Repulpable EVA hot-melt packaging adhesives using polybutenes

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ABSTRACT: In a basic hot-melt formulation consisting of an EVA base resin, wax, tackifying resin, and antioxidant, various molecular-weight grades of polybutene were used to partially replace the tackifying resin. This resulted in lower blend densities and improved hot-melt removal efficiencies in laboratory tests. Polybutene also improved low-temperature flexibility without increasing open time.

KEYWORDS: Adhesives, antioxidants, EVA polymers, formulations, hot melts, packaging materials, paraffin wax, physical properties, polybutylene, polyisobutylene, removal, repulping, synthetic polymers, tackifiers.

EVA-based (ethylene vinyl acetate) hot-melt adhesives are currently used in a variety of packaging applications, with case and carton sealing being the largest of these *(1)*. As more corrugated board and other packaging materials are recycled, we need to formulate adhesives for these applications that are dispersed in the repulping slurry or that can be easily removed physically. In the repulping operation, a paper fiber–water slurry is formed when water is added to recovered paper, and the mixture is mechanically agitated in a Hydropulper. This slurry is then passed through various screens and centrifugal cleaners to remove nondispersed material. Adhesive material that cannot be dispersed or physically removed can potentially form sticky deposits on equipment or result in recycled products of a poorer quality *(2)*.

Separation of adhesive from the paper fiber is reported to be 90% efficient for formulations with specific gravities less than 0.98 *(3)*. Polybutenes, which are available in a wide variety of molecular weight grades, have specific-gravity values that range from 0.83 to 0.90. When polybutene is used in EVA hot-melt formulations to partially replace solid tackifying resins having specific gravity values of about 1.05 to 1.10, the result is a decrease in the overall blend density and an improvement in the hot-melt removal efficiency.

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Experimental

A typical EVA hot-melt adhesive formulation consists of an EVA base resin, paraffin wax, and a tackifying resin. For the control formulations in this study, these three components were blended in equal amounts. A small amount of antioxidant was also added to retard degradation of the adhesive during preparation and application. Three polybutene grades of different viscosity and molecular weight were added at substitution levels of 20% and 50% by weight for the tackifying resins. These formulations are presented in **Table I**.

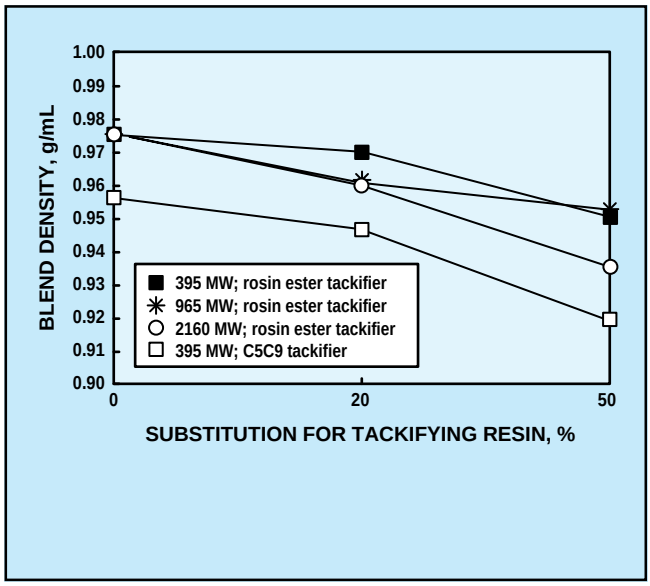
For those tests requiring the application of an adhesive film to a substrate, a two-roll hot-melt applicator was used. The two-roll applicator consisted of a heated top roller, maintained at 177°C, and a water cooled bottom roller. The desired film thickness was controlled by the placement of precision feeler gauges inserted between the two rollers. The substrate was placed between the two rollers, and molten adhesive was then poured onto the substrate on the back side of the rollers. The adhesive coated the substrate as it was pulled through the rollers at a continuous rate.

Results and discussion

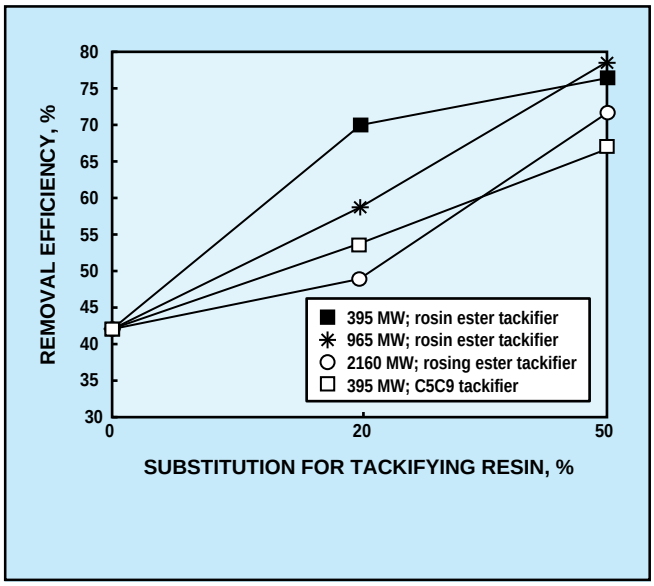
Physical properties of adhesive formulations

The physical properties of the test formulations are presented in **Table**

1. Effect of polybutene concentration on hot melt blend density



2. Effect of polybutene concentration on removal efficiency



I. Test formulations

Components	*Parts per hundred of base resin*									
Formulation no.	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*
EVA base resin, 28% vinyl acetate	100	100	100	100	100	100	100	100	100	100
Paraffin wax	100	100	100	100	100	100	100	100	100	100
Rosin ester tackifying resin	100	80	50	80	50	80	50	...	...	...
C5C9 tackifying resin	...	...	...	...	...	...	...	100	80	50
Antioxidant	3	3	3	3	3	3	3	3	3	3
Polybutene: MW-395 M(n)	...	20	50	...	...	...	...	...	20	50
Polybutene: MW-965 M(n)	...	...	...	20	50	...	...	...	...	...
Polybutene: MW-2160 M(n)	...	...	...	...	...	20	50	...	...	...

II. The addition of polybutene, while reducing the viscosity of the hot melt, appeared to have little effect on Ring and Ball softening point. Blend density was reduced as a function of increased polybutene concentration, as illustrated in **Fig. 1**. This plot also illustrates the fact that lower-molecular-weight grades, which also have lower specific gravity values, are most effective at reducing hot-melt blend density. Density is reduced when either rosin ester or C5C9 tackifiers are used in the formulation.

Hot-melt removal efficiency

A laboratory method was developed that measures the relative hot-melt removal efficiency during the repulping process. This method is intended to determine the relative removal efficiencies of similar adhesive formulations and may not give an accurate representation of the precise level of removal efficiency one can expect in the mill. In this method, a 0.05-mm coating of the adhesive was applied to white laser copier paper. The coated paper was then cut into 2.54-cm squares. About nine of the squares were weighed and then soaked in 250 mL of water for 1 hour. The weight of the coating was determined by weighing nine uncoated squares and calculating the weight difference. The aqueous solution was then agitated at medium speed in a household blender for 2 minutes. The

II. Physical properties of test formulations

Formulation no.		*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*
Polybutene mol. wt.		*...*	*395*	*395*	*965*	*965*	*2160*	*2160*	*...*	*395*	*395*
Percent sub. for tackifier		*0*	*20*	*50*	*20*	*50*	*20*	*50*	*0*	*20*	*50*
Property	*Units*										
Brookfield viscosity at 177°C	Centipoise	540	480	367	532	486	570	580	529	509	329
Ring and Ball softening point	°C	72	71	71	73	72	74	73	73	72	72
Blend density	g/mL	0.975	0.960	0.936	0.961	0.953	0.970	0.951	0.956	0.947	0.920

III. Hot melt removal efficiencies of test formulations

Formulation no.	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*
Polybutene mol. wt.	*...*	*395*	*395*	*965*	*965*	*2160*	*2160*	*...*	*395*	*395*
Percent sub. for tackifier	*0*	*20*	*50*	*20*	*50*	*20*	*50*	*0*	*20*	*50*
Removal efficiency, %	42	70	77	59	79	49	72	42	54	67

IV. Adhesive performance results

		Formulation no.	*1*	*2*	*3*	*4*	*5*	*6*	*7*
		Polybutene mol. wt.	*...*	*395*	*395*	*965*	*965*	*2160*	*2160*
		Percent sub. for tackifier	*0*	*20*	*50*	*20*	*50*	*20*	*50*
Test	*Method*	*Units*							
Mandrel bend flexibility at –12°C	ASTM D-3111	Mandrel diameter, cm	>1.27	0.32	0.32	0.64	0.32	1.27	1.27
Open time	ASTM D4497	Seconds	5	5	5	5	5	5	5
180-degree peel adhesion	See text	Fiber tear/no tear	FT	FT	FT	FT	FT	FT	FT
High-temperature stability at 177°C	ASTM D-4499	Hours	100	100	100	100	100	100	100
Shear adhesion fail temperature	ASTM D-4488	°C	66	62	60	60	61	62	61

resulting slurry was quantitatively transferred to centrifuge tubes and centrifuged at 2000 rpm for 15 minutes. Immediately after centrifuging the top solids layer was skimmed off into aluminum pans and dried in an air-purged oven at 60°C overnight. The pans were then cooled in a desiccator and weighed. The weight percent of recovered adhesive was then determined.

Hot melt removal efficiencies for the test formulations are presented in **Table III**, and graphically illustrated in **Fig. 2**. Only slightly more than 40% of the control formulations, which contained no polybutene, was recovered from the repulping process. The partial substitution of polybutene for the tackifying resins resulted in significant improvements in adhesive removal efficiencies. In the formulations with rosin ester tackifying resin, where several molecular-weight grades of polybutene were tested, low-molecular-weight polybutene grades resulted in the highest removal efficiencies—almost 80%.

Adhesive performance

The results of several key adhesive performance tests are presented in **Table IV**. These tests were chosen to measure the effect of polybutene addition on high- and low-temperature performance and open times of the hot melts. Compatibility of the adhesive components at elevated

temperature was also measured. Additional information on these methods and the results of these tests are discussed as follows.

Low-temperature flexibility. Mandrel bend flexibility at –12°C (ASTM D-3111) was used to measure the effect of polybutene addition on the low-temperature performance of the hot melts. Using this method, plaques were made of each hot-melt formulation that measures 75 mm x 10 mm and that has a thickness of 1.25 mm (± 0.04 mm). The plaques were cooled to –12°C and bent over mandrels of 1.27, 0.64, and 0.32 cm diameter. Plaques that had a fracture failure or crazing were considered a failure. The results presented in Table IV are the smallest mandrel diameters that resulted in a "pass" for each sample. In the control formulation, without polybutene, the sample fractured when tested with the largest-diameter mandrel (1.27 cm). With the addition of low-molecular-weight polybutene grades, the formulations passed the 0.32-cm-diameter mandrel test. Even with the highest-molecular-weight grade of polybutene tested, the samples passed the 1.27-cm-diameter mandrel test, offering improved low-temperature flexibility over that of the control formulation.

Open time. The addition of polybutene to the formulations did not result in extended open times within the limits of this ASTM method. This property is important in packaging applications where fast line speeds are critical.

180-degree peel adhesion. In this method a 0.0381-mm coating of the adhesive was applied to natural kraft paper and heat sealed at 150°C for 1 second with a jaw pressure of 138 kPas. All formulations resulted in substrate failure at ambient temperature. These data indicate that the addition of polybutene does not result in a weakening of the adhesive bonds to this substrate.

High-temperature stability. This test measures the number of hours a hot melt must endure at elevated temperatures without experiencing phase separation, skinning, or unusual discoloration. The test is conducted for a maximum of 100 hours. All test formulations passed the 100-hour maximum without failure.

Shear adhesion fail temperature. This test measures the high-temperature performance of an adhesive by determining the temperature at which overlap kraft paper–kraft paper bonds fail under a weight of 500 g. The paper was coated and heat sealed using the method described under "180-degree peel adhesion." Overlap bonds of the substrate strips measured 2.54 cm^2. The data indicate that there was a slight decrease in the high-temperature performance of those formulations containing polybutene. Other formulation variables which were not investigated, and which may improve high-temperature performance, include selecting different grades of wax or EVA base resin and varying the wax-to-EVA ratio.

Summary

EVA-based hot melt packaging adhesives have been formulated to offer improved removal efficiencies in the repulping process. Higher removal efficiencies using the density difference method were obtained by using polybutene as a partial replacement for solid tackifying resins in the formulations to reduce the overall blend density of the hot melt. Polybutene also improved the low-temperature flexibility of the hot melt without extending open times, decreasing bond strength, or causing compatibility problems. High-temperature performance of the adhesives was reduced slightly.

Literature cited

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