Can Laboratory and Pilot Recycling Trials Predict Adhesive Removal in Commercial Recycling Systems? Results from the USPS Environmentally Benign Stamp Project.

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Abstract

The ultimate goal of the US Postal Service (USPS) Environmentally Benign Stamp Program is to develop stamp laminates, i.e., face paper, adhesive and siliconized liner, that do not cause difficulties in recycling mills. The criterion for success, and the USPS definition of benignity. is the avoidance of process and product quality hardships when such PSA laminates are introduced in significant quantities. However, since it is neither prudent nor cost-effective to test experimental adhesive materials at mill scale, we have developed laboratory scale (360 g pulp) and pilot-scale (112 kg pulp) test methods for determining adhesive performance in recycling environments. Comparison of results from these small-scale trials with mill trial results has shown that there are strong correlations. Furthermore, specifications for environmentally benign stamp adhesives, based on laboratory and pilot results, have been set and accepted by the recycling industry.

Introduction

Before initiation of the USPS Environmentally Benign Stamp Program, no widely accepted test methods existed for determining the compatibility of pressure sensitive adhesives (PSAs) with paper recycling processes. Thus, there was no unified guidance for adhesive formulators in their efforts to develop new products in response to environmental concerns. The current test methods are the product of several years of testing experimental . adhesives submitted by formulators.

During the development of these test methods, several recycling mill representatives gave suggestions for operating parameters and process sequence, and advised adhesive suppliers to formulate their products to be removed at the earliest stages of the recycling process. The USPS drafted test methods detailing process configurations and operating parameters that included conventional recycling process technology. This proposal was reviewed and approved by our industrial cooperators. Although the results from laboratory and pilot-scale experiments must be interpreted using extrapolations and approximations, the participating mill operators have indicated these data allow them to estimate the performance of an adhesive in their mill. Furthermore, experiments conducted using these test methods do provide a uniform basis of comparing different adhesive formulations.

After developing and testing the test methods, many adhesives were evaluated on both laboratory and pilot-scales. These experimental adhesives included a variety of formulations including both acrylic and rubber-based materials. This test set of adhesives provided a range of recycling performance, i.e., some were easily screened from pulp while others were extremely difficult to remove. Thirteen adhesives were ultimately submitted for full mill trials at one of six different US recycling facilities. Since the various recycling mills have process configurations that differed from each other, a single benchmark adhesive material was trialed at each facility before the experimental adhesives were tested. Benchmark trial results provided a basis for normalizing the experimental adhesive results from the six different facilities to a common basis. A total of 20 mill trials were conducted between August 10, 1999 and June 26, 2000.

From the database of laboratory, pilot and mill results, the correlation between results from tests on the various size-scales can be assessed. Comparing and contrasting these results will be the subject of the balance of this report.

Laboratory-Scale Test Method

The USPS laboratory-scale test method[1] consists of 4 major unit operations and two dewatering operations. The first operation is high-consistency, 15 %, pulping of 360 g OD pulp which contains 1% adhesive by weight. After 0.3 and 0.15 mm slotted screening with a Valley Flat Screen, the pulp slurry is further cleaned by flotation

[1]C. Houtman, David Bormett, Nancy Ross Sutherland, and Donald Donermeyer, "Development of the USPS Laboratory and Pilot Testing Protocols", Proceedings to the 2000 TAPPI Recycling Symposium held in Washington DC, March 2000, page 403.

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in a Denver Flotation device. After each unit operation, the pulp was dewatered with a 200 mesh screen. Four pulp samples are taken: after pulping, after 0.3 mm screening, after 0.15 mm screening, and after flotation. These samples are analyzed for residua1 adhesive levels using the USPS image analysis test method. The two screening and flotation reject samples are dried and weighed. Visual inspection is used to estimate the fraction of fiber in these samples, which allows for the estimate of the mass of adhesive removed in each unit operation.

Pilot-Scale Test Method

The USPS pilot plant test method[1] provides six major unit operations: pulping, slotted screening, forward cleaning, flow-through cleaning, flotation and washing.

Although adhesive removal efficiencies from experiments using flat screens can be indicative of product performance, experiments with pressure screens reflect mill practices more accurately. Furthermore, since slotted screening will be essential to removing most adhesive formulations, the pilot test method was developed around commercial pressure screens. The sizes and flowrates in the test method were set to give approximately 1 hour of screen operation. The screen sizes, consistencies and passing speeds are shown in Table 1. Typical pressure screen passing speeds in recycling mills are 0.6 to 2 m/s. Unfortunately, due to down-stream limitations, the passing speeds for the pilot plant trials were 0.35-0.4 m/s. Lower passing speeds are often associated with higher screening efficiencies, at a cost of fiber fractionation.

Table 1: Pressure Screen Operating Parameters

Slot Size	Inlet Csc.	Open Area	Passing Speed
0.3 mm milled	1.10%	4.3 %	0.41 m/s
0.1 mm profile	0.94%	2.3%	0.35 m/s

The total fiber yield was 67%, which is on the low side of typical mill-scale operation. Significant yield improvements could be achieved by adding secondary pressure screens to the system, which is planned for the near future.

During a trial temperatures and flowrates are recorded using a computer data acquisition system. These data are used to monitor the operating parameters. Consistencies are also determined on all the process streams. The physical properties of adhesives are very temperature sensitive. Thus, temperature must be considered an important operating parameter. The design of the pilot plant system includes temperature controllers on all process water streams. Analysis of data shows that temperature is quite repeatable. The pulping temperatures are all between 46-48 °C.

Mill-Scale Test Method

Six US paper mills (P.H. Glatfelter Company, International Paper-Franklin VA, International Paper-Hudson River NY, Mississippi River Corporation, Westvaco Corporation-Tyrone PA, and American Fiber Resources L.P.) agreed to host trials with experimental adhesives. Each facility produces pulp that is suitable for printing and writing grades. and the production rates ranged from 40 to 600 tons per day. Mill trials were conducted as double spikes, typically one trial each day for two days. The mill process was sampled to determine the background contamination, and then adhesive material was added continuously for approximately 2 hours. The amount of adhesive added varied from site-to-site but was in the range of 0.05 to 0.2%. The level of the adhesive loading was set so that the spike was statistically significant with respect to background contamination. Process samples were taken to follow the passage of adhesive through the system. Although there were significant differences in process design and operation, all six mills included pulping, pressure screening and final pulp dewatering, so samples were taken after each of these unit operations. These samples were analyzed both by the mill's QA/QC method and the USPS image analysis test method. Table 2 shows a summary of the processes on the three size-scales.

Laboratory 360 g. per mal	Pilot 112 kg. per trial	Mill 40-600 tons/day
15% csc. pulping	12.5% csc pulping	HC pulping other unit ops.
0.3 mm flat screen dewatering	0.3 mm pressure screen	
0.15 mm flat screen dewatering	0.I mm pressure screen forward cleaners flow-through cleaners	pressure screen other unit ops.
flotation dewatering	flotation dewatering	dewatering

Image Analysis Test Method

PSA particles are often white and have low contrast with the background fibers. To quantify the PSA concentration in handsheets made from pulp samples the particles were dyed to increase the contrast with the background fibers. For samples from these trials, handsheets were dipped in a solution of a hydrophobic dye. The dye associates strongly with any hydrophobic particles in the sample. Since PSAs are often hydrophobic, they tend to retain dye while cellulose fibers, which are hydrophilic, do not. The result of washing with methanol is a handsheet with dark blue PSA panicles and a white or lightly tinted background. Once the PSA particles are made visible by dyeing, the amount of adhesive is quantified by using a flatbed scanner and image analysis software.'

Data Analysis

Inspection of Table 2 shows that laboratory, pilot and mill-scale trials all included pulping, screening. and final dewatering. Samples were taken after each of these unit operations. These pulp samples were tested using the USPS image analysis test method. From this analysis, six results were calculated: pulper PPM, pulper particle size, screen PPM, PPM fraction remaining after screening, final pulp PPM, PPM fraction remaining in final pulp. Fraction remaining is defined as FR = (PPM after)/(PPM before). This quantity is related to efficiency by the formula, Efficiency = (1-FR)x100

Since the mill trials each had different process configurations and adhesive loadings, the data need to be normalized before results could be compared to the lab and pilot data. The benchmark adhesive trial results, at a particular mill site, were used to adjust the candidate mal results. The steps in the data analysis can be summarized as follows:

(1) Use the results of the benchmark trials at the mills to calculate a ratio for each candidate, ratio = (candidate results)/(benchmark results). These ratios were adjusted to account for differences in adhesive loadings between candidate and benchmark trials.

(2) Use the results of benchmark trials at the laboratory and pilot-scale to calculate similar ratios for these two testing venues

(3) Conduct statistical analysis on the calculated ratios

Confirmation of this method of correction was obtained by comparing the rank of one adhesive, Acrylic 1, which was submitted for trials at two different mill sites. The results are shown in Table 3. Although, these two mill sites had rather different process designs, the results, after correction, were very similar. Except for pulper PPM, adhesive Acrylic 1 is placed at the same rank relative to the other adhesive by the results from both mill sites.

Table 3: Rank of Acrylic 1 at two different mill sites

Parameter	Mill B	Mill E	B-E
Pulper PPM	12	5	7
Pulper Size	7	6	1
Screen PPM	10	9	1
Screen FR	3	5	-2
Final PPM	11	11	0
Final FR	3	3	0

Results

Correlation Analysis

Spearman Rank-Order Analysis[2] allows one to statistically determine the probability that two data sets are correlated. It is similar to the correlation coefficient used with linear regressions, but it is generally-considered more robust with respect to data distributions. The method involves sorting the results from lowest to highest and then assigning each the integer corresponding to its position in the list. After ranking the values for the two data sets of interest, the probability of correlation is calculated using standard methods.[2] Table 4 shows the results of the Spearman Rank-Order analysis for pilot versus mill results.

Table 4: Pilot and Mill results correlations

Pilot vs. Mill Correlation			
	Spearman r	t	significance
Pulper PPM	0.489	1.604	91%
Pulper Size	0.643	2.092	98%
Scrn PPM	0.549	1.797	95%
Scrn FR	0.308	1.016	69%
Final PPM	0.511	1.675	94%
Final FR	0.407	1.338	83%

The "significance" column in Table 4 can be interpreted as the probability that a correlation exists between the datasets. Pulper particle size stands out as highly correlated. In fact, subsequent linear regression analysis showed that although particles sizes generated for a specific adhesive are not the same in the pilot and mill systems, the results from the one can be used to predict the size in the other.

Inspection of Table 4 indicates that since there is a high probability of correlation for the pilot and mill data, the adhesives were generally ranked in the same order by both pilot and mill systems. This observation is relevant, particularly in light of the stated intentions of the program. The USPS desires PSAs that repulp to sufficiently large particles so they can be screenable in commercial facilities. Clearly, the 98% significance attributed to pilot prediction of mill pulper particle size confirms that pilot pulper successfully reflects likely particle sizes when these PSAs are introduced into commercial systems. Thus, the USPS objective of large particle size contaminants is ensured if PSAs form large particles in the pilot system. Similar significance is noted of the laboratory pulper particle size, when correlated through rank analysis to the mill results.

Identification of Exceptions

The goal of this work is to identify adhesives that may cause operational problems during paper recycling and thus, the correlations between laboratory, pilot and mill results were tested by identifying the adhesives that performed poorly. Since there was no definition of poor performance before conducting these trials, the database was used to determine those adhesives that behaved significantly worse than the other adhesives.

[2] W.H. Press, S.A. Teukolsky, W.T. Vetterling, and B.P. Flannery, Numerical Recipes in C: The Art of Scientific Computing, Second Ed., Cambridge University Press, Cambridge UK, 1992, page 639.

Probability Plotting is commonly employed to test the goodness of fit of data to an appropriate model.[3,4] Specifically, the data were tested for normality. Using an iterative procedure, the maximum number of experimental PSAs that can be included before a data population becomes non-normal was determined. The primary intent in using this technique is to compare exceptions, those candidates that fall outside the normal distribution of data. In summary, if experimental adhesive X proves to be an outlier with respect to final PPM as determined by laboratory, pilot, and mill trials, then we have demonstrated that it is possible to predict mill trial results from laboratory and pilot trial results.

Table 5 shows a summary of results of a probability plot analysis of the laboratory, pilot and mill dataset. Inspection of Table 5 indicates that when adhesives exhibited poor performance throughout most of the mill process the pilot plant also identified them as poor performance exceptions, as shown by adhesives 9 and 10. Adhesive 10 was identified as a poor performance exception by the laboratory as well. There is also significant agreement between the pilot and mill data with respect to the adhesives that did not cause a problem in the mill.

The results in Table 5 are segregated by base polymer chemistry, acrylic and rubber. Although one rubber-based adhesive was identified as an exception in the mill trials, in general, they presented fewer problems than the acrylic-based materials.

Table 5: Poor performance exceptions as identified by results from laboratory, pilot and mill mal results.

Adhesive	Pulper PPM	Pulper Size	Scrn PPM	Scrn FR	Final PPM	Final FR
Acrylic 1					M	PM
Acrylic 2	P					
Acrylic 3	P		P			
Acrylic 4					P	P
Acrylic 5	P				P	
Acrylic 6					L	
Acrylic 8				M		M
Acrylic 9			PM	PM	PM	PM
Acrylic 10		LPM	LPM	LPM	LPM	LPM
Rubber 1					M	M
Rubber 2				P		LP
Rubber 3	M		PM			
Rubber 4						L

L = Poor performance exception in the laboratory
P = Poor performance exception in the pilot plant
M = Poor performance exception in a mill

The exception analysis shown in Table 5 can be summarized by counting the number of times the laboratory or pilot agreed with the mill in terms of poor performance exceptions. Table 6 shows the percentage of agreement between trials.

Table 6: Agreement in identification of poor performance exceptions

	Agreement	Random
Lab predicts pilot	30%	10%
Lab predicts mill	29%	10%
Pilot predicts mill	65%	26%

[3]B.F. Kimball, "On the Choice of Plotting Positions on Probability Paper", *Journal of the American Statistical Association*, Sept. 1960, page 546.
[4]G.E.P Box, W.G. Hunter, and J.S. Hunter, Statistics for Experimenters: Introduction to Design, Data Analysis, and Model Building, John Wiley and Sons, NY, NY, 1978, page 329.

Conclusions

A series of adhesives were tested in the laboratory, pilot plant, and mill. Analysis shows that there are strong correlations between the pilot and mill results. Two adhesives that caused significant problems in the mill were also identified during the pilot testing. The agreement between the pilot and the mill results was nearly 70%.

The analysis suggests that the laboratory and pilot data are sufficient to identify the adhesives that are likely to present an operational problem at a mill. Recently, with support of representatives from the host mills, the United States Postal Service has added limits on laboratory and pilot recycling results to its specifications for stamp adhesives?

Future Efforts

From this research, several factors were identified that may have reduced the correlation of the laboratory and pilot results with the mill results. These factors include: (1) the use of only clean water for dilution, (2) a screening system with low passing velocities, (3) clean feed stock, and (4) thoroughly cleaning the system between trials. Current efforts are directed towards modification of the laboratory and pilot systems to better reflect mill operation. For example, water clarifiers are currently being tested with both systems. These modifications will allow water closure, which is more representative of commercial mill operation:

Acknowledgements

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[5] USPS specification 1238F, http://stamps.secdog.com/