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SZ, UG).**Published***With international search report.***(54) Title:** TACKIFIED EMULSION PRESSURE-SENSITIVE ADHESIVE**(57) Abstract**

An inherently tacky, pressure-sensitive adhesive having good convertibility and improved low temperature performance at coat weights lower than conventional systems is provided. The pressure-sensitive adhesive comprises (a) an emulsion polymer formed from a monomer mix comprising 35 to 60 % by weight alkylacrylates, 15 to 35 % by weight vinyl esters, 15 to 35 % by weight diesters of dicarboxylic acids, and up to about 5 % by weight unsaturated carboxylic acids, and (b) a tackifier comprising a hydrocarbon resin component and a rosin-based resin component, with an acid number of 30 to 45 and a softening point of 55 to 65 °C. A preferred pressure-sensitive adhesive comprises 8 to 12 parts by weight tackifier per 100 parts by weight pressure-sensitive adhesive polymer, with the preferred tackifier comprising an aromatic modified aliphatic resin containing wood rosin.

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## TACKIFIED EMULSION PRESSURE-SENSITIVE ADHESIVE

### Background of the Invention

5 The present invention relates to emulsion pressure-sensitive adhesives which, when formulated with suitable tackifiers, provide good convertibility and improved low temperature performance at coat weights lower than the conventional systems used in general purpose label applications.

10 In general, pressure-sensitive adhesives are provided as solvent polymers, bulk polymers, and emulsion polymers. Some are pressure-sensitive adhesives in their own right, i.e., inherently tacky, while others require tackification to achieve that end. My earlier patents, U.S. Patent No. 5,164,444, and U.S. Patent No. 5,189,126 (both of which are incorporated herein by this reference), describe emulsion pressure-sensitive adhesive polymers which exhibit excellent performance at room and low temperature, and good  
15 convertibility.

It is known that acrylic-based pressure-sensitive adhesives can be tackified to improve adhesion to non-polar substrates such as polyolefins. However, most of the commercially available tackifiers have low molecular weights, as compared to the adhesive polymer, and their addition to the polymer lowers the shear properties and convertibility characteristics of  
20 the adhesive. Such tackified adhesives often exhibit poor adhesion to other substrates, such as recycled corrugated board. Additionally, tackification of pressure-sensitive adhesives typically improves room temperature performance, but not low temperature performance, and, in most cases, the performance of the tackified adhesive is less than that of the non-tackified pressure-sensitive adhesive. Performance characteristics would thus appear to  
25 depend in large part on the selection of tackifier(s), their functional groups, and concentrations.

The following patents are illustrative of various known types of tackified pressure-sensitive adhesives:

30 U.S. Patent No. 4,418,120 to Kealy, et al. describes a tackified, crosslinked, acrylic adhesive which shows improved adhesion to low energy substrates. The adhesive is based on a solution polymer of isooctyl acrylate and acrylic acid containing from 3-7% by weight of acrylic acid, 100 parts of which is blended with 20-50 parts of a tackifying rosin ester having an acid number less than 20. The adhesive also contains antioxidants and a crosslinking agent, and is coated and dried at 70°C to allow for  
35 crosslinking. The composition is required to be free of surfactants to achieve the designated performance.

U.S. Patent No. 4,726,982 to Trayner, et al. describes a tackified acrylic pressure-sensitive adhesive for application to high solids automotive paints. The adhesive is a solution

1 polymer of an acrylic acid ester of non-tertiary alcohol containing from 4-14 carbon atoms,  
which polymers contain from 10-40 parts of N-vinyl lactam, and is tackified with 5-50 parts,  
based on the weight of the polymer, of tackifying resins such as poly(isobornyl  
methacrylate), rosin ester, and mixed aliphatic/aromatic polymeric tackifier resins. A  
5 combination of the N-vinyl lactam and the tackifier is needed to provide high adhesion to  
automotive paint. Compositions with less than 10% N-vinyl lactam do not provide high  
adhesion. The tackifying resins are either esters or hydrocarbon resins with no carboxyl  
functionality.

U.S. Patent No. 4,988,742 to Moon, et al. discloses tackified acrylic pressure-sensitive  
10 adhesives with improved shear and adhesion to low energy substrates. The adhesives  
comprise from about 60-95 parts of a photo-polymerized polymer of monomers containing  
60-96 parts of an alkyl acrylate, the alkyl groups having an average of 6-12 carbon atoms,  
from about 2-15 parts of strongly polar monomers, and 2-25 parts of second strongly polar  
and/or moderately polar monomers; from about 5-40 parts of a hydrogenated rosin ester  
15 tackifying agent; and from about 0.01-1 parts of a photoinitiator; wherein such adhesive has  
a lower glass transition temperature than would an identical untackified acrylic pressure-  
sensitive adhesive. The tackifying resins disclosed are rosin esters with softening points of  
65-110°C with an acid number of less than 10.

U.S. Patent No. 5,284,891 to Wouters et al. describes the use of tackifiers in  
20 polyacrylate emulsion pressure-sensitive adhesives. Typical compositions have 35-85% by  
weight on a dry basis of the polyacrylate emulsion pressure-sensitive adhesive and from 15-  
70% by weight of a tackifying resin having a softening point of 10-120°C, the resin being  
a copolymer of (i) a feed which is predominantly C<sub>5</sub> olefins and diolefins, and (ii) 10-60  
weight percent of monovinyl aromatic compounds, e.g., styrene. The resins are purely  
25 hydrocarbon resins with no acid functionality.

U.S. Patent No. 5,242,963 to Mao discloses a tackified pressure-sensitive adhesive  
composition with improved adhesion to low density polyethylene, comprising an aqueous  
pressure-sensitive adhesive polymer emulsion and a tackifier. The copolymer emulsion  
consists essentially of 10-25 wt. % vinyl acetate, 10-25 wt. % ethylene, 50-80 wt. % acrylate  
30 monomer, and 5-40 wt. % dialkyl maleate or fumarate, and has a glass transition ranging  
from -70°C to 15°C. The tackifier resin is a rosin ester, a petroleum resin or a terpene  
resin, present in an amount of from 5-60 wt. %, and preferably 20-40 wt. %. The patent  
does not disclose the use of a resin mixture, nor the functional groups present in the resin.  
The resins used either do not have any carboxyl functionality or have an acid number of less  
35 than 20. To see the improvement in performance, the resin needs to be present in an amount  
of at least 30%, based on solids.

European Patent No. 0 303 430 to Martin describes tackified acrylic pressure-sensitive  
adhesives and adhesive tapes cured by ultraviolet radiation, with improved peel adhesion to

1 various substrates. The pressure-sensitive adhesives comprise from about 50-95 parts by  
weight of an ultraviolet radiation-polymerized polymer of one or more monomers which are  
predominantly alkyl acrylate, the alkyl group of which has an average of 4-12 carbon atoms,  
5 or about 0-15 parts by weight of one or more strongly polar copolymerizable monomers,  
or about 0-30 parts by weight of one or more moderately polar monomers; and about 5-50  
parts by weight of one or more tackifying resins, said tackifying resins comprising aliphatic  
polymeric resins derived from C-5 or (C-5)<sub>2</sub> monomer fractions which further contain from  
10 about 1 to about 80 weight percent aromatic components derived from C-9 aromatic  
monomer hydrocarbon fractions. The tackifying resins have a number average molecular  
weight of about 300-2500, a polydispersity index of less than about 5, a glass transition  
temperature of about 40-120°C and a solubility parameter of about 7-9.5, and said adhesive  
having a monomer conversion factor of at least about 95%. The resins used are aromatic  
modified aliphatic polymeric resins.

15 U.S. Patent Nos. 5,013,784; 5,106,902; 5,179,151; 5,095,065 and 5,164,441 describe  
internal resin tackified acrylic polymers containing crosslinkable monomers. The tackified  
acrylic copolymer compositions comprise a hydrogenated hydrocarbon resin, synthetic or  
natural, having a molecular weight of from about 500 to about 5000 and an aromatic content  
of at least 10% by weight; and an acrylic emulsion copolymer comprising from about 1-15  
20 wt. % of a (meth)acrylic acid, from 0 to about 50 wt. % of a lower alkyl (meth)acrylate  
ester and from about 25 to about 99 wt. % of an upper alkyl (meth)acrylate ester, having  
dissolved therein from about 10 to about 100 parts of the resin per 100 parts polymerizable  
monomer, said polymerizable monomer mixture including an effective amount of about 0.1  
to about 2% of a crosslinking monomer. The polymer can also be made free from  
crosslinking monomer and chain transfer agent.

25 In general, the presence of a tackifying resin improves adhesion to polyolefin  
substrates at the cost of reduced adhesion to recycled corrugated board. What is needed is  
a tackified pressure-sensitive adhesive that performs well on a wide variety of substrates, at  
both room and low temperatures.

### 30 Summary of the Invention

The present invention provides pressure-sensitive adhesive compositions that exhibit  
excellent peel and tack adhesion to a wide variety of substrates ranging from polar, relatively  
high energy surfaces such as glass and stainless steel, to nonpolar, relatively low energy  
surfaces such as polyolefins, and to difficult to bond surfaces such as corrugated board.  
35 Performance at room temperature and low temperature is good in spite of tackification. In  
adhesive constructions including facestock and liner, the adhesive provides excellent high  
speed converting characteristics such as die cutting, matrix stripping, and fan folding,  
equivalent to that of the nontackified systems. The adhesive systems provide good

1 performance in sheet fed laser printer applications, with minimal or no contamination, and  
and are free from edge ooze. Superior properties are obtained even at lower than normal coat  
weights. In sum, the adhesives are broad-based, and serve to replace many emulsion-based  
adhesives.

5 The pressure-sensitive adhesives of the present invention comprise an emulsion  
pressure-sensitive adhesive polymer and a tackifier having an acid number of from about 30  
to about 60 and a softening point of from about 50 to about 70°C, with the tackifier being  
present in an amount of between about 5 and 25 parts by weight per 100 parts of the  
polymer. Preferably, the tackifier is present in an amount of between about 8 and 15 — and  
10 more preferably, about 8 to 12 — parts by weight per 100 parts of the polymer.

The pressure-sensitive adhesive polymers used in the present invention are similar to  
the polymers described in my earlier patents, identified above, but with compositional  
modification and no crosslinker. The polymers have a glass transition temperature of less  
than about -30°C and a gel content of from about 50 to about 70% by weight of the polymer,  
15 and comprise, on a polymerized basis and based on the total weight of the polymer, about  
35 to about 60% by weight of at least one alkyl acrylate (more preferably, a mixture of alkyl  
acrylates) containing from about 4 to about 8 carbon atoms in the alkyl group; about 15 to  
about 35% by weight (more preferably, about 15 to 20% by weight) of at least one vinyl  
ester containing from about 2 to about 16 carbon atoms in the alkyl chain of the ester; about  
20 15 to about 35% of at least one diester of dicarboxylic acid, wherein each alkyl group of the  
diester independently contains from about 6 to about 12 carbon atoms; and up to about 5%  
by weight (more preferably, about 1 to about 3%) of an unsaturated carboxylic acid  
containing from about 3 to about 5 carbon atoms.

The emulsion polymers are prepared using conventional surfactants or, more  
25 preferably, by additionally employing a reactive surfactant which polymerizes and becomes  
part of the emulsion polymer, and which has been observed to enhance cohesive strength and  
aid in copolymerization of the monomers in forming the emulsion pressure-sensitive adhesive  
polymers. The reactive surfactant is employed in an amount up to about 0.5% by weight of  
the total monomers, preferably from about 0.1 to 0.25% by weight. The preferred reactive  
30 surfactants are anionic vinyl functional surfactants, such as sodium vinyl sulfonate and  
sodium styrene sulfonate.

The emulsion polymers are prepared with excellent conversions at reaction temperature  
ranging from about 70° to about 85°C, in the presence of from about 0.5 to about 1% by  
weight, based on the weight of the monomers, of a persulfate or equivalent catalyst, with the  
35 monomer mix being fed over a period of about 4 to 5 hours. Reaction pH is from about 2.5  
to about 4.0. Conversion is high, approaching 100% at the reaction conditions set forth  
above.

1           The tackifiers useful in this invention have an acid number of from about 30 to about  
60, preferably 30 to 45, and a ring and ball softening point of from about 50 to about 70°C.  
In a preferred embodiment, the tackifier comprises two parts, namely, a hydrocarbon resin  
5           component, preferably an aromatic modified aliphatic hydrocarbon resin, and a rosin-based  
resin component, with the two components present in a suitable ratio to provide a desired  
balance of properties and good adhesion both to polyolefins and recycled corrugated board.  
Typically, the tackifier is added as an aqueous dispersion. The hydrocarbon resin and rosin-  
based resin component can be mixed together and dispersed in a single process, or,  
10           alternatively, aqueous dispersions of the two components can be blended together in a  
suitable ratio to obtain the desired acid number and softening point.

          The pressure-sensitive adhesives of the present invention are prepared by making an  
emulsion polymer as described above, adjusting the pH to between about 6 and 7 (preferably  
by adding ammonia), and adding an aqueous dispersion of the tackifier to the emulsion  
polymer. When so formulated, the adhesive is ready to be coated on release paper, dried,  
15           and laminated to a desired backing.

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1 **Detailed Description**

5 The present invention provides tackified, emulsion pressure-sensitive adhesives which exhibit excellent convertibility and improved low-temperature performance at coat weights lower than conventional systems used in general purpose label applications. The adhesives exhibit excellent peel and tack adhesion to a wide variety of substrates, from polar, relatively high energy substrates such as glass and stainless steel, to non-polar, relatively low energy surfaces such as polyolefins, and to difficult to bond surfaces such a recycled corrugated board. The adhesives exhibit good room temperature performance and, in spite of the presence of a tackifier, good low temperature performance.

10 As compared to tackified styrene-butadiene adhesives, the adhesives of the present invention exhibit better aging and show no edge ooze or bleed as part of a release liner-adhesive-facestock laminate. In adhesive constructions including a facestock and release liner, the adhesives provide excellent high speed label converting characteristics, such as die cutting, matrix stripping and fan folding, equivalent to that of non-tackified systems. The adhesives provide good performance in sheet-fed laser printer applications, with minimal or no contamination, and are free from edge ooze. Surprisingly, superior properties are obtained even at coat weights of about 20 grams per square meter ( $\text{g}/\text{m}^2$ ), which is significantly lower than coat weights of 23-27  $\text{g}/\text{m}^2$  commonly used in many general purpose labeling applications.

20 The adhesives of the present invention contain a pressure-sensitive adhesive polymer essentially similar in composition to the polymers described in the above-referenced U.S. Patent Nos. 5,164,444 and 5,189,126, and a positive amount of a tackifier having an acid number of between about 30 and about 60 and a softening point of between about 50 and about 70°C. The polymers are prepared by emulsion polymerization, according to the method described in U.S. Patent No. 5,164,444 (col. 5, lines 27-68). The polymers contain, on a polymerized basis and based on the total weight of the polymer, the following monomers: (i) from about 35 to about 60 percent by weight of at least one alkyl acrylate containing from about 4 to about 8 carbon atoms in the alkyl group; (ii) from about 15 to about 35 percent by weight of at least one vinyl ester containing from 2 to about 16 carbon atoms in the alkyl chain of the ester; (iii) from about 15 to about 35 percent by weight of at least one diester of a dicarboxylic acid, wherein each alkyl group of the diester independently contains from about 6 to about 12 carbon atoms; and (iv) from 0 to about 5 percent by weight of an unsaturated carboxylic acid containing from about 3 to about 5 carbon atoms.

35 More specifically, the first monomeric component comprises at least one alkyl acrylate, and preferably a mixture of alkyl acrylates, present in a total amount of from about 35 to 60 percent by weight. Useful alkyl acrylates include n-butyl acrylate, 2-ethylhexyl acrylate, isooctyl acrylate and the like. The preferred first monomeric component is a mixture of 2-ethylhexyl acrylate and butyl acrylate.



1           The second monomeric component comprises vinyl esters present in a total amount of  
from about 15 to about 35 percent by weight, more preferably from about 15 to about 20  
percent by weight, with the vinyl esters containing from 2 to about 16 carbon atoms in the  
alkyl group of the ester. Suitable vinyl esters include vinyl acetate, vinyl butyrate, vinyl  
5           propionate, vinyl isobutyrate, vinyl valerate, vinyl versitate, and the like. Vinyl acetate is  
preferred.

          The third monomeric component of the emulsion polymer comprises one or more  
diesters of a dicarboxylic acid, wherein each alkyl group of the diester independently contains  
from about 6 to about 12 carbon atoms, the diesters being present in a total amount of from  
10          about 15 to about 35 percent by weight. The preferred diesters are dioctyl esters such as  
di-2-ethylhexyl maleate, di-2-ethylhexyl fumarate, and mixtures thereof.

          The fourth monomeric component of the polymer comprises up to about 5 percent by  
weight, preferably from about 1 to about 3 percent by weight, of at least one unsaturated  
carboxylic acid containing from 3 to about 5 carbon atoms. Such unsaturated carboxylic  
15          acids include acrylic acid, methacrylic acid, itaconic acid,  $\beta$ -carboxyethyl acrylate, and the  
like. Acrylic and methacrylic acid are the preferred unsaturated carboxylic acids, with a  
mixture of the two being more preferred.

          Preferably, the emulsion polymers used in the pressure-sensitive adhesives of the  
present invention are prepared in the presence of a reactive surfactant which polymerizes  
20          during formation of the polymer and becomes an integral part of the polymer. Preferred  
reactive surfactants include anionic vinyl functional monomers such as sodium vinyl  
sulfonate, sodium styrene sulfonate, and the like. The reactive surfactant is present as part  
of the total surfactant system in an amount up to about 0.5 percent by weight of the total  
monomers, preferably about 0.1 to about 0.25 percent by weight.

25          The presently preferred polymer contains, exclusive of reactive surfactant monomers  
and on a weight basis, about 48% 2-ethylhexyl acrylate, 27% di-2-ethylhexyl maleate, 19.5%  
vinyl acetate, 3.35% butyl acrylate, 1.2% acrylic acid, and 0.95% methacrylic acid. When  
tackified, this polymer yields a pressure-sensitive adhesive which exhibits excellent  
convertibility and good room and low-temperature performance. It is slightly less expensive  
30          to produce than the second presently preferred polymer, which contains, exclusive of reactive  
surfactant monomers, 51% 2-ethylhexyl acrylate, 26.85% di-2-ethylhexyl maleate, 20% vinyl  
acetate, 1.15% acrylic acid and 1% methylacrylic acid.

          The proportions of monomers are adjusted in such a way that the polymer has a glass  
transition temperature ( $T_g$ ) less than about  $-30^\circ\text{C}$ , giving a good balance of adhesion and tack  
35          at room and low temperatures.

          As explained in U.S. Patent No. 5,164,444 (col. 4, line 61 through col. 5, line 20),  
the above-described emulsion polymers have a gel content (% insolubles) in the range of 50  
to 70% by weight, which provides good cohesive strength without the use of multifunctional

1 monomers. In this regard, gel represents the amount of polymer which is insoluble in tetrahydrofuran, expressed in % by weight and determined by the membrane gel partitioning method.

5 In addition to an emulsion polymer, the pressure-sensitive adhesives of the present invention also include a tackifier. Although the emulsion polymers themselves are inherently tacky, by adding a tackifier having an acid number of from about 30 to about 60 and a ring and ball softening point of from about 50 to about 70°C, pressure-sensitive adhesives exhibiting good convertibility and improved room temperature and low temperature performance are obtained, even at coat weights lower than the conventional systems used in  
10 general purpose label applications, and for a wide range of substrates, including polar, non-polar, and difficult to bond to surfaces.

More particularly, it has been found that excellent performance characteristics are obtained by adding to the emulsion polymer an aqueous dispersion of a tackifier having an acid number of from about 30 to about 60, preferably from 30 to 45, and a softening point  
15 of from about 50 to 70°C. As used herein, the term "acid number" means the number of milligrams of potassium hydroxide required to neutralize the free acids present in one gram of tackifier.

The preferred tackifiers used in this invention contain both a hydrocarbon resin component and a rosin-based resin component. Preferably, the hydrocarbon resin is an  
20 aromatic modified aliphatic hydrocarbon resin, with a ring and ball softening point of from about 60 to about 80°C, and having no reactive functional groups. (Pendant phenyl groups and the like are not considered "reactive" in this context.) This range of softening points has been found to be optimal for the pressure-sensitive adhesive polymer compositions described above. At higher softening points, the adhesive properties can deteriorate, while at lower  
25 softening points, tackifier migration can occur.

Useful hydrocarbon resins include Tacolyn 1070, an aromatic-modified, aliphatic hydrocarbon resin sold by Hercules Incorporated ("Hercules"); XR-3013, an experimental resin from Arizona Chemicals; and similar resins.

The rosin-based resin component of the tackifier is a rosin or rosin derivative which,  
30 when blended with the hydrocarbon resin, yields the desired acid number and softening point. Such rosin-based resin components include wood rosins, gum rosins, tall oil rosins, fully or partially polymerized rosins, disproportionated rosins, fully or partially hydrogenated rosins, fully or partially esterified rosins, and other modified and/or stabilized rosins and the like. A preferred rosin is wood rosin, which is primarily abietic acid, with an acid number of from  
35 about 155 to about 210. Commercially available high-acid tackifiers include Snowtack 301A, 342A, 385G, 348A, and the like, sold by Eka Nobel.

A two component tackifier containing a hydrocarbon resin and a rosin-based resin component can be prepared by mixing a rosin-based resin dispersion with a hydrocarbon

1 resin dispersion, or vice versa, in a suitable ratio such that the resulting tackifier has an acid  
number of between about 30 and 60 (preferably, from 30 to about 45) and a softening point  
of between about 50 and 70°C in order to provide an adhesive which exhibits good adhesion  
both to substrates such as polyolefins and recycled corrugated board. Presently, the  
5 preferred tackifier is ResA 2467, an experimental product developed with Hercules and  
which is an aromatic modified aliphatic resin containing rosin acid, with an acid number of  
from 30 to 45 and a softening point of from 55 to 65°C. Hercules makes the tackifier in a  
single process.

10 The tackifier is added to the emulsion polymer in a positive amount of up to about 25  
parts by weight (dry tackifier) per 100 parts of the polymer (dry weight), with about 5 to 15  
parts tackifier per 100 parts polymer being preferred, and 8 to 12 parts per 100 parts  
polymer being more preferred. Presently, the most preferred adhesive formulation contains  
about 8 to 10 parts by weight of ResA 2467 per 100 parts of the emulsion polymer. A good  
15 balance of adhesive properties and, surprisingly, converting properties equivalent to or better  
than untackified adhesives, is seen with this and similar formulations. Although some prior  
systems reveal tackifier resin levels of between 5 and 50%, the working examples actually  
require greater than about 20% tackifier to see improved performance.

20 The pressure-sensitive adhesives of the present invention are preferably formulated by  
first preparing an emulsion polymer as described above, adjusting its pH to between about  
6 and 7 (preferably by addition of ammonia, with agitation), and adding an aqueous  
dispersion of the tackifier. Adhesive constructions are then prepared as follows: The  
formulated adhesive is coated on a release liner at about 20 to 21 g/m<sup>2</sup> coat weight, dried at  
70 to 90°C for about 10 to 15 minutes, and laminated to a paper (or other) backing,  
preferably, electronic data processing paper (EDP), data systems processing paper (DSX),  
25 high gloss, and/or dual print paper. Other facestocks such as polymeric film can also be  
used.

#### EXAMPLES 1-21C

30 To a one liter reactor equipped with a reflux condenser, a thermocouple, a pitched  
turbine agitator and a nitrogen inlet tube, there was charged a solution containing 78g of  
deionized water, 4g of sodium vinyl sulfonate (25% by weight solution in water) and 0.38g  
of Polystep B-27, an anionic surfactant manufactured and sold by Stepan Chemicals. A  
monomer mix consisting of 240g of 2-ethylhexyl acrylate (2-EHA), 145g of di-2-ethylhexyl  
maleate (DOM), 105g of vinyl acetate (VAc), 6.25g of acrylic acid (AA), and 5g of  
35 methacrylic acid (MAA) was added to 125g of water containing 44.5 g of Polystep B-27, and  
6.5g of Polystep F-9 (manufactured and sold by Stepan Chemicals) and agitated to make a  
pre-emulsion. The reactor charge was heated to 72 to 75°C and there was added 21g of  
4.45% w/w of potassium persulfate solution. 66g of the pre-emulsified monomer and 10.4g

1 of the potassium persulfate were added over a 20-30 minute period. After the temperature  
 reached a steady state, the remaining monomer pre-emulsion and a 1.85% aqueous solution  
 of potassium persulfate buffered with sodium bicarbonate were introduced into the reactor  
 at respective rates of 2.22 and 0.287g/minute, over a period of 270 minutes. The reaction  
 5 temperature was maintained between 79°C and 82°C. After the end of feed, the temperature  
 was raised to 83-85°C. Thirty minutes after the feed, 5.25g of a 4.8% solution of potassium  
 persulfate was added and the reactants maintained at a temperature of 87-90°C for 90  
 minutes. Once the polymerization was complete, the contents were cooled to ambient  
 temperature, neutralized with ammonia to pH 5-7 and discharged. The polymer content was  
 10 59.2% solids with less than 0.02% coagulum. Polymer  $T_g$  was -33°C. 10.15g of an  
 aqueous dispersion of Tacolyn 1070 resin was slowly added, with agitation, to 100g of the  
 above-described emulsion polymer, and the formulation was stirred for 20 to 30 minutes.

Using the above-described procedures, the tackified adhesives and untackified  
 comparative pressure-sensitive adhesive polymers listed in Table 1 (Examples 1-21C) were  
 15 prepared. Control 1 ("Ctl. 1") is an inherently tacky emulsion acrylic copolymer pressure-  
 sensitive adhesive prepared in accordance with U.S. Patent No. 5,164,444, and is  
 commercially available from Avery-Dennison. Control 2 ("Ctl. 2") is a different emulsion  
 acrylic copolymer pressure-sensitive adhesive available from Avery-Dennison, to which was  
 added Snow Tack 301A as a tackifier.

20

	Monomeric Composition (Wt.%)							Tackifier			
	2-EHA <sup>1</sup>	DM <sup>2</sup>	VAC <sup>3</sup>	BA <sup>4</sup>	AA <sup>5</sup>	MAA <sup>6</sup>	% Solids <sup>7</sup>	Tackifier	% Tackifier	Softening Pt. (°C)	Acid No.
Ex. 1	48	29	21	0	1.25	1	59.2	Tac. 1070 <sup>8</sup>	10	70	-
Ex. 2	52.5	27	18	0	1.5	1	60.2	Tac. 1070	10	70	-
Ex. 3	35	25	21	16.5	1.5	1	59.3	Tac. 1070	10	70	-
Ex. 4	48	23	18	8.7	1.5	0.8	59.4	-	-	-	-
Ex. 4A	48	23	18	8.7	1.5	0.8		ResA 2430 <sup>9</sup>	15	60	-
Ex. 4B	48	23	18	8.7	1.5	0.8		11558-48 <sup>10</sup>	12.5	63	42
Ex. 5	49.7	23	16	9	1.5	0.8	60	-	-	-	-

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TABLE 1

	Monomeric Composition (Wt.%)							Tackifier			
	2-EHA <sup>1</sup>	DM <sup>2</sup>	VAC <sup>3</sup>	BA <sup>4</sup>	AA <sup>5</sup>	MAA <sup>6</sup>	% Solids <sup>7</sup>	Tackifier	% Tackifier	Softening Pt. (°C)	Acid No.
Ex. 5A	49.7	23	16	9	1.5	0.8		ResA 2430	12.5	60	-
Ex. 5B	49.7	23	16	9	1.5	0.8		11558-48	12.5	63	42
Ex. 6	49	23	17	8.85	1.25	0.9	59.6	-	-	-	-
Ex. 6A	49	23	17	8.85	1.25	0.9		11558-48	12.5	63	42
Ex. 7	50	24	17	6.8	1.3	0.9	59.9	-	-	-	-
Ex. 7A	50	24	17	6.8	1.3	0.9		11558-48	15	63	42
Ex. 7B	50	24	17	6.8	1.3	0.9		11558-76 <sup>11</sup>	13	60	-
Ex. 8	48	25.05	18	6.85	1.1	1	59.1	-	-	-	-
Ex. 8A	48	25.05	18	6.85	1.1	1		Tac. 1070	12.5	70	-
Ex. 8B	48	25.05	18	6.85	1.1	1		11558-84 <sup>12</sup>	8	63	42
Ex. 9	48	25	18	6.85	1.2	0.95	59.35	-	-	-	-
Ex. 9A	48	25	18	6.85	1.2	0.95		11558-96 <sup>13</sup>	8	61	39
Ex. 9B	48	25	18	6.85	1.2	0.95		11558-84	8	63	42
Ex. 9C	48	25	18	6.85	1.2	0.95		11568-10 <sup>14</sup>	8	NA	42
Ex. 10	50	26.45	19	2.4	1.2	0.95	59.8	-	-	-	-
Ex. 10A	50	26.45	19	2.4	1.2	0.95		11558-76	12.5	60	-
Ex. 10B	50	26.45	19	2.4	1.2	0.95		ResA 2430	10	60	-
Ex. 11	50	26.45	19	2.4	1.25	0.9	59.8	-	-	-	-
Ex. 11A	50	26.45	19	2.4	1.25	0.9		11558-76	12.5	60	-
Ex. 11B	50	26.45	19	2.4	1.25	0.9		11558-84	8	63	42
Ex. 11C	50	26.45	19	2.4	1.25	0.9		ResA 2430	8	60	-

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TABLE 1

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	Monomeric Composition (Wt.%)							Tackifier			
	Z-ENA <sup>1</sup>	DOM <sup>2</sup>	VAC <sup>3</sup>	BA <sup>4</sup>	AA <sup>5</sup>	MAA <sup>6</sup>	% Solids <sup>7</sup>	Tackifier	% Tackifier	Softening Pt. (°C)	Acid No.
Ex. 12	51	26.9	20	-	1.3	0.8	59.6	-	-	-	-
Ex. 12A	51	26.9	20	-	1.3	0.8		11558-76	13.5	60	-
Ex. 12B	51	26.9	20	-	1.3	0.8		ResA 2430	10	60	-
Ex. 12C	51	26.9	20	-	1.3	0.8		11558-84	8	63	42
Ex. 12D	51	26.9	20	-	1.36	0.8		11558-96	8	61	39
Ex. 13	51	26.85	20	-	1.15	1	59.58	-	-	-	-
Ex. 13A	51	26.85	20	-	1.15	1		ResA 2430	10	60	-
Ex. 13B	51	26.85	20	-	1.15	1		11558-84	10	63	42
Ex. 13C	51	26.85	20	-	1.15	1		11558-96	8	61	39
Ex. 14	51	26.9	20	-	1.2	0.9	59.39	-	-	-	-
Ex. 14A	51	26.9	20	-	1.2	0.9		Tac. 1070	12.5	70	-
Ex. 14B	51	26.9	20	-	1.2	0.9		ResA 2430	12.5	60	-
Ex. 14C	51	26.9	20	-	1.2	0.9		11558-84	12.5	63	42
Ex. 15	48	25	18	6.85	1.25	0.9	59.5	-	-	-	-
Ex. 15A	48	25	18	6.85	1.25	0.9		11558-84	12.5	63	42
Ex. 15B	48	25	18	6.85	1.25	0.9		11558-84	10	63	42
Ex. 16	48	25	18	6.85	1.25	0.9	58.63	-	-	-	-
Ex. 16A	48	25	18	6.85	1.25	0.9		ResA 2430	8	60	-

TABLE 1

	Monomeric Composition (Wt.%)							Tackifier			
	2-EHA <sup>1</sup>	DM <sup>2</sup>	VAC <sup>3</sup>	BA <sup>4</sup>	AA <sup>5</sup>	MAA <sup>6</sup>	% Solids <sup>7</sup>	Tackifier	% Tackifier	Softening Pt. (°C)	Acid No.
Ex. 16B	48	25	18	6.85	1.25	0.9		11558-84	8	63	42
Ex. 16C	48	25	18	6.85	1.25	0.9		Tac. 1070	8	70	-
Ex. 17	48	28.85	21	-	1.2	0.95	59.56	-	-	-	-
Ex. 17A	48	28.85	21	-	1.2	0.95		11558-84	8	63	42
Ex. 17B	48	28.85	21	-	1.2	0.95		11558-84	10	63	42
Ex. 18	48	27	19.5	3.35	1.2	0.95	59.4	-	-	-	-
Ex. 18A	48	27	19.5	3.35	1.2	0.95		11558-84	10	63	42
Ex. 18B	48	27	19.5	3.35	1.2	0.95		ResA 2430	10	60	
Ex. 18C	48	27	19.5	3.35	1.2	0.95		11558-84	8	63	42
Ex. 18D	48	27	19.5	3.35	1.2	0.95		ResA 2430	8	60	-
Ex. 18E	48	27	19.5	3.35	1.2	0.95		11575-20 <sup>16</sup>	8	70	38
Ex. 18F	48	27	19.5	3.35	1.2	0.95		11575-33 <sup>16</sup>	8	65	34
Ex. 18G	48	27	19.5	3.35	1.2	0.95		11575-34 <sup>17</sup>	8	67	38
Ex. 18H	48	27	19.5	3.35	1.2	0.95		11575-49 <sup>18</sup>	8	61	36.5
Ex. 18I	48	27	19.5	3.35	1.2	0.95		DM462010G <sup>19</sup>	8	62	10
Ex. 18J	48	27	19.5	3.35	1.2	0.95		DM461044G <sup>20</sup>	8	62	44
Ex. 18K	48	27	19.5	3.35	1.2	0.95		11575-70 <sup>21</sup>	8	56	45

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TABLE 1												
	Monomeric Composition (Wt.%)							Tackifier				
	2-EHA <sup>1</sup>	DOM <sup>2</sup>	VAC <sup>3</sup>	BA <sup>4</sup>	AA <sup>5</sup>	MAA <sup>6</sup>	% Solids <sup>7</sup>	Tackifier	% Tackifier	Softening Pt. (°C)	Acid No.	
Ex. 18L	48	27	19.5	3.35	1.2	0.95		SnowT 301A <sup>22</sup>	8	64	120	
Ex. 18M	48	27	19.5	3.35	1.2	0.95		ResA 2467 <sup>23</sup>	8	63	32	
Ex. 18N	48	27	19.5	3.35	1.2	0.95		XR-3013 <sup>24</sup>	8	80	-	
Ex. 19	48	25	18	6.9	1.15	0.95	59.8	-	-	-	-	
Ex. 19A	48	25	18	6.9	1.15	0.95		11558-84	10	63	42	
Ex. 20	51	26.4	19.3	1.5MM	1.03	0.775	59.9	-	-	-	-	
Ex. 20A	51	26.4	19.3	1.5MM	1.03	0.775		11558-84	8	63	42	
Ex. 20B	51	26.4	19.3	1.5MM	1.03	0.775		11558-96	8	61	39	
Ex. 21	49	25.5	19	4.35	1.2	0.95	59.89	-	-	-	-	
Ex. 21A	49	25.5	19	4.35	1.2	0.95		ResA 2441 <sup>25</sup>	8	NA	42	
Ex. 21B	49	25.5	19	4.35	1.2	0.95		ResA 2430	8	60	-	
Ex. 21C	49	25.5	19	4.35	1.2	0.95		ResA 2438 <sup>26</sup>	8	NA	42	
Ctl. 1	see text							-	-	-	-	
Ctl. 2	see text							SnowT 301A	25-30	64	120	

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- 1           <sup>1</sup> 2-ethylhexyl acrylate
- <sup>2</sup> di-2-ethylhexyl maleate
- <sup>3</sup> vinyl acetate
- 5           <sup>4</sup> butyl acrylate
- <sup>5</sup> acrylic acid
- <sup>6</sup> methacrylic acid
- 10           <sup>7</sup> percent solids in untackified polymer. Tackified samples typically differ by about  $\pm 2\%$  from the untackified base polymer
- <sup>8</sup> Tacolyn 1070, manufactured and sold by Hercules
- <sup>9</sup> Lab made aromatic modified aliphatic resin, made by Hercules
- <sup>10</sup> Lab made experimental product containing Tacolyn 1070 and wood rosin, made by Hercules
- 15           <sup>11</sup> See note 9
- <sup>12</sup> See note 10
- <sup>13</sup> See note 10
- <sup>14</sup> See note 10
- 20           <sup>15</sup> Lab made mixed resin dispersion containing gum rosin, made by Hercules
- <sup>16</sup> Lab made mixed resin dispersion containing wood rosin, with KOH used as a neutralizing agent, made by Hercules
- <sup>17</sup> Lab made mixed resin dispersion containing both wood rosin and gum rosin, made by Hercules
- 25           <sup>18</sup> See note 15
- <sup>19</sup> Lab made experimental resin made by Eka Nobel
- <sup>20</sup> See note 19
- <sup>21</sup> Physical blend of wood rosin dispersion and Tacolyn 1070 dispersion, made by Hercules
- 30           <sup>22</sup> Snow Tack 301A, a high acid rosin ester dispersion made by Eka Nobel
- <sup>23</sup> Production scale-up batch of the lab made resin described in note 10
- <sup>24</sup> Experimental aromatic modified aliphatic hydrocarbon resin made by Arizona Chemicals
- 35           <sup>25</sup> 10 gallon scale-up of lab made resin described in note 10, with surfactant variation, made by Hercules
- <sup>26</sup> 10 gallon scale-up of lab made resin described in note 10, made by Hercules
- NA = not available

1            Table 2 compares the adhesive performance of Examples 1-21  
 and Controls 1 and 2 at room temperature. As indicated, the  
 tackified adhesives of the present invention provide  
 significantly better performance than the existing commercial  
 5 products and the untackified base pressure-sensitive adhesive  
 polymers, even at lower coat weights. Consequently, the  
 adhesives of the present invention are less expensive to use than  
 the commercially available products. The effect of adhesive coat  
 weight on adhesive performance is seen by comparing Examples 18  
 10 and 18', which are identical, untackified emulsion pressure-  
 sensitive adhesives, but which were applied using coat weights  
 of 21 and 23 g/m<sup>2</sup>, respectively.

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TABLE 2								
Sample	Coat Wt. (g/m <sup>2</sup> )	90° Peel Initial lbs/inch			Loop Tack Initial lbs/inch			RTS <sup>3</sup> (min.)
		Glass	HDPE <sup>1</sup>	RCB <sup>2</sup>	Glass	HDPE	RCB	SS <sup>4</sup>
Ex. 1	19	2.1	1.4	1	4.1	2.1	1.6	80c
Ex. 2	19	1.7	1.2	1	3.5	2.5	1.1	137c
Ex. 3	19	2.2	1.3	1.1	3.8	2.1	1.7	90c
Ex. 4	19	1.76	1.15	1.15	3.74	1.9	1.82	186c
Ex. 4A	20.6	2.43	1.06	1.84	3.8	2.05	1.9	69c
Ex. 4B	20.6	2.47	1.08	1.72	4.4	2.46	1.65	65c
Ex. 5	NA	NA	0.66	1.72	NA	1.57	1.75	50c
Ex. 5A	NA	1.7	0.84	1.6	3.28	1.9	1.8	31c
Ex. 5B	NA	1.96	1.03	1.62	3.4	2.05	2.1	28c
Ex. 6	20.4	1.75	0.78	1.46	3.04	1.65	1.07	31c
Ex. 6A	21	2.49	1.12	1.6		2.23	2.05	30c
Ex. 7	21	2.13	0.82	1.63	2.4	1.74	1.58	38c
Ex. 7A	21	2.35	1.06	1.44c	3	2.05	1.66	25c
Ex. 7B	20.4	2.07	0.83	1.42p/c	3	1.45	0.56	28c
Ex. 8	20.2	2.02p/c	0.81	0.94p/c	2.82	1.44	1.07	32c
Ex. 8A	20.2	2.27c	0.72	1.6p/c	2.1	1.87	1.74	17c
Ex. 8B	19.8	2.56	1.23	1.34	3.5	2.15	2.25	23c

TABLE 2

Sample	Coat Wt. (g/m <sup>2</sup> )	90° Peel Initial lbs/inch			Loop Tack Initial lbs/inch			RTS <sup>3</sup> (min.)
		Glass	HDPE <sup>1</sup>	RCB <sup>2</sup>	Glass	HDPE	RCB	SS <sup>4</sup>
Ex. 9	20.4	2.33	0.94	2	3.2	1.62	1.8	65c
Ex. 9A	19.8	2.4	1.37	1.62	3.76	2.2	2.4	21c
Ex. 9B	19.8	2.8	1.43	1.72	4.1	2.12	2.18	26c
Ex. 9C	19.8	2.24	1.3	1.5	3.71	1.94	1.96	26c
Ex. 10	21	2.25	0.96	1.7	3.13	2.15	2.3	52c
Ex. 10A	20.4	2.5	0.89	1.7	3.16	1.9	1.6	32c
Ex. 10B	20.6	2.11	1.13	1.4	3.35	2.2	1.6	38c
Ex. 11	19.4	2.2	0.84	1.4	2.46	1.83	2.03	51c
Ex. 11A	19.8	2.22	1.01	1.53	2.65	1.83	1.09	41c
Ex. 11B	20.4	2.56	1.35	1.64	3.91	2.25	2.28	31c
Ex. 11C	20.4	2.33	1.1	1.31	3	2	1.55	35c
Ex. 12	20.2	2.06	0.95	1.45	3.75	1.95	2.5	90c
Ex. 12A	20.5	1.94	0.91	1.4	3.42	1.93	1.6	46c
Ex. 12B	20.6	2.2	1.26	1.67	4.13	2.06	1.81	43c
Ex. 12C	20.6	2.62	1.4	1.42	3.9	2.6	2.1	42c
Ex. 12D	20.4	2.5	1.41	1.6	4.31	2.3	2.1	36c
Ex. 13	NA	1.9	0.95	1.4	3.75	1.75	0.44	NA
Ex. 13A	20.2	1.92	1.14	1.4	3.7	2.11	1.5	35c
Ex. 13B	20.8	2.73	1.3	1.6	4.13	2.15	2.08	25c
Ex. 13C	20.4	2.7	1.5	1.74	4.4	2.4	1.93	31c
Ex. 14	20.2	2	0.9	1.64	3.75	1.5	1.31	60c

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TABLE 2

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Sample	Coat Wt. (g/m <sup>2</sup> )	90° Peel Initial lbs/inch			Loop Tack Initial lbs/inch			RTS <sup>3</sup> (min.)
		Glass	HDPE <sup>1</sup>	RCB <sup>2</sup>	Glass	HDPE	RCB	SS <sup>4</sup>
Ex. 14A	NA	2	0.91	1.5	3.36	2	0.98	NA
Ex. 14B	NA	2	0.97	1.61c	3.2	2	0.98	NA
Ex. 14C	NA	2.24	1.07	1.4	3.4	1.9	1.9	NA
Ex. 15	NA	1.75	0.95	1.71	2.91	1.9	1.54	NA
Ex. 15A	NA	2.03	1.1	1.51	3.51	2.1	1.24	NA
Ex. 15B	20.6	2.44	1.25	1.81	3.4	2.4	2.03	17c
Ex. 16A	20.4	2.11	1.11	1.44	3.3	2.11	1.65	42c
Ex. 16B	20.2	2.6	1.37	1.4	3.5	2.07	1.74	30c
Ex. 16C	19.8	2.4	1.23	1.1	3.03	2.07	1.93	50c
Ex. 17	20.2	1.9	0.99	0.83	3.32	2	2.1	60c
Ex. 17A	20.4	2.6	1.15	2.08	4.12	2.4	2.33	31c
Ex. 17B	20	2.4	1.07	1.64	4	2.1	1.84	29c
Ex. 18	20.9	1.98	1.1	1.3	3.5	1.97	1.6	138c
Ex. 18'	23.1	2.06	1.2	1.5	4.4	2.1	2.3	125c
Ex. 18A	20.6	2.44	1.25	1.81	3.7	2.2	2.55	22c
Ex. 18B	20.4	2.1	1.2	1.45	3.71	1.85	1.72	NA
Ex. 18C	20.4	2.5	1.5	1.82	4.2	2.2	2.13	35c
Ex. 18D	20.6	2.1	1.34	1.45	3.75	2.2	1.64	44c
Ex. 18E	21	2.36	1.26	0.8	4.61	3.15	1.2	53c

TABLE 2

Sample	Coat Wt. (g/m <sup>2</sup> )	90° Peel Initial lbs/inch			Loop Tack Initial lbs/inch			RTS <sup>3</sup> (min.)
		Glass	HDPE <sup>1</sup>	RCB <sup>2</sup>	Glass	HDPE	RCB	SS <sup>4</sup>
Ex. 18F	20	2.92	1.22	1.05	4.4	2.51	1.21	51c
Ex. 18G	20-21	2.6	1.32	0.64	4.8	2.4	1.35	38c
Ex. 18H	21	2.06	1.12	0.8	4.59	3.36	1.11	43c
Ex. 18I	21	2.25	1.26	1.28	4.42	2.54	1.87	54c
Ex. 18J	21	2.3	1.31	1.5	4.94	2.7	2.33	53c
Ex. 18K	21	2.46	1.26	1.6	4.5	2.4	2.2	56c
Ex. 18L	20.8	2.4	1.2	1.1	4.53	2.4	2.17	70c
Ex. 18M	20.8	2.44	1.4	1.4	4.5	2.8	2.4	53c
Ex. 18N	21	2.7	1.3	1.2	5.1	2.7	2	47c
Ex. 19	20.6	1.65	1.03	1.4	3.25	2.05	1.8	60c
Ex. 19A	20.8	2.5	1.2	1.9	3.5	2.36	2.3	22c
Ex. 20A	20	2.6	1.42	1.76	4.25	2.4	1.9	37c
Ex. 20B	20.6	2.75	1.2	1.5	4.16	2.5	2.15	57c
Ex. 21	21	1.97	1.07	1	3.8	2.22	1.88	120c
Ex. 21A	20.6	1.9	1.3	1.28	3.75	2.33	1.73	72c
Ex. 21B	20.6	1.9	1.2	1.14	3.8	2.4	1.65	74c
Ex. 21C	20.6	2.2	1.4	1.45	4.2	2.35	2	63c
Ctl. 1	23	2	1.3	1.4	4.8	2.6	1.9	116c
Ctl. 2	19-20	1.83	1.1	1	3.7	2.17	1.2	22c

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<sup>1</sup> HDPE = High density polyethylene

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<sup>2</sup> RCB = Recycled corrugated board. All peel measurements on RCB showed slight to moderate fiber pick, unless otherwise noted

<sup>3</sup> RTS = Room temperature shear (PSTC No. 7, 6th Ed. (500g load)

<sup>4</sup> SS = Stainless steel

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c = Cohesive failure

p/c = panel cohesive failure

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Table 3 shows the low temperature (25°F) performance of several examples of the pressure-sensitive adhesives of the present invention compared to the commercially available product, Control 1, the tackified commercial product of Control 2, and three untackified base polymers, Examples 4, 12 and 13. Except where noted, each sample was adhered to glass, high density polyethylene, and recycled corrugated board surfaces, and hand peel from each surface was separately tested three times. The adhesives of the present invention provide better performance than the commercial products and the untackified base polymers.

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TABLE 3									
Sample	25° F Hand Peel <sup>1</sup>								
	Glass <sup>2</sup>			HDPE <sup>3</sup>			RCB <sup>4</sup>		
	#1	#2	#3	#1	#2	#3	#1	#2	#3
Ex. 1	2	2	-	1	2	-	2	2	-
Ex. 2	2	3	-	2	2	-	2	2	-
Ex. 3	1	1	-	1	1	-	1	2	-
Ex. 4	2	3	3	2	2	2	3	3	3
Ex. 6A	2	2	3	2	2	2	2	2	2
Ex. 7A	2	2	2	2	2	2	2	2	3
Ex. 9C	2	2	3	2	2	2	2	2	3
Ex. 12	2	2	3	2	2	2	2	2	2
Ex. 12B	1	1	1	1	1	1	2	2	2
Ex. 12C	1	1	1	1	2	2	3	2	2

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TABLE 3									
Sample	25° F Hand Peel <sup>1</sup>								
	Glass <sup>2</sup>			HDPE <sup>3</sup>			RDB <sup>4</sup>		
	#1	#2	#3	#1	#2	#3	#1	#2	#3
Ex. 12D	2	2	2	2	1	1	3	2	2
Ex. 13	2	2	2	2	2	2	2	2	2
Ex. 13B	2	2	2	1	2	1	2	2	2
Ex. 13C	3	3	3	2	2	2	3	2	3
Ex. 15B	2	2	2	2	2	2	2	2	2
Ex. 16B	3	2	3	2	2	2	3	2	3
Ex. 17A	1	2	2	1	1	1	4	2	2
Ex. 18A	2	2	2	1	2	1	2	2	2
Ex. 18C	2	3	2	2	2	2	5	5	5
Ex. 18D	2	2	2	2	2	2	2	2	5
Ex. 18N	3	3	3	1	1	1	1	2	2
Ex. 19A	3	2	2	2	2	2	3	3	2
Ex. 20A	1	1	1	1	1	1	2	2	2
Ex. 20B	2	1	1	1	1	1	2	2	2
Control 1	2	2	1	1	2	1	2	2	3
Control 2	3	4	-	2	2	2	2	2	3

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<sup>1</sup> Subjective hand peel adhesion is expressed as follows: 1 = light adhesion; 2 = moderate adhesion; 3 = tight adhesion; 4 = very tight adhesion; and 5 = facestock/substrate tear.

<sup>2</sup> Removal of each sample from a glass panel was "clean" in all cases except the following: Control 2 - slight tear.

30 <sup>3</sup> HDPE = highly density polyethylene. Removal of the sample was "zippy" in all cases.

<sup>4</sup> RCB = recycled corrugated board. "Fiber pick" was observed in all cases, except Ex. 12C - slight tear.

35 In sheet fed laser printer applications, by feeding the adhesive construction sheets (8\*12 size) at a rate of 133 sheets per minute, the adhesives of the present invention show fewer spots (20 to 40) per page than the existing commercial product (Control 1), which shows 60 spots per page. In normal practice, the converting speeds at which most known tackified acrylic

1 pressure-sensitive adhesives perform well are lower than that of  
 the corresponding untackified adhesives. Surprisingly, the  
 adhesives of the present invention exhibit convertibility  
 characteristics better than tackified commercial adhesives, and  
 5 equivalent to or better than untackified adhesives. Table 4  
 shows the convertibility characteristics of the adhesives of the  
 present invention. Pressure-sensitive adhesive constructions  
 were prepared using DSX paper as a facestock and a 50# FS release  
 liner.

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TABLE 4			
Convertibility Characteristics <sup>1</sup>			
A) Comco Press; max speed of 500 feet per minute			
Sample	DIE-X	DIE-V	AV. Speed
Ex. 1	500	500	500
Ex. 2	500	500	500
Ex. 3	500	500	500
Control 1	500	450	475
Control 2	420	300	360
B) Mark Andy Press; DIE-X			
Sample	Av. Speed	Flags <sup>2</sup>	Hangers <sup>3</sup>
Ex. 18C	875	840	920
Ex. 18M	825	840	870
Control 1	800	840	860

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<sup>1</sup> Values indicate press speed in feet per minute.

<sup>2</sup> Values denote the press speed (ft./min) at which "flags" first appear, i.e., the speed at which the matrix between two labels breaks on one end (but is still removed with the rest of the matrix) during the converting process

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<sup>3</sup> Values denote the press speed (ft./min) at which "hangers" first appear, i.e., the speed at which the matrix between two labels breaks on both ends (and is not removed with the rest of the matrix, but stays on the web) during the converting process



1 **WHAT IS CLAIMED IS:**

1. A pressure-sensitive adhesive, comprising:

(a) a pressure-sensitive adhesive polymer formed by emulsion polymerization of a monomer mixture comprising, on a polymerized basis and based on the total weight of the monomers:

5 (i) from about 35 to about 60% by weight of at least one alkyl acrylate containing from about 4 to about 8 carbon atoms in the alkyl group,

10 (ii) from about 15 to about 35% by weight of at least one vinyl ester containing from 2 to about 16 carbon atoms in the alkyl chain of the ester,

15 (iii) from about 15 to about 35% by weight of at least one diester of a dicarboxylic acid, wherein each alkyl group of the diester independently contains from about 6 to about 12 carbon atoms, and

(iv) from 0 to about 5% by weight of at least one unsaturated carboxylic acid containing from about 3 to about 5 carbon atoms,

20 said polymer having a glass transition temperature of less than about  $-30^{\circ}\text{C}$  and gel content of from about 50 to about 70% by weight of the polymer; and

(b) a tackifier comprising:

(i) a hydrocarbon resin component, and

(ii) a rosin-based resin component,

25 said tackifier having an acid number of from about 30 to about 60 and a ring and ball softening point of from about 50 to about  $70^{\circ}\text{C}$ .

30 2. A pressure-sensitive adhesive as recited in claim 1, wherein the hydrocarbon resin component comprises an aromatic modified aliphatic resin having a softening point of from about 60 to about  $80^{\circ}\text{C}$ .

35 3. A pressure-sensitive adhesive as recited in claim 1, wherein the rosin-based resin component has an acid number of from about 100 to about 210 and a softening point of from about 40 to about  $90^{\circ}\text{C}$ .

- 1        4.    A pressure-sensitive adhesive as recited in claim 1, wherein  
the tackifier has an acid number of from about 30 to about 45.
- 5        5.    A pressure-sensitive adhesive as recited in claim 1, wherein  
the rosin-based resin component is selected from the group  
consisting of wood rosins, gum rosins, tall oil rosins,  
polymerized rosins, disproportionated rosins, hydrogenated  
rosins, partially esterified rosins, and mixtures thereof.
- 10       6.    A pressure-sensitive adhesive as recited in claim 1, wherein  
the rosin-based resin component comprises a wood rosin having an  
acid number of from about 155 to 210.
- 15       7.    A pressure-sensitive adhesive as recited in claim 1, wherein  
the tackifier is present in an amount of from about 5 to about  
25 parts (dry weight) per 100 parts (dry weight) of the pressure-  
sensitive adhesive polymer.
- 20       8.    A pressure-sensitive adhesive as recited in claim 1,  
wherein the tackifier is present in an amount of from about 8 to  
about 15 parts (dry weight) per 100 parts (dry weight) of the  
pressure-sensitive adhesive polymer.
- 25       9.    A pressure-sensitive adhesive as recited in claim 1, wherein  
the tackifier is present in an amount of from about 8 to about  
12 parts (dry weight) per 100 parts (dry weight) of the pressure-  
sensitive adhesive polymer.
- 30       10.   A pressure-sensitive adhesive as recited in claim 1, wherein  
the tackifier comprises an aromatic modified aliphatic  
hydrocarbon resin and a rosin acid, said tackifier having an acid  
number from about 30 to about 45 and a softening point of from  
about 50 to about 70°C, and is present in an amount of from about  
8 to about 12 parts (dry weight) per 100 parts (dry weight) of  
35       the pressure-sensitive adhesive polymer.

- 1 11. A pressure-sensitive adhesive as recited in claim 1, wherein  
the alkyl acrylate comprises a mixture of 2-ethylhexyl acrylate  
and butyl acrylate.
- 5 12. A pressure-sensitive adhesive as recited in claim 1, wherein  
the vinyl ester comprises vinyl acetate.
- 10 13. A pressure-sensitive adhesive as recited in claim 1, wherein  
the vinyl ester is present in an amount of from about 15 to about  
20% by weight, based on the total weight of monomers.
- 15 14. A pressure-sensitive adhesive as recited in claim 1, wherein  
the diester of a dicarboxylic acid is selected from the group  
consisting of di-2-ethylhexyl maleate, di-2-ethylhexyl fumarate,  
and mixtures thereof.
- 20 15. A pressure-sensitive adhesive as recited in claim 1, wherein  
the unsaturated carboxylic acid is selected from the group  
consisting of acrylic acid, methacrylic acid, itaconic acid,  $\beta$ -  
carboxyethyl acrylate, and mixtures thereof.
- 25 16. A pressure-sensitive adhesive as recited in claim 1, wherein  
the unsaturated carboxylic acid is present in an amount of from  
about 1 to about 3% by weight, based on the total weight of  
monomers.
- 30 17. A pressure-sensitive adhesive as recited in claim 1, wherein  
the monomer mixture comprises about 48% by weight of 2-ethylhexyl  
acrylate, about 27% by weight of di-2-ethylhexyl maleate, about  
20% by weight of vinyl acetate, about 3% by weight of butyl  
acrylate, about 1% by weight of acrylic acid and about 1% by  
weight methacrylic acid.
- 35 18. A pressure-sensitive adhesive as recited in claim 1, wherein  
the monomer mixture comprises about 51% by weight 2-ethylhexyl  
acrylate, about 27% by weight di-2-ethylhexyl maleate, about 20%  
by weight vinyl acetate, about 1% by weight acrylic acid and  
about 1% by weight methacrylic acid.

1 19. A pressure-sensitive adhesive as recited in claim 1, wherein  
the monomer mixture further comprises up to about 0.5% by weight  
of a reactive surfactant, based on the total weight of monomers.

5 20. A pressure-sensitive adhesive as recited in claim 19,  
wherein the reactive surfactant is present in an amount of from  
about 0.1 to about 0.25% by weight, based on the total weight of  
monomers.

10 21. A pressure-sensitive adhesive as recited in claim 19,  
wherein the reactive surfactant comprises an anionic vinyl  
functional monomer.

15 22. A pressure-sensitive adhesive as recited in claim 21,  
wherein the reactive surfactant is selected from the group  
consisting of sodium vinyl sulfonate and sodium styrene  
sulfonate.

20 23. A pressure-sensitive adhesive, comprising:  
(a) a pressure-sensitive adhesive polymer formed by  
emulsion polymerization of a monomer mixture comprising, on a  
polymerized basis and based on the total weight of the monomers:  
(i) from about 35 to about 60% by weight of at least  
one alkyl acrylate containing from about 4 to about 8 carbon  
25 atoms in the alkyl group,  
(ii) from about 15 to about 35% by weight of at least  
one vinyl ester containing from 2 to about 16 carbon atoms in the  
alkyl chain of the ester,  
(iii) from about 15 to about 35% by weight of at least  
30 one diester of a dicarboxylic acid, wherein each alkyl group of  
the diester independently contains from about 6 to about 12  
carbon atoms, and  
(iv) from 0 to about 5% by weight of at least one  
unsaturated carboxylic acid containing from about 3 to about 5  
35 carbon atoms,  
said polymer having a glass transition temperature of less  
than about -30°C and a gel content of from about 50 to about 70%  
by weight of the polymer; and

1 (b) a tackifier present in an amount from about 5 to about  
25 parts (dry weight) per 100 parts (dry weight) of the pressure-  
sensitive adhesive polymer, said tackifier having an acid number  
of from about 30 to about 60 and a softening point of from about  
5 50 to about 70°C.

24. A pressure-sensitive adhesive as recited in Claim 23,  
wherein the tackifier comprises a hydrocarbon resin component and  
a rosin-based resin component.

10 25. A pressure-sensitive adhesive as recited in Claim 24,  
wherein the hydrocarbon resin component comprises an aromatic  
modified aliphatic hydrocarbon resin.

15 26. A pressure-sensitive adhesive as recited in Claim 24,  
wherein the hydrocarbon resin component has a ring and ball  
softening point of from about 60 to about 80°C.

20 27. A pressure-sensitive adhesive as recited in Claim 24,  
wherein the rosin-based resin component is selected from the  
group consisting of wood rosins, gum rosins, tall oil rosins,  
polymerized rosins, disproportionated rosins, hydrogenated  
rosins, esterified rosins, and mixtures thereof.

25 28. A pressure-sensitive adhesive as recited in Claim 24,  
wherein the rosin-based resin component has an acid number of  
from about 100 to about 210 and a softening point of from about  
40 to about 90°C.

30 29. A pressure-sensitive adhesive, comprising:

(a) a pressure-sensitive adhesive polymer formed by  
emulsion polymerization of a monomer mixture comprising, on a  
polymerized basis and based on the total weight of the monomers:

35 (i) from about 35 to about 60% by weight of at least  
one alkyl acrylate containing from about 4 to about 8 carbon  
atoms in the alkyl group,

1 (ii) from about 15 to about 35% by weight of at least one vinyl ester containing from 2 to about 16 carbon atoms in the alkyl chain of the ester,

5 (iii) from about 15 to about 35% by weight of at least one diester of a dicarboxylic acid, wherein each alkyl group of the diester independently contains from about 6 to about 12 carbon atoms, and

10 (iv) from 0 to about 5% by weight of at least one unsaturated carboxylic acid containing from about 3 to about 5 carbon atoms,

said polymer having a glass transition temperature of less than about  $-30^{\circ}\text{C}$  and a gel content of from about 50 to about 70% by weight of the polymer; and

(b) a tackifier comprising:

15 (i) a hydrocarbon resin component, and

(ii) a rosin-based resin component,

said tackifier having an acid number of from about 30 to about 60 and a ring and ball softening point of from about 50 to about  $70^{\circ}\text{C}$ ;

20 wherein, when coated on a release liner and laminated to a flexible backing, the pressure-sensitive adhesive exhibits adhesion to both polyolefins and recycled corrugated board substrates.

25 30. A pressure-sensitive adhesive, comprising:

(a) a pressure-sensitive adhesive polymer formed by emulsion polymerization of a monomer mixture comprising, on a polymerized basis and based on the total weight of the monomers:

30 (i) from about 35 to about 60% by weight of at least one alkyl acrylate containing from about 4 to about 8 carbon atoms in the alkyl group,

(ii) from about 15 to about 35% by weight of at least one vinyl ester containing from 2 to about 16 carbon atoms in the alkyl chain of the ester,

35 (iii) from about 15 to about 35% by weight of at least one diester of a dicarboxylic acid, wherein each alkyl group of the diester independently contains from about 6 to about 12 carbon atoms, and

1 (iv) from 0 to about 5% by weight of at least one  
unsaturated carboxylic acid containing from about 3 to about 5  
carbon atoms,

5 said polymer having a glass transition temperature of less  
than about -30°C and a gel content of from about 50 to about 70%  
by weight of the polymer; and

(b) a tackifier comprising:

(i) a hydrocarbon resin component, and

(ii) a rosin-based resin component,

10 said tackifier having an acid number of from about 30 to  
about 60 and a ring and ball softening point of from about 50 to  
about 70°C;

15 wherein, when coated on a release liner to a coat weight of  
about 20 g/m<sup>2</sup> and laminated to a flexible backing, the pressure-  
sensitive adhesive exhibits a 90° peel of greater than about  
1 lb/in. from both HDPE and recycled corrugated board sub-  
strates.

31. A pressure-sensitive adhesive, comprising:

20 (a) a pressure-sensitive adhesive polymer formed by  
emulsion polymerization of a monomer mixture comprising, on a  
polymerized basis and based on the total weight of the monomers:

25 (i) from about 35 to about 60% by weight of at least  
one alkyl acrylate containing from about 4 to about 8 carbon  
atoms in the alkyl group,

(ii) from about 15 to about 35% by weight of at least  
one vinyl ester containing from 2 to about 16 carbon atoms in the  
alkyl chain of the ester,

30 (iii) from about 15 to about 35% by weight of at least  
one diester of a dicarboxylic acid, wherein each alkyl group of  
the diester independently contains from about 6 to about 12  
carbon atoms, and

35 (iv) from 0 to about 5% by weight of at least one  
unsaturated carboxylic acid containing from about 3 to about 5  
carbon atoms,

said polymer having a glass transition temperature of less  
than about -30°C and a gel content of from about 50 to about 70%  
by weight of the polymer; and

1           (b) a tackifier comprising:  
              (i) a hydrocarbon resin component, and  
              (ii) a rosin-based resin component,  
5           said tackifier being present in an amount of from about 5  
to about 25 parts (dry weight) per 100 parts (dry weight) of the  
polymer and having an acid number of from about 30 to about 60  
and a ring and ball softening point of from about 50 to about  
70°C.

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INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US95/10262

**A. CLASSIFICATION OF SUBJECT MATTER**  
 IPC(6) :C08K 5/15; C09F 1/00, 7/00  
 US CL :524/187, 270, 272  
 According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**  
 Minimum documentation searched (classification system followed by classification symbols)  
 U.S. : 524/187, 270, 272

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US, A, 4,477,613 (EVANS ET AL) 16 October 1984, columns 3-5 and examples.	1-31
Y	US, A, 5,189,126 (BERNARD) 23 February 1993, columns 3-7.	1-31
A	US, A, 4,654,389 (GRAHAM ET AL) 31 March 1987.	1-31

Further documents are listed in the continuation of Box C.  See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Z" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search 04 OCTOBER 1995	Date of mailing of the international search report <b>20 NOV 1995</b>
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