The Use of Compatible Blend of Styrene-Vinylacetate Copolymer/Natural Rubber Latex in Pressure-Sensitive Adhesive Applications by Using Irradiation and Chemical Initiation

M. M. Magida, Y. H. Gad, H. H. El-Nahas

Polymer Chemistry Department, National Center for Radiation Research and Technology, P.O. Box 29, Nasr City, Cairo, Egypt

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ABSTRACT: Thermoplastic elastomers from blends of styrene-vinylacetate copolymer latex (S-VAc) and natural rubber latex (NRL) as a hydrophilic/hydrophobic blend have been prepared by the solution-casting technique. Compatibility between S-VAc and NRL was confirmed at all ratios of the blend mixing through the measurements of water absorption, thermogravimetric analysis, and scanning electron microscope . The prepared blend was studied for pressure-sensitive adhesive (PSA) application. Nonylphenol ethoxylate (NPE) was used as a chemical tackifier to initiate a sticky property for S-VAc. Also, electron beam irradiation was applied as a physical tackifying

INTRODUCTION

Pressure-sensitive adhesive (PSA) agents are used in three forms: as modified aqueous dispersions or solutions in different solvents and as hot-melts adhesives. They exhibit permanent adhesive power and they are able, by the use of low pressure, to produce an adhesive joint between two bonded surfaces. While the solvent-type are used for the production of PSA tapes, the aqueous acrylic dispersions are applied to the manufacture of labels. The product originally used for making PSA tapes was natural rubber.¹ While the used rubber or elastomeric copolymer has decisive influence on elastic properties of PSA, the low molecular tackifier resins or tacky admixtures significantly influence the viscoelastic properties of PSA and its ability to wet the surface of adherent.² Moreover, certain rubbery thermoplastic blends possess a combination of physical and rheological properties that are unavailable in a single polymer.³ PSA are made by irradiating an uncured mixture of polymerizable compounds either by ultraviolet light or by electron beam (EB) to form a polymeric film of an adhesive. In the adhesive cured by EB, high peel and high tack combined with high

agent to promote stickiness for NRL at 12.5 Mrad or at 2.5 Mrad by addition of NPE tackifier. Peel strength, tensile strength, elongation at break, surface hardness, and aging time were investigated. Two types of double-sided PSA were prepared: the first to be applied at room temperature; the second to be applied thermally at 80°C. The obtained PSA adhesive was suggested to be used for adhering the synthetic veneers to the particle wood panels. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 157–165, 2009

Key words: blends; irradiation; pressure-sensitive adhesive

shear strength are especially noteworthy as such properties are difficult to achieve in conventional PSAs.⁴ In earlier references radiation cure⁵ describes EB irradiation at 1-24 Mrad of polymers such as polyacrylates, polyethers, and rubber-tackifier blends to obtain PSA tapes with improved high-temperature shear adhesion. Styrene-grafted natural rubber (styrene-GNR) latex was synthesized via emulsion copolymerization by using cumene hydroxide/tetraethylene pentamine as initiator. It was then mixed with natural rubber latex (NRL) with various latex compounding contents and then prevulcanization was carried out. The mechanical properties and heat, weathering, and ozone resistance of NRL modified with styrene-GNR latex were investigated as a function of the grafted natural rubber content. At high content of styrene-GNR the tensile and tear strength were decreased, whereas Young's modulus and hardness were increased.⁶ PSAs were prepared from Styrene Butadiene Rubber/Standard Malaysia Rubber grade L (SBR/SMRL) blends with a use of coumarone-indene resin and toluene as the tackifier and solvent, respectively. The blend ratio of SBR/SMRL varied from 0 to 100%, whereas the resin content increased from 40 to 120 phr in the adhesive formation. The loop tack and peel strength of paper/polyethylene terephthalate film were measured by using a Lloyd adhesion tester operating at 30 cm/min loading. Loop tack of adhesive indicates a maximum

Correspondence to: Y. H. Gad (yasser2uk@yahoo.com).

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value at 20% SBR. An observation is attributed to the optimum wettability of adhesive on the substrate where formation of mechanical interlocking occurs.⁷ A blend of polymers is applied in a film thickness that is used as PSA and then irradiated by EB. The adhesive is formed directly on the substrate to be used later as desired. For many of the applications, natural rubber-based adhesives are still used because of their good all-around balance of tack and cohesive combined with low cost. The aim of this study is to prepare double-sided polymeric films with PSA properties for use in synthetic veneer–particle wood adhesion applications.

MATERIALS AND METHODS

Materials

Styrene-vinyl acetate copolymer emulsion latex has a total solid content of 50%. The percent composition of styrene in the styrene-vinylacetate copolymer latex (S-VAc) as a solid content is 60% and vinyl acetate is 40%, where the basic formula of S-VAc latex (liquid phase) is 30% styrene, 20% vinyl acetate, 1% hydroxyl ethyl cellulose (thickener) in 48% water, and 1% emulsifying agent. Glass transition temperature (T_{o}) of S-VAc is $+8^{\circ}$ C; molecular weight of S-VAc is 85,000 g/mol, and specific gravity is 1.1 g/cm³. pH is 5-7, and viscosity at 20°C (Brookfield HAT, spindle 4, 20 rpm) is 2700 cP. Emulsifying agent by anionic with average particle size ca. 0.1 µm was purchased from Grandy (Cairo, Egypt). NRL with a trade name TOPTEX contains 60% solid content (Kuala Lumpur, Malaysia). Nonyl phenol ethoxylate (tackifier) was acquired from ICI (London, England).



Preparation of S-VAc/NRL blend

Blending of S-VAc and NRL was carried out by mixing different ratios from S-VAc and NRL (as a latex form) as follows: 25% S-VAc/75% NRL, 50% S-VAc/ 50% NRL, and 75% S-VAc/25% NRL. Stirring was applied by using a mechanical stirrer for 5 min at 120 rpm at room temperature. The degassed solution of each ratio of the blend was cast at room temperature and allowed to dry; after 2 days, drying was completed under vacuum at 20°C.

Tensile strength and elongation at break

The measurements of tensile strength (T_b) and percentage of elongation at break (E_b) were carried out by using an Instron Model 1195 (UK) at a crosshead speed of 50 mm/min.

The preparation of samples for peel test

- 1. The sample of double-sided PSA films were prepared by a casting process of formulated NRL and S-VAc and their blends on paper were siliconized and left to dry for 2 days at room temperature. Then drying was completed under vacuum at 20°C.
- 2. The dried cast was also covered by siliconized paper and then cut into 2.5 cm \times 14 cm strips with 0.2 to 1 mm in film thickness.
- 3. The irradiation of film samples was applied directly on the covered film strips.
- 4. The peel test was applied by removing the siliconized paper from the film strip and putting it beneath a particle wood piece and synthetic veneer; pressure was then applied for 20 kg/cm² for 30 s at room temperature or at 80°C for 10 s according to the type of PSA film strip was used. The peel strength was measured according to the following equation:

$$P = wt_a \sigma$$

where *P* is the peel strength, *w* is the width of adhesive film sample, t_a is the adhesive film thickness, and σ is the tensile strength. The test results were reported in Newtons per meter (N/m).

Peel test

Peel stripping strength of adhesive bond requires that one of the adherents be flexible enough so that it can essentially hold back on itself. Peel strength was measured at 23°C by 180° peel test by using a tensile tester with a cross-head speed of 25.4 cm/ min as described in ASTM D 903-49 180° peel test. The average force after the initial peak load was taken as the peel strength.

Hardness test

Surface hardness was measured by using ASTM D 2240 specifications, model 306L type A, D durometer for soft and hard plastic.

Water absorption percentage

Water absorption measurements were made by using clean and dried samples of known weight, which were immersed in distilled water for 24 h at 25°C. The samples were removed, blotted with absorption paper, and quickly weighted. The water absorption (%) of film was calculated as:

Water absorption (%) = $(W_e/W_d) \times 100$

where W_e is the weight of film at equilibrium and W_d is the weight of dried film before swelling.

Thermogravimetric analysis

The TGA studies were carried out on a Shimadzu-30 (TGA-30) at a heating rate of 10°C/min in air over a temperature range from room temperature up to 500°C. The primary TGA thermograms were used to determine the different kinetic parameters such as activation energy and order of thermal decomposition reaction.

Scanning electron microscope

The morphology of the prepared films was examined to visualize the microstructure of the blend and evaluate their structure morphology. Sample filaments were fractured in liquid nitrogen and then were coated with gold for 15 min by using a vacuum sputter coater prior to observation and scanned by using a SEM technique (Jeol, Japan), taking the micrographs with a JSA-5400 instrument.

Electron irradiation

The equipment used for electron irradiation was an electron accelerator (USA) operating at 1.5 MeV. The samples were put on a mobile plate in the electron accelerator and the selected dose was obtained by varying the number of passages through the scanned electron beam. Every passage resulted in a dose of 2.5 Mrad at speed of 2 m/min.

RESULTS AND DISCUSSION

Research has revealed that the basic criteria that play an important role in determining the miscibility of polymers may be summarized as follows:

- 1. The difference between solubility parameters of polymers.^{8–10}
- 2. The behavior of mixtures of solutions of the respective polymers.^{11–14}
- 3. The difference between polarities of polymers.¹⁵
- 4. The single compound behavior of the polymerpolymer blends.¹⁶
- 5. The regular distribution of the functional groups along the chain and the degree of crystallinity.¹⁷

This system of the blend of NRL and S-VAc was selected as PSA laminated films to adhere synthetic veneer to particle wood panels for two reasons: NRL as a dry film is considered a radiation-sensitive polymer as it degrades to an oxidized and sticky property on the surface of the film without affecting the internal film, keeping in turn some mechanical properties of the irradiated NRL film and also in the blend prepared to act as self-substrated PSA film. This system could be put between two pieces of siliconized paper (released paper) for use as a doublesided PSA film. The second reason is selecting S-VAc, where it contains styrene, which is considered more sensitive, to give a sticky property by adding NPE only without applying irradiation.

Water absorption

Figure 1 shows the water absorption of unirradiated and irradiated dry films of S-VAc and NRL blanks and blends. All films obtained from emulsion polymers usually show more or less pronounced water absorption when dipped in water or exposed to humid environments. The amount of water absorbed depends on the hydrophilic nature of the polymer, irradiation process, and also to some extent, on the type and amount of the emulsifying agent. SVAc/ NRL blend represents a combination of two different phases of polymer behavior, hydrophilic and hydrophobic, respectively. A partial hydrophilic behavior can seen for all the ratios of blending due to the addition of S-VAc and decreased slightly by using the irradiation process as a result of crosslinking,¹⁸ as shown in Figure 1.

Thermal decomposition behavior (TGA)

Figures 2–5 show the TGA thermograms of unirradiated and polymer blends containing different ratios



Figure 1 Effect of dipping time on water absorption of unirradiated and irradiated S-VAc/NRL blend films at different ratios.

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120

100

80

60

40

20

Û

D

100

Weight remaining %

Figure 2 TGA thermograms of unirradiated S-VAc/NRL polymer blend.

300

Tennerature(⁰C)

400

500

600

1120

1100

80

60

40

20

0

0

200

of S-VAc and NRL before and after they had been exposed to electron beam irradiation at different doses (25, 75, and 125 kGy). Table I summarizes the percentage weight loss at different decomposition temperatures for the unirradiated and irradiated S-VAc/NRL polymer blends taken from TGA thermograms. It can be seen that pure S-VAc and NRL polymers lose about 10% of their weight at 310 and 250°C, respectively. This behavior indicates that the initial decomposition reaction for S-VAc begins at a slightly higher temperature than for NRL. Thus, it may be concluded that the unirradiated S-VAc polymer is more stable against thermal decomposition than the unirradiated NRL over the entire range of studied temperature. On the other hand, the thermal stability of unirradiated S-VAc/NRL blends at any composition is higher than those of pure components, especially polymer blends rich in S-VAc content on the basis of percent weight loss. Thermal



100 %NRL 25% S-VAc 100 50 % S-V/Ac 75% S-VAc 100 % S-\/Ac Weight remaining(%) 80 60 40 20

Figure 4 TGA thermograms of unirradiated S-VAc/NRL polymer blend irradiated to 7.5 Mrad.

stability of any polymer material is largely determined by the strength of the covalent bond between the atoms forming the polymer molecules. The calculated dissociation energy for the different covalent bonds C-H, C-C, C-O, C=O, and C=C was reported to be 98, 81, 87, 174, and 145 kcal/mol, respectively.¹⁹ Based on these values, it can be calculated that the average complete dissociation energy of S-VAc is calculated to be about 102.1 and 97.6 kcal/mol for NRL. The lower complete dissociation energy of NRL compared to S-VAc is certainly due to the lack of the C=C bonds of (cis-1,4-polyisoprene) in the NRL molecule. In the case of irradiated polymer blends, it can be seen from Table I that the (75% S-VAc/25% NRL) polymer blend is thermally more stable than (50% S-VAc/50% NRL) and (25% S-VAc/75% NRL) up to 290°C at irradiation doses 2.5, 7.5, and 12.5 Mrad except at the temperature range from 310 to 390°C, in which the ratio (50%

> 100% NRL 75%NRL

50%NRL

25%NRL

100%S-VAc



300

Temperature (⁰C)

400

500

600

200

100







Polymer blend composition	Irradiation dose (Mrad)	Weight loss (%)					
		210°C	250°C	290°C	330°C	370°C	410°C
100% NRL	0	3.31	10.49	28.55	56.92	75.16	82.05
	2.5	2.37	4.09	10.89	22.45	30.62	37.42
	12.5	1.98	2.77	3.96	7.12	38.34	86.96
25% S-VAc	0	0.63	2.49	7.46	26.71	82.72	89.55
75% NRL	2.5	2.24	3.74	7.47	22.39	70.15	85.83
	12.5	2.39	2.39	41.67	89.29	93.66	94.85
50% S-VAc	0	0.82	2.46	5.74	26.77	65.58	75.41
50% NRL	2.5	1.87	2.8	3.73	11.63	48.38	79.46
	12.5	1.37	2.74	6.17	23.98	67.61	84.25
						63.7	
75% S-VAc	0	1.51	2.52	4.53	20.61	59.8	71.86
25% NRL	2.5	1.43	2.86	5.00	26.43	64.29	76.43
	12.5	1.2	3	5.39	26.35	67.07	78.45
100% S-VAc	0	2.06	3.43	5.48	35.62	69.87	73.29
	2.5	3.63	4.67	7.26	39.9	68.92	72.03
	12.5	2.76	3.45	5.18	18.97	66.56	72.07

 TABLE I

 Weight Loss (%) at Different Decomposition Temperatures for Different Polymer Blends of Various

 Ratios of S-VAc and NRL Unirradiated and Irradiated at Different Doses of Electron Beam Irradiation

SVAc/50% NRL) showed higher thermal stability than the ratios (75% S-VAC/25% NRL) and (25% S-VAc/75% NRL). Finally, we can conclude that there



Figure 6 Scanning electron micrographs of (a) unirradiated S-VAc, (b) irradiated S-AVc, (c) unirradiated NRL, (d) irradiated NRL, (e) blend of unirradiated 75% S-VAc/25% NRL, (f) blend of irradiated 75% S-AVc/25% NRL.

is a marked increase in the thermal stability of irradiated polymer blends especially for the blends rich in S-VAc. This increase is attributed to the presence of an aromatic ring in S-VAc, which offers a considerable degree of radiation protection of macromolecules. On the other hand, the thermal stability of unirradiated S-VAc/NRL blends at any composition is higher than those of pure components, especially polymer blends rich in S-VAc content on the basis of percent weight loss. This is because rubber thermoplastic blends possess a combination of physical and rheological properties that are unavailable in a single polymer.^{3–5} Moreover, NRL is highly unsaturated and the double bonds remaining in the monomeric unit (polyisoprene) after polymerization are susceptible to degradation by heating. However, the blend contains S-VAc, which has an aromatic ring that possesses a higher thermal stability.

Scanning electron microscopy

From figure (6), it can be seen that micrograph (a) represents the fracture surface of S-VAc blank, where a homogenous phase appeared as white and dark areas. Micrograph (b) shows the irradiated S-VAc films, where a continuous phase was noticed and the molecular chains of S-VAc as styrene and vinylacetate became more adherent to each other due to crosslinking, with the disappearance of a white area due to phase contraction compared to the unirradiated sample. Micrograph (c) shows NRL film as a blank sample appearing as a corrugated surface and the presence of textured pores.

Figure 7 Effect of irradiation dose on peel strength of synthetic veneer from particle wood by using NRL and S-VAc as laminated adhesive films.

However, the shape of the NRL domains appears less regular as a spongy surface shape. After the irradiation process, NRL produced a smooth and soft surface with slight sticky properties, increasing in parallel with the increase in the irradiation dose, as shown in micrograph (d). In the blended solution, the high mobility in solution prior to evaporation of the water allows the blend to separate and causes a macroheterogeneous morphology consisting of rather large zones of dispersed polymer.14-16 Micrograph (e) shows a blend of 75% S-VAc/25% NRL with a complete embedding of NRL particles into the S-VAc matrix with a compatible phase appearing in water absorption, and thermal and mechanical properties. Moreover in micrograph (f) a large plate appeared after irradiation of the S-VAc/NRL blend, giving a pronounced crosslinking in S-VAc with

1400 NRL unirradiated 1200 Peel strength (N/m) NRL at 2.5 Mrad NRL at 12.5 Mrad 1000 NRL at 30 Mrad S-VAc 800 600 400 200 0 10 20 30 40 NPE concentration (%) Figure 8 Effect of NPE concentration percent on peel

strength of synthetic veneer from particle wood by using NRL at different irradiation doses and unirradiated S-VAc as laminated adhesive films.

Figure 9 Effect of S-VAc concentration (%) on tensile strength of unirradiated and irradiated blend of laminated adhesive films.

softening and smoothing in NRL, which gave more adherence and compatibility.

Factors affecting PSA properties of NRL and S-VAc (unblended)

NRL-peel strength

Figure 7 shows the effect of irradiation dose on the peel strength of NRL and S-VAc separately without blending, where the dry film of NRL is given at 30 Mrad maximum peel strength (539), but by adding 20% nonyl phenol ethoxylates at an irradiation dose of 2.5 Mrad, a maximum peel strength (1470) was obtained, as shown in Figure 8. On the contrary, no peel was obtained by adding NRL without applying the irradiation process. At a ratio of 10% NPE added to the NRL with an irradiation dose of 12.5 Mrad, the peel strength was 1372, but at maximum



S-VAc concentration (%)







1600

irradiation dose of 30 Mrad, the maximum peel strength (539) was obtained at 0% NPE. The peel strength of NRL as a tackifier material and S-VAc through the mechanism of decreasing the glass transition of the polymer almost equals the room temperature degree, giving in this case a soft polymer with tacky properties. From Figure 8 it was concluded that no effect of NPE on peel strength of NRL was noticed; this may be because NRL is considered a highly unsaturated polymer (polyisoprene). After the irradiation process, the double bonds that remain in the monomeric unit of polyisoprene are susceptible to breaking down, giving a saturated polymer; this change leads to a successful reaction between NRL and NPE, which leads in sequence to a softer NRL polymer than the irradiated NRL, without adding NPE (as shown in Fig. 7). 5

S-VAc peel strength

No peel strength was obtained when the dry film of S-VAc was irradiated at different doses as shown in Figure 7. Figure 8 shows the effect of NPE added at different ratios on peel strength of S-VAc where the maximum peel strength was recorded 1470 at 25% of NPE. From Figures 7 and 8, it was concluded that a maximum peel strength of NRL was obtained at 20% NPE and 2.5 Mrad; also a maximum peel strength of S-VAc was obtained at 25% NPE and no effect of applying the irradiation process.

Mechanical properties of S-VAc/NRL blend

Figures 9–11 represent the mechanical properties of S-VAc/NRL blend samples unirradiated and irradiated at the doses of 2.5 and 12.5 Mrad, respectively. It can be observed that the tensile strength increases with the increase of S-VAc added and also with the



Figure 11 Effect of S-VAc concentration (%) on hardness of laminated adhesive unirradiated and irradiated films.



Figure 12 Effect of nonyl phenol (%) on peel strength of synthetic veneer from particle wood panel at blend of laminated adhesive film of 25% NRL-75% S-VAc ratio.

increase of irradiation dose; this was due to the crosslinking behavior of S-VAc toward the irradiation process, as shown in Figure 9. On the contrary, elongation at break exhibited a weak ratio with increasing S-VAc added and also decreases with the increase of irradiation dose, as shown in Figure 10. Hardness was increased with the increase of S-VAc added but not increased with the increase of irradiation dose, as shown in Figure 11. Therefore, it can be concluded that a low dose of electron beam irradiation (at 2.5 Mrad) enhances the mechanical properties at the blend ratio of 75% S-VAc/25% NRL.

Factors affecting the adhesion promotion of the blend

Radiation process and addition of NPE as tackiness were considered factors for promotion such pressure-sensitive adhesive properties for the prepared blend at the ratio of 75% S-VAc/25% NRL.

Effect of radiation on natural rubber

The presence of 1,4 units of polyisoprene in natural rubber contributed to the occurrence of chain scissions. Oxygen has been found to increase the rate of scission and to decrease the degree of crosslinking.^{20,21} Also, there is some evidence that the natural rubber molecule does not consist exclusively of carbon and hydrogen atoms but also contains minor amounts of oxygen, probably in the form of carbonyl groups.^{22,23} One effect of the presence of such groups is that crosslinking gradually occurs, due to interaction with active methylene groups on adjacent polymers chains. This crosslinking contributes significantly to the irreversible hardening,²³ which occurs in natural rubber during prolonged storage. A

Figure 13 Effect of temperature on peel strength of synthetic veneer from particle wood panel at different concentrations of NPE in the blend of laminated adhesive.

further cause of irreversible hardening arises from the presence of certain peroxides in natural rubber latex, which form redox systems with some of the other constituents. The consequence can be both crosslinking and chain scission. NRL is also highly unsaturated and the double bonds that remain in the monomeric unit of polyisoprene are extremely susceptible to attack by oxygen ozone and electron irradiation, resulting in general degradation by free radical mechanisms and softening of the surface layers of NRL, becoming slightly sticky and increasing in sequence the adhesion properties²⁴ of NRL, as shown in Figure 12.

Effect of tackifier

NPE as a tackifier reacts directly with styrene in the copolymer (SVAc) with a side reacted chain of the vinyl acetate promoting tacky properties (PSA properties). Also, the additions of NPE increase the PSA properties of NRL²⁵ at low doses, as shown in Figure 12.

Effect of temperature

The advantage of applying the heat process at different degrees of temperature is mainly to promote the adhesion of PSA properties at a low ratio of NPE (tackifier) added and also at low doses of irradiation. This type of process is classified as hot-melt pressure-sensitive adhesive, as shown clearly in Figure 13.

CONCLUSION

The following conclusions may be made from the study:

- 1. A blend of S-VAc and NRL was prepared by casting technique at ratios of 25, 50, and 75%.
- Compatibility of the blend has occurred at each ratio of mixing of S-VAc and NRL and was confirmed by the following investigations:
 - a. Water absorption of the blend showed that the hydrophilic properties of SVAc and hydrophobic properties of NRL possessed a partially hydrophilic blend at all ratios of mixing (25, 50, and 75%), indicating that a full miscibility and homogeneity of the prepared blend have occurred.
 - b. TGA asserted that the complete interaction was observed and increased with increasing S-VAc in the prepared blend at 75%.
 - c. SEM showed an intermediate morphological domain between S-VAc and NRL.
- 3. The prepared blend at ratio of 75% S-VAc/ 25% NRL was chosen to apply it as a film with double PSA properties by using two different methods as follows:
 - a. Chemical adhesion initiation method, by adding nonyl phenol ethoxylate (NPE) as a tackifier to initiate the PSA property of S-VAc.
 - b. Physical adhesion initiation method, by using the irradiation (EB) to induce a degradation of NRL at high doses (30 Mrad), resulting in an oxidation and breakdown of the double bond in the basic units of NRL (isoprene), leading to stickier PSA properties. Also, by adding a small ratio of NPE to NRL, the same strength of PSA properties was obtained at low doses.
- 4. The study indicated that the prepared of double PSA properties can be used to adhere veneer to the particle wood panels.

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