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Effect of Amphiphilic Additives on the Behavior of Water-Based Acrylic Pressure-Sensitive Adhesives during Paper Recycling

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Pressure-sensitive adhesives (PSAs) in recovered paper reduce efficiency and increase operating costs for paper recycling mills. Increased PSA fragmentation during pulping and the corresponding reduction in screening efficiency are indications that a PSA will likely interfere with paper recycling. Water-based PSAs, which dominate the label market, have complex formulations that include several amphiphilic materials, i.e., emulsifiers, dispersants, and wetting agents. Increasing the amount of these surface-active materials increased adhesive fragmentation during pulping, and thus reduced screening removal efficiencies. Accompanying the reduction in size was a distinct change in morphology of adhesive particles, which assumed a less collapsed structure during repulping. The presence of surface-active materials also appeared to facilitate the removal of fiber from PSA films during repulping, reducing the importance of paper facestock properties in determining fragmentation behavior. The findings presented here combined with results reported previously provide more complete guidelines for the synthesis and formulation of recycling-compatible acrylic water-based PSAs.

Introduction

Contamination of postconsumer waste by pressure-sensitive adhesive (PSA) presents a significant obstacle to increasing recycling rates.[1−5] These materials break down during repulping, which involves the use of water and mechanical energy to fiberize paper products. Fragmented pieces of adhesive films are mostly removed with conventional screening and cleaning equipment.[6,7] However, the PSA not removed is passed on to the remaining recycling operations and causes deposits on equipment and defects in the final paper product. The cost of the problems created by the presence of PSA contaminants is substantial and has compelled much research on methods for its control. The most promising and widely accepted of these is the redesign of pressure-sensitive (PS) label products to inhibit the fragmentation of PSA films during pulping.

Together, water-based and hot-melt PSAs account for 80% of adhesives used on paper labels, with a 5:1 ratio of water based to hot melt. Previously, it was reported that the removal efficiency of hot-melt PSA is strongly dependent on the properties of the facestock onto which it is attached.[8] For example, the screening removal efficiency of a commercial label-grade, hot-melt PSA was nearly 50% lower when attached to a commercial facestock containing sizing and wet strength additives versus untreated control. It was hypothesized that increased adhesion between the PSA film and facestock in an aqueous environment resulted in films that possessed a less collapsed, more open structure during repulping, which allowed for greater stress to be induced and resulted in more fragmentation. Relative to hot-melt formulations, water-based acrylic PSAs are more complex. After emulsion polymerization, a single adhesive can be modified with tackifying dispersions, wetting agents, rheology modifiers, and other additives to meet a variety of product specifications. Here, the influence of water-based PSA design on adhesive behavior during repulping is examined.

In an earlier publication, the fragmentation of water-based acrylic PSA was shown to be strongly dependent on the monomer composition of the base elastomer.[9] Specifically, it was shown that the presence of both vinyl acetate and acrylic acid monomers produced PSAs that readily broke down during repulping operations resulting in a low screening removal efficiency. This conclusion is based on a study of a model system for which the hard and functional monomers of a commercial formulation that readily fragments during repulping were replaced with more hydrophobic components. For example, in one formulation, methacrylic acid replaced acrylic acid, and in another, methyl methacrylate replaced vinyl acetate. Each of these substitutions produced a significant reduction in fragmentation and a substantial increase in screening removal efficiency. It was also shown that the water-based films rapidly swell in water and that the swelling kinetics likely play a small role in determining fragmentation behavior. The most salient point from this work is that the strength of water saturated PSA films determines their fragmentation behavior during repulping operations and thus the extent to which they can be controlled with mill screening equipment.

In this work, we focus on amphiphilic additives and their influence on PSA behavior during recycling. This includes emulsifiers used during the synthesis, wetting agents used to aid in the coating of PSA films, and surfactants used in tackifying dispersions. We will present information on how these additives change the structure of these films, their mechanical properties and morphologies when fragmented. Furthermore, given that facestock properties affected hot-melt behavior,

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we will show results for water-based PSAs coated on different facestocks. These findings extend previously published results to provide a more comprehensive understanding of how PSA synthesis, formulation, and pressure-sensitive (PS) product design impact the behavior of adhesives during recycling operations.

Experimental Section

Chemicals and Materials. Copper (II)−ethylenediamine complex (1 M solution in water) was purchased from Acros Organics (Pittsburgh, PA). Boise Cascade Corporation (International Falls, MN) provided papermaking fiber and commercial facestocks. Acrylic water-based PSAs were synthesized and formulated at Franklin International (Columbus, OH). Model water-based PSAs were synthesized using commercially available monomers including *n*-butyl acrylate, 2-ethylhexyl acrylate, vinyl acetate, methyl methacrylate, acrylic acid, and methacrylic acid and commercially available sulfated nonylphenol ethoxylate (ammonium salt) and nonylphenoxypoly(ethyleneoxy)ethanol emulsifiers. Many of the adhesive emulsions utilized sodium persulfate or ammonium persulfate initiators, *tert*-butyl hydroperoxide oxidizer, sodium formaldehyde sulfoxylate reducer, an ammonia buffer, and commercial biocides. The wetting agent utilized in the coating package is 50% FX-1 solution purchased from Eagle Sales (St. Louis, MO). Its composition is described as 21% 2-butoxyethanol and 4% ethanol. The tackifying dispersion was obtained from Arizona Chemical (Jacksonville, FL). It is a commercial product, Aquatac 6085, described by the supplier as an aqueous dispersion of rosin glycerol ester with 2−4% surfactant content. In order to cast uniform films without wetting agent, a rheological modifier, Alcogum AN-10, was added at about 1%. Alcogum AN-10, manufactured by Alco Chemical (Chattanooga, TN), is a sodium polyacrylate thickener with solid content of 10% and viscosity of 200−400 cP. The affect of a common commercial defoamer was also examined as part of this study. Foam Blast 325 supplied by Emerald Performance Materials (Cuyahoga Falls, OH) was used in studies at levels of 0.15−0.6%. Emulsions were coated on two-sided release paper using a draw-down coater and dried in an 82 °C oven for 10 min. For most samples, film thickness was targeted at 1 mil or 25.4 μm.

Characterization of PSAs. Procedures used in characterizing the PSAs have been described in detail elsewhere.[3,8,9,15] Here these descriptions are repeated in an abridged form. All performance testing was carried out on 1 mil (25.4 μm) thick films under controlled temperature and humidity conditions, at 23 °C and 50% relative humidity. Peel strengths of water-based PSAs were measured using an IMASS Inc. (Accord, MA) 180° Slip/Peel Tester Model SP101B at a peel rate of 12 in./min. One-inch-wide poly(ethylene terephthalate) (PET) backed films were peeled from PSTC grade polished stainless steel panels (Ryerson Steel, Chicago, IL) rolled with a 4.5 lb ASTM quality manual roller. The average peel force from three tests was reported for each sample. Shear test strips were prepared with a contact area of 0.5 in. × 0.5 in. between PSTC grade polished stainless steel panels and PET-backed films transferred with a 4.5 lb ASTM quality manual roller. Five-hundred-gram weights were clipped to the bottom of the test strips. The average time in minutes for the weight to pull the PET-backed films from the plate for three tests was reported for each sample. Loop tack strengths of water-based PSAs were measured using an Ametek Corporation (Paoli, PA) Chatillon with a DFM 10 Digit Force meter. PSA was direct coated onto PET to form 1 in. × 5 in. test strips, which, for testing, were looped with the adhesive facing out and used to form a contact area of 1 in. × 1 in. on PSTC grade polished stainless steel panels. A dwell time of 1 s and upward travel speed of 12 in./min were used for testing. The average maximum force for three tests was reported for each sample. Glass transition temperatures (T_g's) were determined using the TA Instruments (New Castle, DE) DMA 2980 dynamic mechanical analyzer. Tested samples were composite structures formed by dipping a glass support cloth (30 mm × 10 mm × 0.3 mm) into the liquid adhesive emulsion. The thermal locations of the peaks in the loss modulus (E'') for scans carried out at 1 Hz were reported as the T_g. Force−displacement curves for PSA films were determined using an Instron (Norwood, MA) 5542 tensile tester at a crosshead speed of 10 mm/min. For the dry tests, two pieces of PET films (1 in. wide by 3 in. long strips) were used as a sample holder. A 1 mil adhesive film was transferred from a release liner over the joint between the two PET films, and they were placed in the tensile tester grips. All the dry tests were carried out at 23 °C and 50% humidity. For water-submerged testing, the tensile tester was equipped with an in-house temperature-controlled bath. At least 20 measurements were made for each sample under both dry and wet conditions, and the average maximum forces were reported for each sample. Surface energies of water-based adhesives were determined from contact angle measurements for selected liquids.

Procedure for Testing the Removal of PSA Labels. A known amount of pressure-sensitive adhesive film (≈1.5 g) was transfer coated onto paper facestock (≈5 g) using a heavy roller to produce a label system. Subsequent to the lamination process, the release liner was removed and the PSA/substrate system (≈6.5 g) was attached by hand to envelope-grade paper (≈8 g). These laminates were intermixed with copy paper (287 g). All paper samples were conditioned at 23 °C and 50% relative humidity prior to use. The mixture of copy paper and laminates was then cut into 0.25 in. wide strips using a commercial shredder. Tap water (3 L) that had been heated to the selected temperature with an immersion heater was combined with the shredded sample in an Adirondack 450H laboratory pulper (Adirondack, NY) and mixed at 60 Hz for 30 min. The repulper was equipped with a heating/cooling jacket connected to a recirculating water bath to maintain temperature during testing. The temperature change of the contaminated fiber slurry for a 30-min repulping experiment was determined to be ±2 °C over the range of temperatures investigated. The resulting fiber slurry was passed through a Valley vibrating flat screen equipped with a 15-cut slotted screen (i.e., slotted openings of 0.38 mm), which is a size that provides strict requirements for separation during recycling operations. Screening rejects containing adhesive particles and fiber were removed from the screen plate. Rejected PSA particles were isolated from fibrous material for mass analysis using copper(II)−ethylenediamine (CED) mixed with a magnetic stir rod for approximately 8 h to dissolve cellulose fiber. Adhesive particles were isolated via filtration and dried at 105 °C to a constant weight. Rejected PSA was reported as a removal efficiency (RE), which is the percentage of PSA added to the repulper that is rejected at the screen. The reproducibility of removal efficiency measurements was found to be ±3%. This procedure was described in previous publications,[8,9,15] and was recently shown to correlate well with standard government pilot scale testing for removal efficiencies.[16]

Results and Discussion

Properties of Films Produced from Adhesive Emulsions. Table 1 shows the monomer composition for the model system

Table 1. Monomer Compositions of Model Water-Based PSA Formulations Which Were Synthesized with *n*-Butyl Acrylate (BA), 2-Ethylhexyl Acrylate (EHA), Methyl Methacrylate (MMA), Vinyl Acetate (VA), Methacrylic Acid (MAA), and Acrylic Acid (AA)

	monomer composition					
	soft monomer		hard monomer		functional monomer	
PSA	BA	EHA	MMA	VA	MAA	AA
PSA1	80.8		16		3.2	
PSA2	70.8	10.0	16			3.2
PSA3	70.8	10.0		16.0		3.2

Table 2. Performance Properties, Wet Tensile Strengths, and Removal Efficiencies (REs) Measured at 50 °C for the Model PSAs

PSA	loop tack (N/25 mm)	180° peel (N/25 mm)	shear (min)	wet tensile strength (N)	RE (%)
PSA1	17.8	13.0	214	0.39	90
PSA2	12.2	10.8	2684	0.21	75
PSA3	10.2	10.5	1069	0.05	2

of PSAs used in this study, which are denoted PSA1, PSA2, and PSA3. These are based on a common commercial formulation used in the production of PS labels. The adhesive polymers in water-based acrylics combine so-called soft, hard, and functional monomers to achieve a variety of performance properties. “Soft” and “hard” refer to the glass transition temperatures (T_g’s) of hompolymers produced with these monomers. Soft and hard monomers would produce homopolymers with T_g’s of below about −40 °C and above 30 °C, respectively. Functional monomers are used to stabilize the latex and provide the PSA polymer chain with functional groups where additional chemical reactions can occur. Table 2 lists the performance properties of the model adhesives, wet tensile strengths, and screening removal efficiencies measured at 50 °C. These three PSAs were selected for study to provide a range of fragmentation behaviors. As described in detail previously, monomer composition appears to be the dominant factor controlling their fragmentation during repulping operations.[9] In general, replacing softer monomers with harder ones or using less hydrophilic monomers increases the removal efficiency. Improvements in screening removal efficiencies appear to be associated with increased wet strengths (Table 2). These data suggest that additives that change the interactions with water are likely tied to changes in the strength of the films.

The results presented above represent the behavior of the base PSA formulation. The additives used to form these films (e.g., sodium polyacrylate rheology modifier, various biocides, and defoamers) do not impact the fragmentation behavior of the adhesives.[9] The results reported below are for the additives that are believed to have the more significant impacts on measured removal efficiencies.

Coating Package and Removal Efficiencies. Pressure-sensitive labels are commonly manufactured with water-based PSAs by a process known as transfer coating, in which the formulated adhesive latex is first applied to a release liner and dried to produce a film. Then this liner-backed adhesive film is pressed onto facestock to produce the label stock. Casting uniform coatings of water-based formulations on release liners requires that the surface tension of the aqueous dispersion be reduced. This can be understood through the spreading coefficient ($S_{l/s}$)

$$S_{l/s} = \gamma_s - \gamma_l - \gamma_{ls} \tag{1}$$

which is negative of this free energy change between the final and initial states of a spreading process in which a liquid with a surface tension γ_l is spread on a solid with a surface energy γ_s to form an interface with energy γ_{ls}.[10] A positive spreading coefficient indicates that the liquid spontaneously wets the solid. In the case of water-based PSA formulations, the process of interest involves the wetting of release liner, which has a low surface energy.

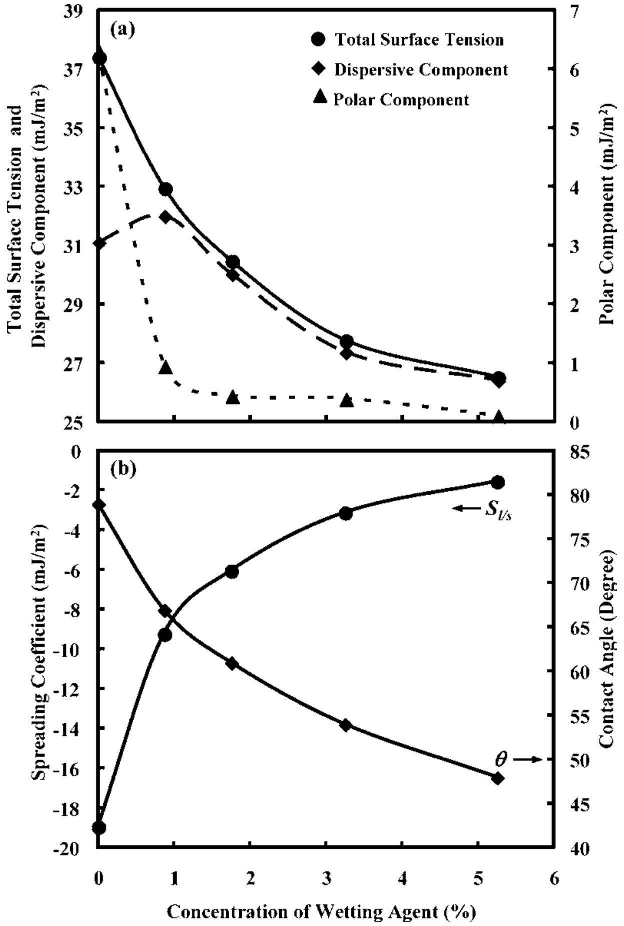

Figure 1. (a) Surface tension and polar and dispersive components of PSA3 with variable levels of wetting agent. (b) Spreading coefficient of PSA3 and the contact angle of this emulsion on release liner with variable levels of wetting agent.

A method for gauging surface energy of a solid is through a series of contact angle measurements using solvents of known surface tensions, a procedure that was described in previous publications.[11,12] This approach assumes that the total surface energy for phase i (γ_i) can be split into contributions from polar (γ_i^p) and dispersive (γ_i^d) interactions, i.e., $\gamma_i = \gamma_i^d + \gamma_i^p$. With these values, interfacial energies between two phases i and j can be estimated using[13,14]

$$\gamma_{ij} = \gamma_i + \gamma_j - 2[(\gamma_i^d \gamma_j^d)^{1/2} + (\gamma_i^p \gamma_j^p)^{1/2}] \tag{2}$$

Measurements done on a variety of release liner indicate that it has surface energy values in the range of 18−30 mJ/m^2 consisting almost solely of dispersive contributions, i.e., $\gamma_i \approx \gamma_i^d$. Combining eqs 1 and 2 and using this assumption, we have

$$S_{l/s} = 2[(\gamma_l^d \gamma_s)^{1/2} - \gamma_l] \tag{3}$$

This indicates that a more positive spreading coefficient is obtained by lowering the surface tension of the liquid phase, while maintaining its dispersive component as high as possible. Figure 1 shows the influence of a commercial wetting agent on the latex for PSA3. Figure 1a shows the effect of the wetting agent on the surface tension and its polar and dispersive

Table 3. Effect of Wetting Agent on Surface Energies, Surface Energy Components, and Work of Self-Adhesion in Water of Model PSA Films; Also Removal Efficiencies (REs) Measured at 50 °C

PSA	wetting agent (%)	surface energy (mJ/m^2)	polar component (mJ/m^2)	dispersive component (mJ/m^2)	work of adhesion (mJ/m^2)	RE (%)
PSA1	0	30.4	0.1	30.3	94.6	90
	0.87	30.9	0.0	30.9	103.6	89
	1.75	37.1	7.7	29.4	39.3	83
	3.50	63.3	32.1	31.2	6.0	74
	5.25	64.4	30.7	33.7	7.7	72
PSA2	0	23.2	0.2	23.0	89.7	75
	0.87	26.3	0.1	26.2	93.6	62
	1.75	34.1	7.6	26.5	38.9	47
	3.50	58.6	30.7	27.9	5.9	23
	5.25	59.8	31.7	28.1	5.4	18
PSA3	0	25.1	0.3	24.8	87.1	2
	0.87	24.6	0.1	24.5	93.3	5
	1.75	25.1	0.0	25.1	96.6	10
	3.50	34.2	7.9	26.3	37.9	2
	5.25	57.6	30.5	27.1	5.8	0

components, while Figure 1b shows how its addition changes both the spreading coefficient and the contact angle for the latex on a release liner with $\gamma = \gamma^d = 25.1$ mJ/m^2. Wetting agents are used in PSA formulations sparingly, usually at levels of 1% or less. This is done to limit the foaming tendency of the formulation, but it also appears to correspond to where the additive has the greatest affect.

Table 3 list the surface energies along with their dispersive and polar components for the model PSA films formulated with various levels of a common wetting agent. Also shown are the cohesion values (maybe better thought of as the work of self-adhesion) and measured screening removal efficiencies. The reversible work, W_A, to separate PSA surfaces in an aqueous environment is simply $2\gamma_{sl}$, where the solid (s) is the adhesive film and water is the liquid (l). It appears that the surfactant retained in the dried PSA films substantially raises the polar components of their surface energies when placed in water and this reduces the screening removal efficiencies for the model PSAs. Insights on how the wetting agent functions to reduce removal efficiencies can be found in the residual particles screened out of the fiber slurry. Figure 2 shows adhesive particles obtained subsequent to repulping for PSA2 with three different wetting agent concentrations. (These are optical images of adhesive particles isolated from a repulping slurry.) It can be seen that films take on a more open structure and, less obviously, have a smaller size as the wetting agent concentration is increased. This is consistent with the expected lower adhesion between PSA surfaces, and the lower removal efficiencies are consistent with the assertion that the greater the surface area of the adhesive film, the greater the fragmentation during repulping operations.

Figure 3 is a plot of the screening removal efficiencies of the three model PSAs as a function of their wetting agent concentrations. It appears that the addition of wetting agent has a significant affect on the removal efficiency of PSA2, but little impact on those of PSA1 and PSA3. The impact of the wetting agent on particle structure during repulping appears to be qualitatively similar for all three PSAs. It is likely that the property most responsible for the observed differences is the wet tensile strength. It can be seen from Table 2 that the wet tensile strength of PSA2 is substantially higher than that of PSA3 and about half that measured for PSA1. The addition of wetting agent did not decrease these values. This suggests that the observed differences come from the base strength of the PSA films. That is, for strong films such as PSA1, the presence of wetting agent has a small impact on its fragmentation behavior because, even in its open configuration, it is strong enough to not break down. The opposite is true for exceedingly weak films such as PSA3; regardless of the additive level, it readily fragments.

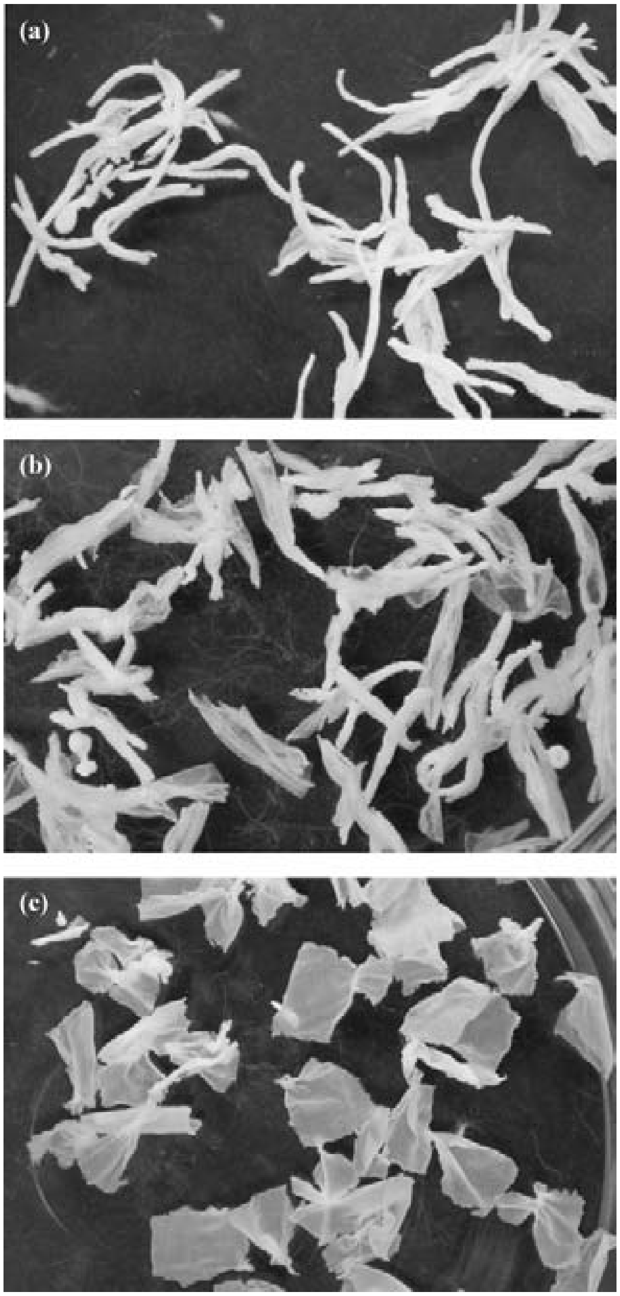

Figure 2. Morphology of adhesive particles after repulping (a) without any additional wetting agent; (b) with 1.75% wetting agent and (c) with 5.25% wetting agent.

Tackifying Dispersions and Removal Efficiencies. Tackifers are used in a PSA formulation to increase the tack of formed films. Table 4 lists the properties of PSA1 and PSA2 produced with tackifying resin concentrations ranging from 0 to 30% (w/w). (PSA3 was left out because its already low removal efficiency means little or no change is possible.) These are levels that are commonly used in water-based acrylic PSAs. A significant increase in tack is observed with the addition of the dispersion. Accompanying this increase is a decrease in measured removal efficiencies. This would appear to indicate that tackifier is reducing the strength of the films. However, the measured wet tensile strengths are shown to increase with tackifier levels.

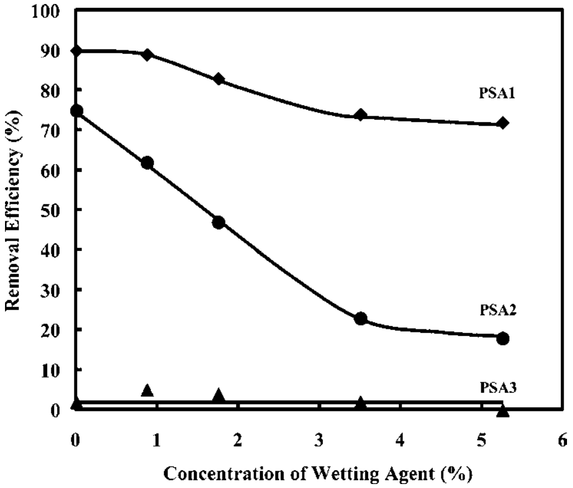

Figure 3. Removal efficiency at 50 °C of model PSAs as a function of the wetting agent concentration used in their formulation.

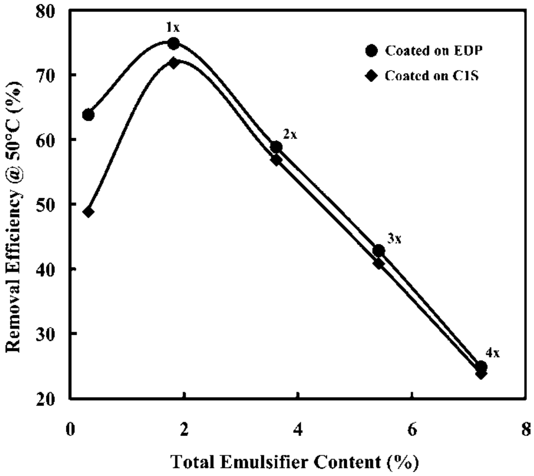

Figure 4. Removal efficiencies for PSA2 coated on EDP and C1S as a function of the emulsifier content. The labels on the data indicate the amount of emulsifier that is part of the latex relative to what is typically used in its synthesis, e.g., 1x is the amount of the emulsifier used in the polymerization and 2x indicates that emulsifier has been added subsequent to synthesis to raise the concentration to twice its typical level in the latex.

Table 4. Effect of Tackifier Dispersion on Loop Tack Value, Glass Transition Temperature, Wet Tensile Strength, and Self-Work of Adhesion in Water of Model PSA1 and PSA2; Also Removal Efficiencies (REs) Measured at 50 °C

PSA	tackifier dispersion (%)	tack (lbs)	T_g (°C)	wet tensile strength (N)	work of adhesion (mJ/m^2)	RE (%)
PSA1	0	2.00	−10.5	0.39	96.8	90
	5	2.46	−9.4	0.40	99.6	91
	10	2.79	−5.9	0.43	100.6	91
	15	2.85	−2.1	0.45	98.8	89
	20	3.11	−1.3	0.46	93.8	88
	30	2.96	−2.1	0.48	45.6	85
PSA2	0	1.70	−22.3	0.21	90.0	75
	5	1.96	−21.1	0.23	95.2	71
	10	2.84	−18.7	0.25	89.2	66
	15	2.75	−16.2	0.28	87.0	60
	20	3.33	−12.1	0.31	84.4	56
	30	3.62	−6.5	0.35	45.4	52

Table 5. Mechanical and Surface Properties of Three Paper Facestocks Used in the Study and Removal Efficiencies at 50 °C of Model PSAs Laminated onto These Facestocks

facestock	basis weight (g/m^2)	tensile loss (%) machine direction	tensile loss (%) cross direction	surface energy (mJ/m^2)	RE at 50 °C (%) PSA1	RE at 50 °C (%) PSA2	RE at 50 °C (%) PSA3
untreated paper	60	98 ± 4		71.1	94	79	10
EDP	77	96 ± 4	96 ± 3	39.7	90	75	2
C1S	77	94 ± 4	94 ± 4	39.7	90	71	4

This result, which appears to contradict our previous findings, can be explained upon closer investigation. The strength of the films does increase with the addition of tackifier. This particular tackifying resin is a glycerol ester rosin, which is a relatively hydrophobic species and has a T_g of about 40 °C, so when it is combined with PSA1 ($T_g = -10.5$ °C) or PSA2 ($T_g = -22.3$ °C) it acts to increase the T_g of the adhesive polymer without increasing its susceptibility to swelling with moisture. However, because the tackifying resin is added to the adhesive polymer in the latex, it is dispersed to form a stable colloid with a surfactant. In this case, the tackifying dispersion contains 2−4% (w/w) surfactant, so the addition of 30% (w/w) tackifier dispersion to the latexes, which have approximately 50% solids levels, introduces as much as 1.2% (w/w) surfactant to the PSA films, which as was described in Table 3 would be expected to decrease the removal efficiencies for the films. Thus, it is reasonable to assume that the benefits to removal efficiencies from the increase in wet strength are offset by the open morphology of the films induced by the accompanying surfactant. Evidence for this explanation is found in the work of cohesion data calculated from the surface energy data (Table 4). It can be seen that, at 30% (w/w) tackifier dispersion levels, this quantity drops for the model PSAs to levels similar to those found when about 1−2% (w/w) wetting agent is added, but the drops in removal efficiencies are not as large. It was also observed that the model PSAs containing high levels of tackifier dispersion had open adhesive films.

Influence of Commercial Facestocks on Removal Efficiencies. Table 5 shows the properties of three paper facestocks used in a previous study examining their influence on the fragmentation behavior of attached hot-melt PSAs.[8] In that study, it was demonstrated that both the tensile loss and surface energy of the facestocks determine how much of an influence they have on removal efficiencies of attached hot-melt PSA. (Tensile loss is the percentage drop in tensile strength with the saturation of paper in water, which is controlled primarily by the wet strength of the paper.) It was proposed that both properties impact the extent to which fibrous materials are retained on film surfaces. A higher wet strength will inhibit the fiberization of the paper, and a lower surface energy results in a higher work of adhesion for removing paper fibers from the PSA film in aqueous environments. In the table it can be seen that the C1S (coated one side) has the lowest surface energy and tensile loss. This is followed by the EDP (electronic data processing) and the untreated paper handsheets.

Listed in Table 5 are the removal efficiencies measured for the model water-based PSAs when attached to the different facestocks. What is interesting is that these changes are not nearly as substantial as those found for the hot-melt PSAs.[8] It is likely that this is due to the composition and structural differences between hot-melt and water-based acrylic PSAs. Although the PSAs used in the facestock study were not formulated with wetting agent, water-based PSAs are produced via emulsion polymerization and thus contain a high content of surfactant. The concentration of surfactant in a water-based acrylic PSA film can be as high as 10% (w/w). This surfactant will likely diminish the adhesion between the PSA film and paper facestock to reduce or eliminate its impact. This hypothesis can be tested with some modifications to PSA2. Figure 4 shows the removal efficiency of PSA2 as a function of the emulsifier content in the PSA formulation. The near 0 sample was produced through the rinsing of films to remove surfactants and their redrying. The 2x, 3x, and 4x samples were produced by simply adding the surfactant to the latex prior to coating. No wetting agent was used in these formulations. It can be seen that removing the emulsifier increases and distinguishes the influences of the C1S and EDP facestocks substantially. The partial removal of the surfactant leads to a decrease in removal efficiencies of more than 20% for the C1S facestock and more than 10% for the EDP. The curves then pass through a maximum, followed by a significant drop in removal efficiencies with increasing emulsifier content in which little or no difference is seen between the influences of the different facestocks. The data again demonstrate a balance between two mechanisms. The impact on the film morphology from the commercial facestocks is diminished by the surfactant at low levels, but with increasing surfactant concentrations the films are dispersed open to an even greater extent resulting in a substantial drop in removal.

Summary and Conclusions

When the results of this study are combined with those reported in previous publications, a general model emerges for the fragmentation of the PSAs commonly used to produce labels. It appears that two major factors govern in large part the extent of the fragmentation and thus screening removal efficiencies. The first is the underlying strength of the film. As discussed, for hot-melt PSAs, reductions in strength sufficient to produce extensive fragmentation are triggered by phase transitions, which are thermally located near common recycling temperatures. The second prevalent factor is the morphology of the film during repulping operations. It appears that the more collapsed the film, the larger the residual particles. This is consistent with a well-established observation that PSAs with the greatest removal efficiencies produce fiberlike residual particles that are tightly wound such as those shown in Figure 2a.

The surfactants (e.g., emulsifiers and wetting agents) found in water-based formulations have an impact on morphology of adhesive films during repulping. Increasing the concentrations of these amphiphilic species produces a substantial reduction in measured removal efficiencies for PSA2, which possesses a moderate wet strength. Far less significant reductions were observed for PSA1, which has the greatest wet tensile strength of the model PSAs, and little change was observed for the weakest model PSA, PSA3. It is theorized that these more open or less wound films promote the induction of greater fragmentation forces during repulping, which has the maximum impact on those films possessing strengths near threshold values. Surfactants were also shown to counter the impact of laminate design. It was observed that the use of commercial paper facestocks, which produce a significant decrease in the removal efficiencies of hot-melt PSAs, had only a minor influence on the water-based PSAs. For these systems it would seem that the surfactant counters the influence of the paper by promoting its removal from adhesive film surfaces. Rinsing and drying the adhesive films prior to lamination results in a decrease in the removal with EDP facestock and a large decrease with C1S facestock. With the addition of higher emulsifier concentrations (post-synthesis), the removal also drops, but little difference is observed between laminates produced with the different facestocks.

This work provides guidelines for the development of recycling-compatible PSAs. The study focused on common commercial additives to demonstrate the reported effects. It is expected that more detailed investigations of any of the additives discussed above will lead to the identification of more effective structures or addition techniques, proving greater flexibility in designing recycling-compatible PSAs.

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