THE EFFECTS OF ADHESIVE PROPERTIES ON THE REMOVAL OF PRESSURE SENSITIVE ADHESIVE CONTAMINANTS IN PAPER RECYCLING

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Abstract

Pressure sensitive adhesive (PSA) materials have significant negative impact on paper recycling. PSA materials are hard to remove in recycling and deposit on equipment and are retained in the product, severely hampering operations. This research had as an objective to evaluate if some PSA properties could be predictors of the removal of PSA materials with pressure screens in paper recycling. Several PSA materials were pulped, characterized for particle size and then subjected to a single slot screening experiment. The PSA materials properties of glass transition temperature, contact angle, yield strain, yield stress, and modulus were determined. Statistical modeling determined that the particle size after pulping was not significantly related to any individual PSA property. Statistical modeling determined that particle passage in a pressurized screen was negatively related to both yield stress and particle dimensions. A hot melt adhesive PSA had a very different screenability than did acrylate based adhesives. The hot melt adhesive was found to fold upon itself into a more spherical shape and to also to contain more attached fibers, in agreement with a higher tack for this adhesive than acrylate adhesives. The passage of the PSA adhesives particles could be modeled with moderate accuracy using a simple extrusion model. The model had yield stress as a fitting parameter and the calculated yield stress from the model was in agreement with the yield stresses measured for the PSA materials. These findings indicate that particle size and cohesive energy are critical to the design of PSA materials that will not extrude through a pressurized screen.

Introduction

Adhesive Contaminants and Paper Recycling

Paper recycling is an integral part of the paper industry, providing about ½ of the fiber used in papermaking in the US. Approximately 100 million tons of paper is produced in the US per year, with 50 million tons of paper recovered for recycling per year. The unrecovered paper is disposed of in landfills. Paper is the largest component existing in landfills by weight being approximately 35% of the total municipal solid waste [1]. Paper recycling is an important service to society and is profitable.

One of the greatest challenges in paper recycling is the complete removal of adhesive contaminants [2-6]. Adhesive contaminants, often referred to as stickies in the paper industry, cause serious problems in paper processing and in final paper properties. These adhesive contaminants deposit on paper recycling and paper making equipment, fouling and plugging the equipment. Acrylate based pressure sensitive adhesive particles that have deposited on a papermachine forming fabric are shown in Figure 1.
This is a considerable issue in papermaking. Typical papermaking involves the distribution of a low consistency stream of fibers in water onto a fast moving plastic wire mesh to drain water, followed by the mechanical pressing of the paper between two press rolls to remove more water, and then heating the paper against steam heated metal drum dryers to evaporate the remaining water. Deposits of the adhesive on the wire mesh, on the press fabrics that absorb the pressed water, and the dryer fabrics that hold the paper tight against the dryer drums and the dryer drums decrease the speed at which the machine can produce paper and necessitate costly down-time to clean the machine. Further, these deposits on wires, fabrics, and drums cause defects in the paper, causing the paper to be rejected. If the adhesive contaminants appear in the paper as observable specks, the paper is often rejected by the customer for visual appearance. Further, if the adhesive is visible or non-visible, the adhesive can interfere with subsequent printing and converting operations. The cost of adhesive contaminant problems has been estimated as 690 million dollars per year in 1996 [2] and 840 million dollars per year in 2005 [3].

Pressure sensitive adhesives PSA are especially troublesome. The paper recycling process is a multi-operation process due to the fact that many different contaminants exist in paper with different properties and different operations with different removal strategies are needed to deal with the myriad of contaminants [7]. As an example, the basic operations involved in converting mixed office waste paper into copy paper will be described here with respect to PSA materials. It is important to realize that in a recycling operation will process around 200-1000 tons per day of material typically. This necessitates extremely intense pumping and mixing. Also, the retention times in typical contaminant removal operations are in the range of seconds. This throughput is necessary to make the process economical but sacrifices removal efficiencies of individual operations.

The first step in recycling is pulping, the disintegration of paper into separated fibers and contaminants in water by the mechanical action of a spinning rotor in a large tank. In this step, PSA containing products are broken up into small particles ranging from sub-visible size and up. This is a critical step in that the smaller the particle is, the more difficult it is to remove. A single PSA label is estimated to be broken into 100’s of particles. Typically the pulp product is then screened and cleaned next.
Screening involves separating large contaminants from fibers by using slots or holes to block the contaminants [8-10]. The size of the slot or hole relative to the size of the contaminant is the main parameter that determines removal. The finest screens available use a 0.004 inch slot (0.1 mm) which is near the limit for screening because fibers have a diameter of about 0.03 mm and smaller slots would cause too much plugging of fibers. For deformable contaminants like PSA’s, the pressure applied to push material through the slot, the temperature and the flow conditions in the screen also can significantly affect the removal. It has been shown that the pressure drop across a slot in a screen has an important effect on screening efficiency [11,12]. Further, the turbulence needed in the screen to keep material fluidized can break the adhesive particles [9,13,14,15]. Screening is the most effective method to remove PSA’s. However, the removal efficiency is typically from 50-80% [8,9]. Most research in adhesive contaminant removal has been involved in understanding how to improve screening.

Centrifugal cleaning is a process in which the pulp (fibers, contaminant and water) are spun at high speeds in a cleaner cone and a separation of solids occurs due primarily to density differences with shape and size being secondary factors. Many designs of cleaners are used, contaminants of both high and low density relative to fibers can be removed. PSA’s, waxes, and plastics are typically of lower density than fibers and are partially separated from fibers with cleaners but with varying and low efficiency. Designing PSA’s with high or low density may not be a solution to removal in cleaners as tacky adhesives adsorb other materials during recycling (inks, clays, water, starch) that can alter the net density of the aggregate.

Washing de-inking is a process in which pulp is diluted with water, small contaminants are dispersed in the water phase, and then the water is removed through a wire mesh with openings of about 0.150 to 0.250 mm. Washing removes small particles, typically less than 0.025 mm in diameter. Most PSA’s do not disperse to this size in water so washing is ineffective in removing PSA.

Flotation de-inking separates the contaminants from fibers based on hydrophobicity differences. In flotation, air bubbles rise through the pulp collecting the hydrophobic contaminants such as inks and toner whereas the more hydrophilic fibers are left. The bubbles form a foam at the surface that is separated from the pulp. The air bubbles collect small particles, 0.010-0.20 mm in diameter, larger particles fall off the bubbles due to shear. Most PSA’s do not fall in this size range and thus are not removed appreciably in flotation.

There has been discussion and efforts to make PSA’s dispersible to be removed in washing and flotation but the paper industry feels that this is not a solution. The dispersed adhesive material can accumulate in the re-circulating waters (water is constantly being re-used to minimize fresh water usage) and potentially cause more problems with processing and product quality.

Other operations in paper recycling include bleaching (the chemical attack of chromophores in the fiber) and dispersion (mechanical action to continue the pulping process) but these operations have minimal impact on the PSA’s.

To summarize, the contaminant removal operations in paper recycling mainly depend on size, density, and hydrophobicity. With respect to PSA’s, screening the large PSA contaminants is the most effective operation but has a highly variable removal efficiency that can often times be low. The pulping process initially determines the size of the PSA particles and can significantly effect the removal in subsequent screening. Thus, characteristics of PSA’s that prevent breakage in pulping and prevent the passage
through slots in screening are the most important routes to developing PSA’s that are readily removed in paper recycling.

**Governmental Efforts to Address the Issue of Adhesives in Paper Recycling**
The government is very interested in promoting paper recycling. By 1995, pressure sensitive postage stamps were becoming widely used and the U.S. Postal Service (USPS) decided to take proactive measures to alleviate recycling problems associated with the PSA materials in the stamps [16]. Members of the paper industry, adhesive industry, converting industry and the USDA’s Forest Products Laboratory worked together to address this issue. This program culminated on July 19, 2001 with the issuing of the USPS P1238-F specification for environmentally benign pressure sensitive adhesive postage stamps. Jointly this team of government and industry experts defined an environmentally benign adhesive (EBA) for stamps and paper label products, developed testing protocols, identified qualified environmentally benign PSA’s (EBPSA’s) and published a listing of them in the Qualified Products List (QPL) in USPS P1238-F [3]. Legislation has been passed to assist the paper recycling industry. Document E.O. 13148, “Greening the Government Through Leadership in Environmental Management”, was signed on April 21, 2000. Section 702 of this Executive Order mandates the use of the EBA’s listed in the USPS Specification’s Qualified Products List for all Government agency paper to paper labeling applications. Currently all postage stamps utilize EBA [3]. However, EBPSA’s are not used exclusively in other tape and label products.

**The Effects of Screening Conditions on PSA Screening Efficiency**
The effects of screening conditions on the passage of PSA particles through a pressure screen has been investigated in this research laboratory [4,5,6,9,11,12,15]. A unique aspect of this research involved the use of a single-slot laboratory device that allowed for the manual control of the pressure across the slot and the direct observation of particle passage [11,12]. Most laboratory research has been done at room temperature with atmospheric screens that do not simulate the pressure drop across an industrial screen. It was found that the dominant variable that predicts particle passage was the size of the particle, Fig 2.

![Figure 2](image)

**Figure 2.** PSA particle (acrylate based) passage through a 0.180 mm slot versus particle area for different temperatures. Particle area is defined as the length times the width of a particle [4,12].
To be more specific, the width of the particle (the second largest dimension) was found to be a critical dimension related to the passage of the particle through the slot whereas the length (longest dimension) for the particles examined did not have as strong a correlation with passage [4,12]. The particle size was determined to be very sensitive to the pulping conditions that generated the particles, and as such is considered to be a pseudo screening processing variable [4,11,12]. Significant amounts of PSA particles with their width of up to four times the slot width (second largest dimension) were able to pass through the slot. The pressure across the slot was determined to be the most important screening condition and its impact on screening PSA’s for a range of commonly found pressures differences in commercial screens is shown in Figure 3.

![Figure 3](image.png)

**Figure 3.** Cumulative PSA particle (acrylate based) passage through a 0.180 mm slot (the percentage of all of the particles tested that were not rejected by the slot) versus pressure difference for different operating temperatures [4,12].

For a given pressure difference across a slot, increased temperature significantly promotes particle passage, Figure 3. In another study, it was found that shear forces in a pressure screen, can cause significant breakage of PSA particles and decrease the screening efficiency [8, 9]. This is especially important for higher fiber consistency [13] in which high shear and elongational stresses are produced.

To summarize, particle size is the most dominant effect in the screening of PSA’s and this can be affected by the pulping process. Increased pressure drop across the slot and higher temperatures both decrease the screening efficiency by promoting the deformation of particles and extrusion through the slot.

**Research Efforts to Predict Screening Efficiency from PSA Properties**

Severtson and co-workers have extensively studied the removal efficiency of hot melt type pressure sensitive adhesives in screening as a function of PSA properties [17-19]. In the studies, the PSA materials investigated consisted of a base polymer that is a block copolymer of styrenic segment of high glass transition temperature ($T_g$) and a low $T_g$ segment consisting of ethylene-propylene, isoprene, butadiene or other. Also present in the formulation are tackifying resin, plasticizer and stabilizer. By comparing lab scale screening results (atmospheric screening and room temperature) at different temperatures and making dynamic mechanical measurements of several PSA formulations, it was found that the screening removal efficiency for all formulations increased to 100% if the pulping temperature...
was much lower than a temperature identified between the glass transitions for the styrenic segments and the rubbery segments [18,19]. The screening experiments executed in the research are labor intensive. To provide PSA manufacturers with a method to rapidly predict screenability without having to do screening experiments, a relationship was developed between screenability of a PSA and the shear adhesive failure temperature (SAFT) and a term $\Delta T$. SAFT is a standard ASTM D-4498 adhesive test that determines the temperature at which a film under stress fails. $\Delta T$ is a measure of the temperature difference between the $T_g$ values of the two phases of the PSA obtained from dynamic mechanical analysis. Figure 4 shows that the screening efficiency can be predicted in terms of these measurable parameters for several hot melt PSA formulations.

![Figure 4](image)

**Figure 4.** Screening removal efficiency of various hot melt adhesives. Left: raw data. Right: results plotted according to model [18]

The removal efficiency was concluded to be related to the cohesive strength of the PSA film if the adhesive is separated from the paper face stock during pulping. The temperature of pulping thus impacts the cohesive strength. Another important factor in the overall strength of the PSA film is the thickness, with thicker films being more screenable [19].

Further, the label backing that the PSA is coated onto has an important impact on the removal efficiency [17,19]. If the label backing is made of an unpulpable material, such as poly(ethylene terephthalate) and the PSA film is retained on the backing, 100% removal efficiency is expected. This is a possible route to make a label benign in recycling. To improve adherence of the PSA film to the backing, the work of adhesion between the backing and PSA film can be increased or the PSA film formulation can be changed to increase the tan delta of the adhesive (a measure of the ability to dissipate energy). For paper backings it was found that by adding sizing agent to make the paper more hydrophobic, the work of adhesion between paper and adhesive was increased. Further, the papers could be made unpulpable by adding a wet-strength agent to the paper during paper production. However, these backings were not representative of commercially available backings.

For common paper backings that disperse during pulping, less than 100% removal efficiency is observed for the backing [19]. For common backings that disperse on pulping, increased sizing (addition of a functional chemical to decrease the surface energy of the paper) decreased removal efficiency. It was suggested that the PSA film is extended on the sized paper for longer times in the pulper and thus is broken into more smaller particles. Another explanation was that for the sized paper, individual fibers were often attached to the PSA film and this prevented the agglomeration and folding of the PSA film causing the PSA film to be extended in the pulper and broken more. It is important to note that the results of the initial experimental backings [17] and the results on the more realistic backings [19] were
very different, suggesting that extreme caution should be used in interpreting lab/experimental results to predict industrial/commercial processes.

Deng and co-workers have studied a large set of emulsion type, polyacrylate-based PSA formulations [20]. Laboratory screening (atmospheric screening and room temperature) was correlated to several common PSA characteristics. In general, the researchers concluded that PSA’s that maintained large size were more screenable. The screenability was found to increase with increased loop tack (Figure 5) and peel strength. Both of these reflect the adhesive and/or cohesive strength of the PSA. The explanation put forth was that (1) the high tack indicates an ability to absorb shear forces during repulping and cause the PSA not to break or (2) PSA materials agglomerated or folded and adhered to itself more, maintaining a large size during pulping.

Figure 5. Screenability versus loop tack for several PSA formulations [20].

Both the shear strength and the tensile strength did not correlate with screenability, presumably because PSA film (thickness less than .025 mm) strength is well below the shear forces in a pulper. The screenability did increase with an increase in elongation index; suggesting that PSA’s with high elongation index had the ability to deform and not break under the forces in a pulper. Increases in the contact angle of water on the PSA film in air (hydrophobicity) were also associated with increased screenability. A possible explanation was that the more hydrophobic PSA will fold on itself or agglomerate during pulping and maintain a large size. Venditti and coworkers also found that detackification of PSA was associated with smaller particle sizes generated during pulping [21, 22].

Various pressure sensitive adhesive materials of known composition were tracked across unit operations in a commercial recycling process producing fine paper from mixed office waste and the number of particles in the accept stream of each operation was reported [23]. The adhesives analyzed consisted of an acrylate, a styrene acrylate, two styrene butadiene rubbers, and a styrene isoprene copolymer based material. The process operations investigated consisted of a pulper, primary screens, secondary screens, and flotation. The acrylate adhesive was the most persistent, having the most particles out of the pulper and after the screens, while the styrene isoprene copolymer had the fewest particles out of the pulper and after the screens. The screens removed about 55% of the acrylate adhesive and effectively 100% of the styrene isoprene copolymer adhesive. The reason why the different adhesives performed differently was not elucidated but this report illustrates that the PSA formulation significantly affects the behavior of PSA particles in industrial screening operations.

In this paper, several properties of acrylate based PSA materials are determined and correlations between these properties and the particle characteristics produced from the pulping operation as well as the subsequent removal efficiency in pressure screens are developed.
Experimental

Adhesive Formulations
Pressure sensitive adhesive (PSA) solutions were prepared with two base polymers from Union Carbide (UCAR9165 and UCAR9175) and two tackifiers from Akzo Nobel (Snowtack 775A (Tack A) and Snowtack 780G (Tack G)). UCAR9165 contains 92% butyl acrylate (BA) and UCAR9175 contains 50% butyl acrylate and 50% 2-ethylhexyl acrylate (BA-EA). Six different adhesive formulations were considered and are shown in Table 1. The concentrations of the base polymers and tackifiers in Table 1 are based on the solids content of the adhesive solution.

<table>
<thead>
<tr>
<th>Sample</th>
<th>BA, %</th>
<th>BA-EA, %</th>
<th>Tack A, %</th>
<th>Tack G, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>BA</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>BA:A</td>
<td>60</td>
<td>0</td>
<td>40</td>
<td>0</td>
</tr>
<tr>
<td>BA:G</td>
<td>60</td>
<td>0</td>
<td>0</td>
<td>40</td>
</tr>
<tr>
<td>BA-EA</td>
<td>0</td>
<td>100</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>BA-EA:A</td>
<td>0</td>
<td>60</td>
<td>40</td>
<td>0</td>
</tr>
<tr>
<td>BA-EA:G</td>
<td>0</td>
<td>60</td>
<td>0</td>
<td>40</td>
</tr>
</tbody>
</table>

The adhesive mixtures were prepared in 200 g batches with solids concentrations of 50%. The mixtures were stirred for 15 minutes in 400 mL beakers before being poured into 500 mL labeled plastic bottles until film preparation.

Also considered in these experiments were two industrial pressure sensitive adhesives, an acrylic based (AC) adhesive film and a styrene-isoprene-styrene (SIS) block copolymer based adhesive film, both provided by Avery Dennison. The exact formulation for these two adhesive films is unknown. Both of these adhesives were provided as films in rolls with the adhesive film between two release liners.

Adhesive Film Preparation
Adhesive films were prepared for each of the six different adhesive solutions by applying the adhesive solution to paper using a coating draw down apparatus. The rod used in the apparatus was a 0.5 inches diameter, #28 wire rod. A sheet of litho paper was secured on a draw down apparatus and a thin strip of adhesive solution was slowly poured in front of the rod. The bar was then drawn over the sheet of litho paper, distributing the adhesive solution into a thin film over the sheet. The adhesive film coated sheet of paper was removed from the coating draw down apparatus and placed in an oven at 60°C to dry for 30 minutes. A sheet of release liner was applied to the adhesive film to protect it until use. Three sheets of adhesive film were prepared for each of the six adhesive solutions. The adhesive films were allowed to condition before being cut into 10 cm by 10 cm labels.
Physical Property Analysis
Some of the mechanical properties, such as the modulus, were determined for each of the adhesive formulations using the Rheometrics Solids Analyzer II (RSAII). A sample containing about 0.030 g of an adhesive solution was applied to the end of a stainless steel rod with a diameter of 7.9 mm. The sample was dried in an oven at 60°C. The rod containing the dried sample was inserted into the top fixture of the RSAII with another rod in the bottom fixture, Figure 6.

![Figure 6. Tensile test sample holder fixtures. Drawing not to scale.](image)

The system was set up in cylindrical tension/compression mode. A dynamic strain sweep in log sweep mode was conducted at a temperature of 50°C, a frequency of 1.0 rad/second, an initial strain of 0.1%, and a final strain of 80.0%. Stress-strain curves were obtained from the RSAII for each of the eight different adhesive samples. Two samples were analyzed for each of the adhesive formulations containing Tack A and four samples were analyzed for the BA-EA base polymer adhesive. The yield strain, yield stress, and modulus were determined from the stress-strain data for each of the adhesive samples. The yield point was determined by fitting the stress strain data in the elastic region to a straight line and then determining the point at which the stress value was at least 1% less than the extrapolated line.

The glass transition temperature ($T_g$) was determined by Differential Scanning Calorimetry (DSC) for each of the dried adhesive formulations using a DSC Q1000 system. The contact angle was determined for each of the eight different adhesive formulations. A 10 cm by 5 cm sample of an adhesive film on paper was secured on the horizontal surface in a goniometer. A drop containing 2 mL of deionized water was dropped onto the surface of the adhesive film using a 100 cm³ syringe. The lines in the scope were aligned with the surface of the film and the edge of the drop of water. The contact angle was recorded at 10, 20, and 30 seconds after the water was dropped onto the adhesive film surface. The tack was evaluated for the acrylic and a SIS pressure sensitive adhesive film. The adhesive films, between two sheets of silicone release liner, were cut into 2.0 cm wide strips. One sheet of release liner was removed from a 2.0 cm wide adhesive film. The strip was applied to a square area of 2.0 cm x 2.0 cm on a 1.2 kg stainless steel weight. The combination of the adhesive strip and weight were placed on a balance. The adhesive strip was slowly pulled up from the weight, without the weight lifting off of the balance surface, until the adhesive released from the weight. When the adhesive released from the weight, the reading on the balance was recorded. The measurement was conducted five times for each
of the adhesive films. The tack was calculated as the force required for the adhesive film to release from the weight per unit area of the adhesive film in contact with the weight.

**Pulping**
Six 10 cm by 10 cm adhesive labels were applied to copy paper and then pulped in a 450H pulper. The operating parameters for the 450H pulper were 450 OD g of pulp, 12% consistency, 50°C, and 415 rpm, for 30 minutes. Pulp samples were removed from the 450H pulper at 10 and 30 minutes of pulping time. Handsheets were prepared from the pulp samples for dyeing followed by image analysis.

The dyeing procedure that was used is as follows. Each handsheet in a set was submerged in Morplas Blue-Heptane solution for ten seconds and then hung up to dry. Once the set of handsheets was dry, each handsheet was submerged in 95% Heptane for ten seconds and then hung up to dry. Once the set of dyed and rinsed handsheets was dry, the set was removed and placed in a labeled plastic bag.

Image analysis was used to determine the stickies content of the samples for the different sets of handsheets. For image analysis, the SpecScan 2000 program by Apogee Systems Inc. was used to scan each set of handsheets. The scanner was a UMAX PowerLook III with a 600 dpi resolution. Both the felt and wire sides of each of the handsheets in a set were scanned. The parts per million (ppm), the number of particles in 1 m², and the average particle size were determined for each side of each handsheet. Also, a histogram of particle sizes in terms of area and count were recorded. Particle size was compared to the physical properties of the different adhesive materials using SAS version 8 from the SAS Institute.

**Single Slot Experiments**
Individual PSA particles for each of the adhesive formulations were analyzed in a single slotted screening device for particle passage, Figure 7. The single slotted screening device consists of a metal plate, a plastic Buchner funnel, a vacuum flask, and a vacuum pressure gauge. The metal plate has a slot cut in the middle that is 0.007 inches (0.18 mm) wide and 1 inch long. The metal plate containing the slot is 0.25 inches thick. The metal plate is secured to the Buchner funnel, with the middle cut out of it, by silicone and all of the holes around the metal plate are filled with silicone. The Buchner funnel fits in the tip of the vacuum flask. The vacuum flask is attached to a house vacuum valve by rubber tubing, with the vacuum gauge in between the flask and the valve.

Adhesive particles were removed manually from a pulp sample produced in the pulping experiments. The length, width, and thickness (largest, middle, and smallest dimension, respectively) were measured for each adhesive particle under a microscope. Each adhesive particle was then allowed to soak in deionized water at 50°C for five minutes, before being placed on the 0.007 inches wide slot of the single slot device. The length of the particle was aligned with the length of the slot. The device was then filled with 1000 mL of deionized water at 50°C. A vacuum was applied to the single slot by slowly opening the vacuum line. If the particle passed through the slot, the vacuum pressure at which the particle passed was recorded and the particle was recovered from the device. The maximum pressure drop across the slot was 74 kPa. The particle passage was determined for each of the adhesive formulations, based on the number of particles that passed through the slot compared to the total number of particles that were analyzed.
**Results and Discussion**

**Pulping of Pressure Sensitive Adhesive Films**

The image analysis results from the pulping experiments for the eight different PSA films are shown in Table 2. The results consist of the parts per million, number of particles in 1 m², and average particle size for particles larger than 0.040 mm². In comparing the two base polymers, BA-EA produced significantly larger particles than BA. AC, the commercial label product, produced the largest average particle size out of the pulper after 10 and 30 minutes. The average particle size of SIS did not change between 10 and 30 minutes in the pulper, while the average particle size decreased for all of the other adhesive films between 10 and 30 minutes in the pulper.

In comparing the AC and SIS adhesive films to the adhesive films with known formulations, AC had the largest average particle size and SIS had the fewest number of particles in 1 m² (Table 2). This is in agreement with an industrial trial that showed acrylate PSA to produce significantly more particles than SIS PSA after pulping [23].
Table 2. Image Analysis Results for Eight Different PSA Materials

<table>
<thead>
<tr>
<th>Sample</th>
<th>Pulping Time, minutes</th>
<th>Parts Per Million, ppm</th>
<th>Number of Particles in 1 m²</th>
<th>Average Particle Size, mm²</th>
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<tbody>
<tr>
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<td></td>
<td>30</td>
<td>2040</td>
<td>1880</td>
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</tr>
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<td></td>
<td>30</td>
<td>2880</td>
<td>5570</td>
<td>0.52</td>
</tr>
<tr>
<td>BA:G</td>
<td>10</td>
<td>1800</td>
<td>1870</td>
<td>0.96</td>
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<td></td>
<td>30</td>
<td>2140</td>
<td>2860</td>
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</tr>
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<td>BA-EA</td>
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<td></td>
<td>30</td>
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<td>1690</td>
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</tr>
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<td>30</td>
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<td>1660</td>
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</tr>
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<td>SIS</td>
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<td>860</td>
<td>950</td>
<td>0.90</td>
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<td></td>
<td>30</td>
<td>1160</td>
<td>1280</td>
<td>0.91</td>
</tr>
</tbody>
</table>

A plot of cumulative particle size distribution from image analysis for the pulps out of the pulper at 30 minutes is shown in Figure 8. This plot shows that BA-EA and AC had the fewest particles less than 0.50 mm² of the eight different adhesives analyzed. The adhesive formulations containing the same tackifier had similar particle size distributions, regardless of the base polymer. Also, the adhesives containing tackifiers had fewer large particles than the base polymer or commercial adhesives.

Figure 8. Cumulative Number of Particles Versus Particle Size Based on Image Analysis of Dyed Handsheets
An AC and SIS based adhesive particle are shown in Figure 9. The images were captured with a microscope at 50x magnification. The two images show a considerable difference in the shape of the particles. The AC particle is significantly longer and more string like than the SIS adhesive particle, characteristics that should have a negative impact on screening. The SIS particle is more 3-dimensional with a much larger second larger dimension than the AC particle. The SIS particle also has a fiber attached to it. From inspection of many particles it was determined that the majority of the SIS particles had fibers attached. In contrast, a small minority of the AC particles had fibers attached.

The tack of the acrylic and SIS adhesive films was measured to be 2.5 kN/m² and 4.4 kN/m², both with a standard deviation of 0.2 kN/m². It is reasonable to suggest that this had two interesting impacts during pulping/screening. First, the SIS particles with higher tack folded upon themselves into tighter more 3-dimensional particles that could be blocked in screening (see later). Also, the fibers still attached to the more tacky particles offered an extra resistance to passing through the slots.

Figure 9. Acrylic (left) and SIS (right) based PSA adhesive particle.

Physical Properties of the Pressure Sensitive Adhesive Films

The physical analysis results for the eight different PSA films are shown in Table 3. The contact angle values are those measured at 10 seconds after the drop was on the sample surface. In comparing the two base polymers, BA-EA has a lower Tg and a lower modulus than BA. However, BA-EA has a higher yield strain and a higher yield stress than BA. In fact, BA-EA has the highest yield stress and yield strain of any of the adhesive materials analyzed. In comparing the adhesive formulations containing tackifiers, the samples containing the same tackifier have similar Tg, yield strain, and yield stress values. In general, the addition of a tackifier reduces the strength properties of the adhesive material. Among all eight of the adhesives, SIS has the highest modulus. In comparing the average particle size in Table 2 and the properties in Table 3, there does not appear to be a strong direct relationship between the average particle size out of the pulper at 30 minutes and any single property by itself, Figure 9. This was confirmed using SAS Version 8 statistical package from SAS Institute [4].
Table 3. Properties of Different PSA Materials

<table>
<thead>
<tr>
<th>Sample</th>
<th>T&lt;sub&gt;g&lt;/sub&gt;, °C</th>
<th>Contact Angle, °</th>
<th>Yield Strain, %</th>
<th>Yield Stress, kPa</th>
<th>Modulus, MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>BA</td>
<td>-37</td>
<td>80</td>
<td>2.9</td>
<td>25</td>
<td>0.84</td>
</tr>
<tr>
<td>BA:A</td>
<td>-20</td>
<td>67</td>
<td>0.78</td>
<td>3.0</td>
<td>0.38</td>
</tr>
<tr>
<td>BA:G</td>
<td>-12</td>
<td>78</td>
<td>3.5</td>
<td>18</td>
<td>0.51</td>
</tr>
<tr>
<td>BA-EA</td>
<td>-49</td>
<td>98</td>
<td>19</td>
<td>56</td>
<td>0.30</td>
</tr>
<tr>
<td>BA-EA:A</td>
<td>-23</td>
<td>75</td>
<td>0.67</td>
<td>1.5</td>
<td>0.22</td>
</tr>
<tr>
<td>BA-EA:G</td>
<td>-16</td>
<td>92</td>
<td>6.4</td>
<td>20</td>
<td>0.31</td>
</tr>
<tr>
<td>AC</td>
<td>-28</td>
<td>80</td>
<td>0.86</td>
<td>6.0</td>
<td>0.69</td>
</tr>
<tr>
<td>SIS</td>
<td>-27</td>
<td>92</td>
<td>3.1</td>
<td>44</td>
<td>1.4</td>
</tr>
<tr>
<td>Tack A</td>
<td>17</td>
<td>70</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Tack G</td>
<td>33</td>
<td>74</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Passage of the Pressure Sensitive Adhesive Particles Through a Slot

The percentage of particles passing through the slot and some average size characteristics are listed in Table 4. Particle area is the particle length times the particle width, as viewed under the microscope. Particle volume is the particle area times the particle thickness. Among the six adhesives with known formulations, this data shows that the base polymers had the longest and widest particles out of the pulper, and they provided the largest average particle area and the largest average particle volume. The adhesive formulations containing the same tackifier resulted in similar particle passage, regardless of the base polymer.

Figure 9. PSA characteristics versus average particle size after pulping.


**Table 4. Particle Passage and Average Particle Dimensions for Eight Different Adhesive Formulations**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Length, mm</th>
<th>Width, mm</th>
<th>Thickness, mm</th>
<th>Area, mm²</th>
<th>Volume, mm³</th>
<th>Particle Passage, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>BA</td>
<td>3.08</td>
<td>0.36</td>
<td>0.31</td>
<td>1.15</td>
<td>0.38</td>
<td>38</td>
</tr>
<tr>
<td>BA:A</td>
<td>1.91</td>
<td>0.35</td>
<td>0.32</td>
<td>0.68</td>
<td>0.22</td>
<td>70</td>
</tr>
<tr>
<td>BA:G</td>
<td>2.21</td>
<td>0.33</td>
<td>0.27</td>
<td>0.77</td>
<td>0.22</td>
<td>30</td>
</tr>
<tr>
<td>BA-EA</td>
<td>2.72</td>
<td>0.38</td>
<td>0.33</td>
<td>1.06</td>
<td>0.38</td>
<td>5</td>
</tr>
<tr>
<td>BA-EA:A</td>
<td>1.61</td>
<td>0.31</td>
<td>0.27</td>
<td>0.50</td>
<td>0.14</td>
<td>75</td>
</tr>
<tr>
<td>BA-EA:G</td>
<td>1.44</td>
<td>0.35</td>
<td>0.30</td>
<td>0.48</td>
<td>0.16</td>
<td>30</td>
</tr>
<tr>
<td>AC</td>
<td>2.68</td>
<td>0.46</td>
<td>0.36</td>
<td>1.27</td>
<td>0.50</td>
<td>30</td>
</tr>
<tr>
<td>SIS</td>
<td>1.31</td>
<td>0.44</td>
<td>0.37</td>
<td>0.57</td>
<td>0.22</td>
<td>10</td>
</tr>
</tbody>
</table>

A plot of cumulative particle size distribution for the particles used in the single slot experiments, with the dimensions measured manually under a microscope, is shown in **Figure 10**. It is clear that the formulation differences are accompanied by different size distributions of particles. It is of interest that the SIS and AC commercial products have a very different size distribution.

![Figure 10](attachment:image.png)

**Figure 10. Cumulative Particle Size Distribution for Particle Area Measured Manually Using a Microscope**

**Figure 11** is a plot of cumulative particle passage versus particle area for the eight different adhesive formulations considered in the single slot. For a given formulation, as the particle area increases, particle passage decreases. This confirms that particle size is a very significant factor in particle passage. However, as the particle area increases, the particle passage decreases at different rates for different adhesive formulations due to the different particle size distributions. The SIS particle passage is considerably lower than other formulations at the same particle size (**Figure 11**).
Plots of particle passage versus adhesive material yield strain, yield stress, and modulus are shown in Figure 12, 13, and 14, respectively. The material with the lowest yield strain has the highest particle passage and the material with the highest yield strain has the lowest particle passage (Figure 12). The material with the lowest yield stress has the highest particle passage and the material with the highest yield stress has the lowest particle passage (Figure 13). It also appears that the material with lowest modulus has the highest particle passage and the material with highest modulus has one of the lowest passage (Figure 14). However the correlation between modulus and passage in the data is weak. These results indicate that as the PSA becomes more deformable (as reflected in lower yield stress and strain and modulus), the particle passage increases, a reasonable finding.
Particle passage was compared to the overall average dimensions of the particles and the physical properties of the adhesive materials using SAS. The overall average dimensions of the particles alone were not significant in the prediction of particle passage [4]. As an example, the SIS average particle size was small but the passage was low (Table 4). Note that this does not mean that the size of individual particles is not a major factor in particle passage for any given formulation. For particles of a certain formulation, increased size decreases passage (see Figures 2 and 11). One must also consider that the average particle size does not reflect the particle size distributions, a PSA with significant fraction of small particles may be less screenable than expected from the average particle size because of the small particle sub-group contribution. Also, the $T_g$ and contact angle were not significant in predicting particle passage [4].

It was interesting to note that the contact angle was not significantly related to particle passage under pressurized screening conditions, but previous work by Deng and coworkers [20] found a trend between screenability in a non-pressurized screen and contact angle. From the work of Severtson and coworkers [18], it would be expected that the $T_g$ of the PSA materials may have some correlation with the screenability. However, the screening temperature in this study was at least 60°C higher than the $T_g$ values of all of the acrylate based adhesives, with no higher $T_g$ phase being present. It stands that at temperatures so high above the $T_g$ that the deformability of the particles as reflected in the yield stress...
and strain and modulus is more important than the $T_g$ value. It is known that at temperatures above the $T_g$ for polymer solutions or melts that the modulus of a material depends on effective crosslinks of the polymer (either from physical entanglements of high molecular weight species, or chemical covalent bonds or physical bonds such as ionic bonds). These effective crosslinks that act to provide resistance to stress relaxation and deformation may influence the passage of the PSA through the slot but not the absolute value of the $T_g$.

It is important to note that the SIS adhesive has two $T_g$ values, a lower one corresponding to the isoprene block (as reported in Table 3) as -27°C and a higher one corresponding to the styrene block (not reported in Table 3) of around 100°C for amorphous styrene. The styrene blocks form a second minority phase of glassy islands providing an effective crosslink up to 100°C. From Figure 11 it is clearly observed that the SIS at the same particle size has a lower particle passage than the acrylates. It stands to reason that this is due to the SIS being the only PSA material with significant crosslinks active at the screening temperature of 50°C.

Note that the report herein includes the direct comparison of acrylate and hot melt adhesives whereas the reports by Deng and coworkers [20] only considers acrylate and the reports by Severtson and coworkers [17-19] only considers the hot melt adhesives. The direct comparison of these two classes of PSA’s in this study confirm the industrial trial results of the more benign behavior of the hot melts relative to the acrylic based PSA [23].

For the mechanical properties, yield stress was the individual property that was most significant in predicting the particle passage. The equation for particle passage as a function of the yield stress is:

$$\text{Particle Passage} = 59.25 - 1.08 \times \text{Yield Stress}$$

where the units are percent for particle passage and kPa for yield stress. For this equation, the $R^2$ value is 0.72 and the $F$ value is 15.30, indicating that 72% of the variability of the passage can be attributed to the yield stress. The calculated versus actual particle passage based on this model is shown in Figure 15.

![Figure 15](image-url)  
**Figure 15.** Calculated versus actual particle passage using model with yield stress only.
The most predictive model containing two variables consisted of yield stress and average particle area:

\[
\text{Particle Passage} = 99.52 - 2.54 \times \text{Yield Stress} + 1.74 \times \text{Yield Stress} \times \text{Area} - 50.18 \times \text{Area}
\]

where the units are percent for particle passage, kPa for yield stress, and mm\(^2\) for average particle area. For this equation, the \(R^2\) value is 0.90 and the F value is 21.41. The calculated versus actual particle passage based on this model is shown in Figure 16.

![Figure 16](image)

**Figure 16.** Calculated versus actual particle passage using model with yield stress and average particle area

This model of particle passage as a function of yield stress and average particle area is physically reasonable. The particle passage decreases as the yield stress increases because particles with a higher yield stress are less likely to deform in order to pass through the slot than particles with a lower yield stress. This has been modeled and experimental data is in agreement with an extrusion model that used yield stress as a primary fitting parameter [4]. The particle passage decreases as the average particle area increases because larger particles require more deformation than smaller particles in order to pass through the slot.

**Conclusions**

Adhesive formulation can significantly impact the PSA particle size produced from pulping and the screenability. Statistical modeling determined that the particle size after pulping was not significantly related to any individual PSA property. Statistical modeling determined that particle passage in a pressurized screen was negatively related to both the yield stress and the average particle size. The passage of the PSA adhesives particles could be modeled with moderate accuracy using a simple extrusion model that indicated that yield stress was an important factor in passage.
References


