PILOT TESTING AND RECYCLING EVALUATION OF NEWLY DEVELOPED ENVIRONMENTALLY BENIGN PRESSURE SENSITIVE ADHESIVES

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ABSTRACT

As a result of an initiative by the U.S. Postal Service (USPS), a team is working cooperatively to help solve the problem of pressure sensitive adhesives (PSAs) in recovered paper. The team consists of the USPS; the USDA Forest Service, Forest Products Laboratory (FPL); Springborn Testing and Research (STR); and industry representatives including papermakers, paper recyclers, paper collectors, equipment manufacturers, and adhesive and chemical suppliers The team's goal is to work with industry to develop new environmentally benign PSAs for postal applications that will fulfill both USPS specifications and be compatible with the USPS environmental strategic plan. Environmentally benign PSA is a form of adhesive that places no sigificant additional burden on plants that are using recycled fiber.

Based on the criteria set by the paper industry, the adhesive industry is now striving to formulate new PSAs that are mostly removable in the screening step of the recycling process. As a result of these combined efforts, a pilot-scale separation sequence has been developed to assess the removal of adhesives from various feed stocks containing PSA. This pilot-testing protocol simulates a typical recycling operation, including high consistency pulping, coarse and fine pressure screening, forward and reverse cleaning, washing, and flotation deinking. An optical protocol using image analysis was developed to measure residual adhesive. Handsheets made from pulp accept samples taken after each unit operation and on paper samples from the paper machine runs were analyzed by this protocol. The recycling protocol simulates a typical recycling operation and uses both preconsumed, postconsumed, and control stocks and was further refined to reflect optimum operating conditions, such as temperature, consistency, screen size, repulping energy, and chemical additives.

In this paper, the development of the protocol is explained and data analysis is given for control trials and a trial using experimental PSA that was screenable and appears to meet established criteria for recyclability.

INTRODUCTION

Removing contaminants from recovered paper pulps is one of the biggest technical barriers to paper recycling. Contaminants from adhesives are an undesirable recovered paper component that comes from pitch, ink, plastic films, convening aids, paper coatings, and adhesives. Adhesives are either hot melt or pressure sensitive. Pressure sensitive adhesives (PSAs) come from products such as labels, tapes, and some postal materials. There are several methods of quantifying contaminants from adhesives, but there is no agreement on a standard method. Despite the advances made during the last few years, contaminants from adhesives, called stickies by papermakers because they stick to paper machine felts and wires, are a major problem during both the processing of recovered paper and the papermaking operation. Closing the water loop by recycling water within mills and reusing and shifting to an alkaline-based papermaking made the problem even more difficult

As a result of an initiative by the U.S. Postal Service (USPS), which currently purchases about 12% of domestic PSA production, a team consisting of the USPS, the Forest Products Laboratory (FPL) (USDA Forest Service), and industry representatives is working cooperatively to help solve the problem of PSAs in recovered paper. Industry representatives include papermakers, paper recyclers, paper collectors, equipment manufacturers, and adhesive and chemical suppliers. The team's goal is to work with the industry to develop new environmentally benign PSA for postal applications that will fulfill USPS specifications and be compatible with the USPS environmental strategic plan. Environmentally benign PSA places no significant burden on plants that are using recycled fiber.

The USPS, in conjunction with FPL, Springborn Testing and Research (STR). and paper industry representatives. organized several national PSA meetings to incorporate the input of the adhesive manufacturing industry and the paper manufacturing industry. The issues included defining the problem, communication, education, source of quality control, recyclability standards for PSAs, paper industry perspective, end-product specifications, and the role of technology. The meetings concluded that problems caused by stickies are best resolved through the combined efforts of the adhesive industry, the paper industry, and the process equipment manufacturers. Based on the criteria set by the paper industry, the adhesive industry is striving to formulate new PSAs that are mostly removable in the screening step. The latest meeting was held at FPL to discuss and decide on the image analysis and pilot plant testing protocols needed to evaluate the newly produced adhesives on pilot scale. The team agreed that due to a lack of information on the removal efficiencies during recycling unit operations, there is a need for a standard testing method and a need to develop a reasonable method combining pilot testing and image analysis.

As a result of these combined efforts. a pilot-scale separation sequence was developed to assess the removal of adhesives from a feed stock containing PSA. This pilot-testing protocol, simulating a typical recycling operation, included high consistency pulping, coarse and fine pressure screening, forward and reverse cleaning, washing, and flotation deinking. An optical protocol using image analysis was developed to measure residual adhesive. Handsheets made from pulp accept samples taken after each unit operation and on paper samples from the paper machine runs were analyzed by this protocol. The recycling protocol simulating typical recycling operations and using both preconsumed, postconsumed, and control stocks was further refined to reflect optimum operating conditions, such as temperature, consistency, screen size, and repulping energy.

METHODS

This section discusses the development of the protocol and includes information on the control trials, operating conditions, system cleaning, and testing. A copy of the protocol and a flow diagram are given at the end of this paper.

Pilot Testing Protocol Development

As mentioned, the pilot testing protocol was developed as a joint effort by the USPS, FPL, STR, and various industries representing adhesive manufacturers, papermakers, stamp paper suppliers, and recyclers. The unit operations were high consistency pulping (14% consistency, 43 C, 20 min), two-stage (0.3 and 0.15 mm) slotted screening, forward cleaning, two-stage reverse cleaning, flotation, washing, and pressing. A final pulp resulting in a din count of 3 parts per million (ppm) or less was set aside for a experimental papermachine run. From those final pulps having a dirt count >25 ppm, a sample was subjected to the additional cleaning operations of dispersion followed by flotation and washing to determine whether the residual adhesive would break down into smaller pieces and be removed by the additional flotation stage. A complete, derailed copy of the protocol/procedure is attached.

Sample Contents, Preconsumer, and Postconsumer Simulation

Fourteen stamp stocks containing experimental adhesives were evaluated, with each trial using 90% copy paper plus 10% stamp stock. For the preconsumer tials, the 10% stamp stock was weighed out as received, including the release paper. For the postconsumer trial, the same amount of stamp stock was weighed out but the release liner was removed, the stamp paper was affixed to copy paper, and an extra amount of copy paper equal to the weight of the release liner was added. This 5% stamp stock represents a much greater amount than expected in a typical printing and writing recovered paper stock.

Benchmark and Control Trials

A series of control trials were conducted to provide a reference for assessing the effectiveness and repeatability of the cleaning and screening operations. The four control runs included the following:

- 100% copy paper to determine the best level of cleaning achievable.
- 90% copy paper plus 10% stamp face paper to simulate the best cleaning level without adhesive.
- 90% copy paper plus 10% conventional PSA linered stamp stock to provide a basis of comparison for the experimental adhesives and representing a “preconsumer” blend.
- 95% copy paper plus 5% linerless PSA printed stamp stock to provide information on how this new construction would compare with the standard linered stamp, especially the effect of the presence of ink.

Operating Conditions

All operating conditions used for the protocol were to be typical operating conditions. Three temperatures (43°C, 48.5°C, and 60°C) were used for pulping during benchmarking trials. A lower temperature was selected for use in the protocol because it gives a higher pressure screening efficiency and represents a typical operating temperature. Pulping temperature can vary from one mill to another, depending on a panicular water treatment strategy including the extent of the water loop closure and the integration of the operation. The 14%consistency and 20-min pulping were based on visual observation and pulping energy measurements.

System Cleaning

For stock preparation, after each trial the system was thoroughly flushed with hot water, followed by scrubbing with clean copy paper. Copy paper was then pulped and passed through the system as an additional scrubbing. With a sample taken after sidehilling, 10 handsheets were made, dyed, and checked for residual dirt or adhesive particles. If dirt count exceeded 15 ppm, then an additional scrubbing with copy paper was made. Baskets from the pressure screen were manually cleaned of any adhering adhesive, If the adhesive was more than a trace, the removed adhesive was dried and weighed. The weight was included as part of the rejects for the respective stage. The baskets were then cleaned with a pressure washer plus a manual scrubbing with an organic solvent to remove all remaining traces of adhesive. For the paper machine, the clean copy paper stock from the final stock preparation system cleaning sequence was used to thoroughly clean the paper machine. Samples from the reel were checked for residual dirt. A dirt count of >15 ppm off the reel indicated that an additional cleaning sequence needed to be made. Clean copy paper was pulped and used for additional cleanings.

Testing

Each material was evaluated in two trials, one simulating post-consumer loading (no release liner present) and the other representing a pre-consumer loading (release liner included). During each trial, an accepts pulp sample was collected at each unit operation outflow. Handsheets made from these samples were dyed and read optically for din count, i.e., the adhesives content. Weight, consistency, and flow rate were measured and calculated as necessary to provide a mass balance and yield information for each trial. Energy input to the pulper was also collected for each trial and, if pulp was cleaned sufficiently; paper machine runs were made. A PIRA deposition apparatus was used to conduct tests on all final pulps. Biochemical oxygen demand (BOD) on the wastewater from washing and mineral metal analysis of the fresh water used and the waste water from washing were collected. Visual observation on the behavior of each experimental adhesive during recycling was noted at each stage. Equipment, especially the pressure screens, were visually observed after each run to examine if adhesive deposition was a problem or nor. Blank runs were made with clean pulp until the dirt count of the final pulp was <10 ppm.

DISCUSSION

The behavior of both control (standard) and one experimental adhesive is discussed and analyzed for each unit operation of the protocol. The results of the speckcheck analysis are shown in Table 1 and Figure 1.

Pulping and Screening

The objective of the pulping stage is to fiberize the paper and keep the contaminants large. Generally, this is made easier if the conditions are right to agglomerate adhesives without blinding the screen.

Preliminary trials indicated that a 20-min pulping time at pH 10 would be satisfactory for both the copy paper and the stamp face paper, A shorter time left “tags,” which are small pieces of paper. Longer pulping times tended to break down the contaminants into smaller particles plus waste energy and increase batch turnaround time. Energy measurements were made to make sure that 20 min was the optimum time needed to liberate the fibers and keep the contaminants large and screenable.

The separation of fibers from the contaminants is desirable for high recovery pulp yields. Having a few fibers attached to a contaminant may be desirable if their presence enhances the particles removal in a unit operation, such as screening where bulk is important. Their presence can be detrimental if the fibers shift the effective density towards 1.0, thereby nullifying the effectiveness of centrifugal cleaning. Entanglement of the attached fibers with free-flowing fibers in a forward cleaner will also impede the movement of the particles in the forward cleaner.

Table 1: Speckcheck analyals (ppm basis)

	Trial	Contaminant level (ppm)							
		Initial pulped slurry	0.30-mm pressure screen accepts	0.15-mm pressure screen accepts	Forward cleaner accepts	Through-flow cleaner accepts	LaMort flotation accepts	Sidehill washing accepts	Pressing/ shredding accepts
	Controls					-pass 1 -pass 2			
127	Copy paper only	1.21	3.17	2.91	0.86	1.85 2.12	1.34	5.82	9.78
128	Face paper only	2.09	1.66	2.01	1.31	2.06 1.27	2.11	1.59	2.17
107	Linerless stamp stock (43 °C)	913	54	9.4	6.4	10 1.6	2.1	0.6	5.2
106	Linerless stamp stock (48.5 °C)	1,506	63	21	31	52 6.7	2.4	2.7	1.6
110	Linerless stamp stock (60 °C)	1,734	338	101	49	26 23	12	18	10
109	Standard PSA stamp stock w/o release liner	2,093	601	42	31	20 21	13	10	19
108	Standard PSA stamp stockwith release liner	3,002	999	112	94	79 84	20	26	21
	Experimental adhesives								
131	Adhesive A w/o release liner	2,120	424	35.3	13.4	22.44 22.68	0.51	7.51	7.45
132	Adhesive A with release liner	2,622	85	41.6	24.3	57.93 13.13	13.99	11.84	12.24

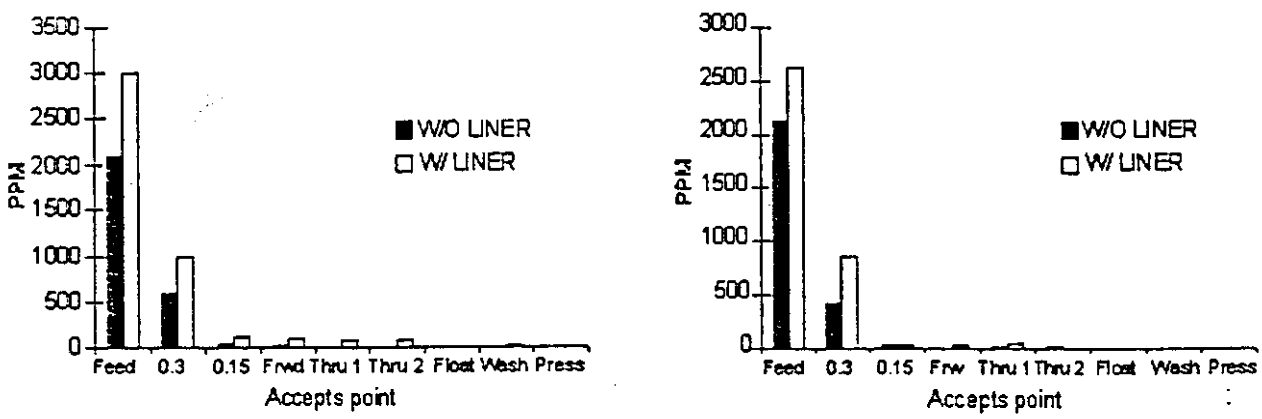

Fig. 1. Contaminant level via image analysis; (left) standard PSA construction, (tight) experimental adhesive A.

In this study, little evidence of fiber attachment was found on any adhesive. Adhesive particles rejected by the pressure screen and retained by the flatscreen all seemed to be free of fiber. The fiber observed through a 30x lighted magnifier appeared to be sandwiched between adhesive particles as they were wadded together during collection. The cleanliness of the adhesive seemed to depend on how thorough a wash the adhesive was given on the flatscreen. Tags were retained by the flatscreen and were prominent in the rejected adhesive. but individual fibers were generally not retained and relatively few showed up in the adhesive. Had the adhesive maintained the paper/adhesive bond during the pulping, there should have been adhesive particles that were virtually coated with fiber. This was not evident for any of the adhesives.

The presence of fibrous tags increased the dried weight of the collected adhesive, giving in some cases a false high value of rejects and a false sense of screening effectiveness. In the same manner, there are particles of adhesive that pass through the 0.2-mm flatscreen, an action that lowers the weight of the rejected adhesive, thereby decreasing the apparent screening efficiency. For these reasons, it was determined that the decrease in din count for the screening accepts relative to the screening feed is a better indicator of the true screening efficiency and yield in general.

For the preconsumer trials where release liner was included. the collected adhesive generally showed a smaller grain structure than for the postconsumer trials where the release liner was absent. As the paper disintegrated into fiber during pulping, if you assume that an adhesive in both cases is (1) released into the slurry having the same particulate size and form, then (2) proceeds to reagglomerate into larger particle groupings, the finer structure of the preconsumer adhesives would imply that one or more components of the release coating (e.g., silicon, clay) impedes the reagglomention. If this scenario is correct, the adhesive groupings should be three-dimensional, implying that for the same amount of adhesive, the summation of the cross-sectional areas of the larger groups should be less than the summation of the areas of the smaller groups. This implies that the din count ppm should be greater for the smaller particle grouping than for the larger particle grouping. That is, the handsheets from the pulper (before screening) for the trials with release liner should have a higher ppm than the same handsheets for the no-liner trials. This was the case for all but one adhesive used in this study.

Standard Adhesives

Control adhesives used for benchmarking are the linerless and the standard PSA stamp stock with and without liner. The linerless stamp stock results are shown in Table 1. Trial 107 (linerless at 43°C) shows a very high pressure screening efficiency. from initial dirt count of 913 ppm to about 10 ppm after two-stage screening, and a very clean final pulp of about 2 ppm after flotation. Results from standard PSA construction (trials 108 and 109) are also shown in Table 1. After two stages of pressure screening, the dirt count for the standard stock without liner (postconsumer simulation) went from initial 2,093 to 42 ppm and gave final pulp, after flotation, with 13 ppm. The trial with liner gave much lower pressure screening efficiency, apparently due to some material in the liner (clay, silicon), causing some pacification. One objective of this project was to evaluate the new experimental adhesives and determine if they screen better and are less problematic during processing. One experimental adhesive called adhesive A is analyzed in this paper.

Experimental Adhesive

Experimental adhesive A was primarily removed by the 0.30-mm screen with only a few fiber tags present. The adhesive dried to a gritty consistency, free of fiber except for the tags. Examination of the dried adhesive using a 30x magnifier showed the adhesive panicles to be glass-like beadlets. A similar examination for the rejects showed a similar appearance. Interestingly, the magnified view also revealed the presence of many tiny colored particles in the linered trial pulp. It was assumed chat these were ink particles, indicating the presence of recycled fiber in the base sheet of the release liner. These particles were small enough to have been washed through the slots of the flatscreen. That they were still present on the adhesive indicated an affinity for the ink by the adhesive.

Adhesive removal efficiencies more than 100%, based on weight of dried rejects from the flatscreen, are dubious. These negative removals reflect the presence of fibrous tags in the rejects. A removal efficiency based on residual adhesive in the accept streams as a ratio of the feed concentration given as ppm is a more reliable indicator. For this adhesive, the removal efficiencies using this technique were 80.0% and 98.330% for the 0.30 and 0.15 mm, respectfully, for the trial without liner, and 67.43% and 98.41% for the trial with liner. Figure 1 shows the dramatic decrease in adhesive contents of the accept streams. Actual image analysis values are presented in Table 1. The early removal of most adhesive, 90% to 95% via the screens, is important in that the smaller amount of residual adhesive left in the pulp has a higher probability of responding to the removal action of the subsequent unit operations.

The response of this adhesive is excellent, despite the lower initial removal in the 0.3-mm screen when the liner was included. Apparently the basic adhesive particle is fairly massive and rejectible by the 0.15-mm screen even if it does not reagglomerate after pulping.

Forward Cleaning

Adhesive A was apparently a denser adhesive (a "sinker") as it responded to the forward cleaning. Without liner, the pulp decreased its ppm count from 35.3 to 13.38 (62% removal), and with liner from 41.6 to 24.3 (42% removal). The moderate decrease in dirt count indicates a density of just over 1.0. Generally, there is about a doubling of the particle count for the linered trial versus the no-liner trial after pulping, indicating a retardation of the agglomeration of the adhesive particles in the linered pulp. For this adhesive, there was only a small dirt count increase (2120 to 2622 ppm). The retardation may be due to a component of the release liner. For this adhesive, with its larger particle size, there should be a much lower surface area to adhesive mass. If a coating of the particle is what retards the agglomeration, then clay may be the coating component. There might be sufficient clay in the copy paper to coat the large particles and any additional amount coming from the release coating would not have much of an effect; i.e., the particles would not agglomerate even if the release backing is excluded. The very gritty appearance of the adhesive for both trials would indicate this. For the forward cleaner, the small additional coating of clay may, for a nearly neutral density adhesive, be just enough to nearly neutralize the cleaner action.

Reverse Cleaning

Since adhesive A is apparently a denser adhesive, it should not respond to the action of the reverse cleaner. which is designed to remove low density particulates. The dirt counts for this trial reflect this anticipated result. For the linered pulp, the results were mixed but imply a possibly slight increase in dirt count. If the adhesive is indeed coated with clay, the slight shift in density could explain the cleaner action.

Flotation

Adhesive A seemed to respond to flotation in the trial without liner. decreasing the dirt count to 0.51 ppm. There was virtually no change in the trial with liner. Both results must be taken cautiously because with large particles the dirt count is accounted for by only a few panicles, and a numerical change of just a few particles can swing the dirt counts significantly. For the no-liner trial, this numerical change was from 43 to 3 particles, these being in the 0.02 to 0.03 mm^2 range. For the liner pulp, the change was from 25 to 11 particles in a size range up to 0.8 mm^2. These results imply that the coating would seem to be interfering with the flotation action.

Washing, Pressing, and Shredding

Washing removes very small particulates: clay, fiber fines, etc. Adhesive floaters tend to remain with the pulp thereby becoming more concentrated as material is washed away. Adhesive sinkers are remove only if present as very small particles. Typically, sidehill washing shows an increase in din count. Adhesive A seems to behave in this manner. increasing in concentration for the no-liner pulp and maintaining its level for the linered trial. Again, the few particles present can cause wide swings in the dirt count with a change in the number of only a few particles. Pressing and shredding of the cleaned pulp should have no effect on dirt count. For both trials the final pulps are about the same as the sidehill.

System Cleaning

Cleanup of the equipment was not a problem with this experimental adhesive. Adhesive residue on the tanks washed off easily with 85°C hot water. Dirt counts of the cleanup (flushing) pulp coming off the sidehill screen were about 3 ppn for both postconsumer and preconsumer trials.

CONCLUDING REMARKS

Adhesive A appears to be a desirable adhesive for recycling. The majority of the particles were removed by the 0.3-mm slots. with virtually all of the rest being rejected by the 0.15-mm slots. The basic particle seems to be very large such that the normally encountered failure to agglomerate in the presence of the release liner is no problem. If there is a problem, it is the density apparently being too close to 1.0 such that the cleaners are neutralized and the flotation is too sensitive to slight shifts in density as a result of possible coating action of contaminants. A slight

shift to a lighter density might be recommended, providing the change does not affect the size distribution of the particles

We recommend the following to change the protocol:

1. Change fine screening basket from 0.15 to 0.1-mm (0.006-in cut to 0.004-in. cut) screen to improve screening efficiency and better simulate typical recycling operation.
2. Add second stage of forward cleaning to better remove high density PSAs and be similar to the two stage reverse cleaning for low density removal.
3. Examine and change surfactant addition point
4. Improve yield by
   - Passing pressure screen rejects through a 0.1-mm (4 cut) flat screen with the accept from flat screen recycled back to the primary system,
   - Installing secondary fiber recovery system for forward cleaners, and
   - Changing sidehill screen size from 70 to 100 mesh.

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