean square curvilinear path diffused by the chain $\langle l^2 \rangle$ is given



FIGURE 4 Typical force chart showing stick slip failure of the HNBR-Nylon control joint.



FIGURE 5 Plot of peel strength vs interfacial thickness.

by

$$\langle l^2 \rangle = 2D_r t \tag{6}$$

Equation (5) could be written as

$$G_a \sim \langle l^2 \rangle^{1/2} \tag{7}$$

However, $\langle l^2 \rangle^{1/2}$ is proportional to half the thickness of the interfacial region in Figure 2b as shown. It may be pointed out that Bister *et al.* suggested a similar dependence of tack strength of virgin rubber-virgin rubber joints on contact time and diffusion coefficient¹⁷ ($G_a \sim (\rho/M)^{2/3} D^{1/2} t^{1/2}$, where ρ is the density). Hence,

$$G_a \sim \lambda$$
 (8)

showing the same first power dependence of G_a on λ .

CONCLUSIONS

- 1. Compatibilizers increase the peel strength of NR-PE, Si-EPDM and HNBR-Nylon joints and the lap shear strength in the case of the Si-EPDM joint. The degree of enhancement depends on the nature and the level of the compatibilizers.
- 2. The peel strength (G_a) results for the compatibilized systems could be explained with the help of increased diameter (d) of the strained interfacial region, calculated using the relationship,

$$G_a \approx d \cdot W_t$$

where W_t is the work-to-break/unit volume of the interfacial region. The diameter increases from 14 µm to 34.4 µm in the case of the NR/PE joint. There is a 270% increase in the effective diameter of the strained interphase of the Si-EPDM joint when compatibilizers are used.

3. From reptation theory and the mechanics of the joint, the peel strength has been shown to have a first power dependence on the interfacial thickness. The preliminary results on the HNBR-Nylon joint confirm this.

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