Compatibility of Pressure Sensitive Adhesives With Recycling Unit Operations

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Removal of pressure sensitive adhesives (PSAs) from recovered paper is a major problem facing the paper recycling industry. As a result of a United States Postal Service (USPS) initiative, which currently purchases about 12% of domestic PSA production, a team was formed consisting of representatives from the USPS, the Forest Products Laboratory, Springborn Testing and Research, paper recovery companies, paper recyclers, adhesive manufacturers, and chemical suppliers. This large team of cooperators has the common goal of formulating PSAs that can be efficiently removed during the fiber screening operations. To evaluate new PSAs, an initial pilot-scale separation sequence and an operating protocol were developed. This fiber sequence involved high-consistency pulping, pressure screening, forward cleaning, reverse cleaning, flotation and washing. The sampling protocol allowed the evaluation of the PSA removal efficiency of each of the unit operations, An initial trial of 14 adhesives has been completed. These data indicate that some PSA formulations can be easily removed during fiber screening, but others are only poorly removed.

INTRODUCTION

Since 1994, Springborn Testing and Research (STR) and the Forest Products Laboratory (FPL) have been working with the United States Postal Service (USPS) to evaluate newly developed pressure sensitive adhesives (PSAs) for postage stamp applications that do not adversely affect the environment. To successfully achieve this objective, an understanding is required of two areas: (1) postage stamp requirements including performance, production, mail processing, and philatelic and (2) paper recycling issues as they relate to adhesive contaminate removal.

Upon consultation with the paper and adhesive industries, it was concluded that although several adhesive manufacturers were actively working on developing recyclable adhesives, no universally accepted recycling test method or protocol is available.

In 1995, the USPS sponsored a conference [1] during which the adhesive industry was invited to participate in the USPS Environmentally Benign Pressure Sensitive Adhesives for Postage Stamp Applications program. Thirteen companies participated by submitting adhesive samples. To determine whether an adhesive was recyclable, a protocol needed to be developed. The results of this work led to a three-tier evaluation process with the development of laboratory, pilot-scale and mill-scale recycling protocols. The laboratory- and pilot-scale protocols were developed and implemented at STR and the FPL, respectively. Both the laboratory- and pilot-scale protocols will be incorporated into a new USPS recyclable PSA stamp

specification. All adhesive samples participating in the program that successfully meet all the qualification requirements of the specification, including the newly developed recycling protocols, will be placed on the USPS qualified PSA stamp products list. The development of the FPL pilot testing protocols is described in this report.

PILOT TESTING PROTOCOL

Removing contaminants from recovered paper pulps is one of the biggest technical barriers to paper recycling [2]. Contaminants are undesirable components that come from pitch, ink, plastic films, converting aids, paper coatings, and adhesives. Adhesives are either hot melt or pressure sensitive. Pressure sensitive adhesives are applied to labels, tapes, and some postal materials. 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 processing recovered paper and papermaking. Furthermore, closing the water loop by recycling water within mills and shifting to alkaline-based papermaking make the stickies problem worse.

The pilot testing protocol was developed as a joint effort by the USPS, FPL, STR, and various industries. The unit operations were high consistency pulping (14% consistency, 43°C, 20 min.), two-stage (0.3- and 0.15mm) slotted screening, forward cleaning, two-stage reverse cleaning, flotation, washing, and pressing. A final pulp with a dirt count of 25 parts per million (ppm) or less was set aside for a experimental papermachine run. From those pulps, having a dirt count

>25 ppm, a sample was subjected to the additional cleaning operations of dispersion, flotation, and washing to determine whether the residual adhesive would break down into smaller pieces and be removed by the additional flotation stage.

Stamp stocks containing 14 experimental adhesives were evaluated, with each trial using 90% copy paper plus 10% stamp stock. For the preconsumer trials, the 10% stamp stock was weighed out as received, including the release liner. For the postconsumer trial, the same amount of stamp stock was weighed out but the release liner was removed, the stamp stock was affixed to copy paper, and an extra amount of copy paper equal to the weight of the release liner was added. The 5% stamp stock level represents a significantly higher loading than a typical recovered paper stock. A stamp on a First Class letter with one sheet of paper represents approximately 0.5% by weight.

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.

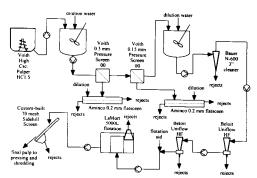


Figure 1. Flow diagram for the experimental system.

- 0% copy paper plus 10% stamp face paper to simulate the best cleaning level without adhesive.
- 990% 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

A flow diagram for the experimental system is shown in Figure 1. For clarity, intermediate holding tanks were omitted from this diagram. A process temperature of 43°C was used for pulping during the trials. Although temperatures of 30°C, 43°C, 49°C, and 60°C were tested, the moderate temperature was chosen as a balance between screening efficiency and ease of cleaning the equipment. At lower temperatures, many of the PSAs were more tacky, while at higher temperatures the screening efficiency decreased. 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 pulping and processing of clean copy paper. With a sample taken after sidehill washing, 10 handsheets were made, dyed, and checked for residual dirt or adhesive particles. If dirt count exceeded 15 ppm, then an additional run of 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 and washed with mineral spirits 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.

Testing

Each material was evaluated in two trials, one simulating postconsumer 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 according to TAPPI T-205 om-88, except that only one 2-min. pressing was done. After drying, the handsheets were dyed with Morplas blue dye in heptane and scanned on an Optomax Speckcheck Dirt Counter to determine the level of contaminants [3]. 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 had a low contaminant level, paper machine runs were made. 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 inspected after each run to determine adhesive deposition. Blank runs were made with clean pulp until the dirt count of the final pulp was < 10 ppm.

DISCUSSION

The behavior of both controls (linerless stamp stock and standard PSA stamp stock) and 14 experimental adhesives was studied and analyzed for each unit operation of the protocol. The controls and two experimental adhesives were chosen to represent the range of responses of the other 12 to the processing conditions and are discussed in this report. Both experimental adhesives are acrylic-based formulations.

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, while maintaining efficient separation of fiber from contaminants. Having a few fibers attached to a contaminant may be desirable if their presence enhances the particle removal in screening where size is important.

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 tree of fiber. Tags were retained by the flatscreen and were prominent in the rejected adhesive, but individual fibers were generally not retained. Had the adhesive remained bonded to the paper 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. Furthermore, there were particles of adhesive that passed through the 0.2-mm flatscreen, thereby decreasing the apparent screening efficiency. For these reasons, it was determined that the decrease in dirt count for the screening accepts relative to the screening feed is the best indicator of the screening efficiency.

For the preconsumer trials where release liner was included, the collected adhesive generally showed a smaller grain structure than for the postconsumer trials, when the release liner was absent. As the paper disintegrated into fiber during pulping it is likely that, in both cases, the adhesive was released into the slurry with the same particle size, and then proceeded to reagglomerate into larger particles. The finer structure of the preconsumer adhesives would imply that one or more components of the release coating (e.g., silicon, clay) impedes the reagglomeration. Unfortunately, the dirt count ppm depends on size distribution. If the particles are smaller, they show a higher dirt count than an equivalent weight loading of larger particles.

Standard Adhesives

Control adhesives were the linerless and the standard PSA stamp stock with and without liner. The linerless stamp stock results are shown in Table 1. Trial 107 shows a very high pressure screening efficiency (Table 2). The initial dirt count of 913 ppm was decreased to about 10 ppm after two stages of screening and to about 2 ppm after flotation. The high screening efficiency was likely due to the formation of relatively large adhesive particles during pulping. 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

Table 1. Speckcheck analysis (ppm basis)

	Contaminant level (ppm)								
Tria	ı	Initial pulped slurry	0.30-mm pressure screen accepts	0.15-mm pressure screen accepts	Forward cleaner accepts	Through- flow cleaner accepts -pass 1 -pass 2	LaMort flotation accepts	Sidehill washing accepts	Pressing/ shreadding accepts
	Controls								
127	Copy paper only	- 1	3	3	1	2			
						2	1	6	10
128	Face paper only	2	2	2	1	2			
						ı	2	2	2
107	Linerless stamp stock	913	54	9	6	10			
400	0: 1 1 704 1					2	2	1	5
109	Standard PSA stamp stock w/o release liner	2,093	601	42	31	20			
108						21	13	10	19
100	with release liner	3,002	999	112	94	79 84	20	26	21
	Experimental adhesives								
131	Adhesive A	2,120	424	35	13	22			
	w/o release liner	2,120	72.7	33	13	23	1	8	7
132	Adhesive A	2.622	854	42	24	58	•	ū	
	with release liner					13	14	12	12
133	Adhesive B	6968	3776	1082	1039	470			
	w/o release liner					289	229	354	260
134	Adhesive B	7128	5226	1169	1182	747			
	with release liner					686	512	607	478

standard stock without liner (postconsumer simulation) went from initial 2,093 to 42 ppm and gave final pulp after flotation, with 13 ppm, as shown in Figure 2. The trial with liner gave much lower overall pressure screening efficiency, as shown in Table 2, apparently due to some material in the liner (clay, silicon), causing some pacification.

Experimental Adhesives

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, tree of fiber except for the tags. Examination of the dried adhesive using a 30x magnifier showed the adhesive particles to be glass-like beads.

For adhesive A, the removal efficiencies were 80% and 92% for the 0.30 and 0.15 mm, respectively, for the trial without liner, and 67% and 95% for the trial with liner. Figure 3 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, 98%, at the screens, is important in that the smaller amount of residual adhesive has a higher probability of being removed in subsequent unit operations. The response of this adhesive is excellent, despite the lower initial removal in the 0.3-mm screen when the liner. Apparently the adhesive particles are fairly large and rejected by the 0.15-mm screen even if it does not reagglomerate after pulping.

When compared with adhesive A, adhesive B was much less screenable. The cleaning efficiencies were 46% and 71% for the two screening operations (Table 2). The ineffective screening is largely due to the particle size and the lack of tackiness of adhesive B. It broke in to very small platelets during pulping and did not reagglomerate. This behavior is illustrated in Figure 4. The initial dirt count is much higher due to the smaller particle size, and there is still approximately 1000 ppm of adhesive in the pulp after screening.

Table 2. Cleaning efficiencies

	Trial	0.30-mm pressure screen accepts	pressure screen	Forward cleaner accepts	Through flow cleaner accepts
107	Linerless stamp stock	94%	83%	33%	0%
109	Standard PSA stamp stock w/o release liner	71%	93%	26%	35%
108	Standard PSA stamp stock with release liner	67%	89%	16%	16%
131	Adhesive A w/o release liner	80%	92%	63%	0%
132	Adhesive A with release liner	67%	95%	43%	0%
133	Adhesive B w/o release liner	46%	71%	4%	55%
134	Adhesive B with release liner	27%	78%	0%	37%

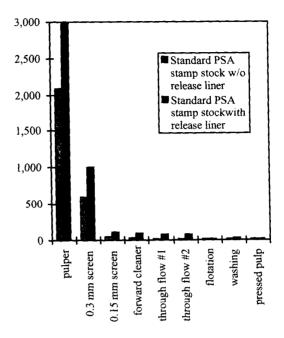


Figure 2. Contaminant level via image analysis standard PSA construction.

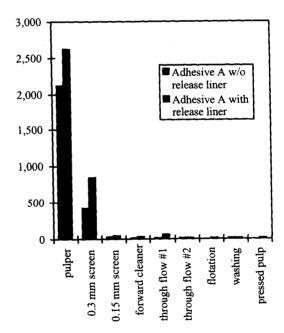


Figure 3. Contaminant level via image analysis experimental adhesive A.

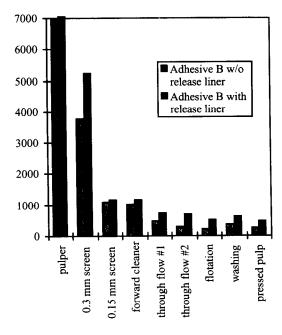


Figure 4. Contaminant level via image analysis experimental adhesive B.

Forward Cleaning

Adhesive A was apparently denser than the pulp slurry because it responded to the forward cleaning, which is designed to remove higher density particles. Without liner, the pulp ppm count decreased from 35 to 13 (63% removal), and with liner from 42 to 24 (42% removal), as shown in Tables 1 and 2. Adhesive B, however, appears to be nearly the same density as the pulp slurry since the dirt count did not change significantly during forward cleaning.

Reverse Cleaning

Since adhesive A was apparently a denser adhesive, it did not respond to the action of the reverse cleaner, which is designed to remove low density particles. Adhesive B was removed selectively during the first flow-through stage. Without liner, the pulp ppm count decreased from 1039 to 470 (55% removal), and with liner from 1182 to 747 (37% removal), as shown in Tables 1 and 2. The reduced performance in the presence of liner may indicate that the particles were coated with a denser material.

Flotation

Adhesive A seemed to respond to flotation in the trial without liner, decreasing the dirt count to 1 ppm. There was virtually no change in the trial with liner. This seems to indicate a change in the surface chemistry when the release agents of the liner are present in the pulp. Both results must be taken cautiously because the sampling error at these low dirt count levels is significant. For the without-liner trial, this numerical change was from 43 to 3 particles, these being in the 0.02 to 0.8 mm² range. For the with-liner pulp, the change was from 25 to 11 particles. These results imply that the coating would seem to be interfering with the flotation action. The particles from adhesive B did not seem to be surface active in flotation and were not selectively removed during flotation.

Washing and Final Processing

Washing removes very small particles: clay, fiber fines, etc. Adhesives tend to remain with the pulp thereby becoming more concentrated as material is washed away. Typically, sidehill washing shows an increase in dirt count. Adhesives A and B seemed to behave in this manner. After the final unit operation, the dry mass of the pulps were measured. Typical yields for these trials were between 30% and 40%.

System Cleaning

Cleanup of the equipment was not a problem with experimental adhesive A. 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 ppm for both postconsumer and preconsumer trials. System cleaning after processing pulp containing adhesive B was problematic. In all the equipment, there was residue that had to be removed with mineral spirits. And several runs were required to bring the dirt count below the required limit.

System Improvements

Finally, since the completion of this trial, the pilot equipment and process has been redesigned to improve the efficiency and yield. A 0.1-mm pressure screen basket was substituted for the 0.15-mm basket and a second stage of forward cleaning was added. Also, fiber recovery systems for the pressure screens and forward cleaners were installed. Initial trials with this new system indicate similar overall cleaning efficiencies with yields between 75% and 85%. This new system will be used to evaluate commercially prepared PSA stamp stocks.

CONCLUDING REMARKS

The ordering of the recyclablity of the four adhesives described in this report is linerless stamp stock, adhesive A, standard PSA stamp stock, and adhesive B. The other 14 experimental adhesives studied responded in the range spanned by linerless stamp stock and adhesive B.

Linerless stamp stock appears to be the most desirable due to its ability to agglomerate during pulping. The formation of large non-tacky particles allows for efficient screening.

Adhesive A appears to also be a desirable adhesive for recycling. Although it did not form as large of particles as the linerless stamp stock, the majority of the particles were removed by the 0.3-mm slots, with virtually all the rest being rejected by the 0.15-mm slots. The initial particle seems to be large enough that the failure to agglomerate in the presence of the release liner is no problem.

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LITERATURE CITED

- Fatah, A.A., and J.Y. Peng, "Environmental Objectives of the Stamp Program," in Proceedings from joint conference, "Environmentally Benign Pressure Sensitive Adhesives for Postal Applications," Windsor Locks, CT (February 14, 1995).
- AbuBakr, S., "Recycling: A TAPPI Press Anthology of Published Papers 1992-1997," TAPPI PRESS, Atlanta, GA (1997).
- Sutherland, N.R., "Stickies Dirt Count and Laboratory Methods," in "Proceedings, "Environmentally Benign Pressure Sensitive Adhesives for Postal Applications," Washington DC (July 7-9, 1997).