Validation of Laboratory-Scale Recycling Test Method of Paper PSA Label Products

Carl Houtman and Karen Scallon
USDA Forest Service, Forest Products Laboratory

Richard Oldack
Dyna-Tech Adhesives, Inc.

ABSTRACT
Starting with test methods and a specification developed by the U.S. Postal Service (USPS) Environmentally Benign Pressure Sensitive Adhesive Postage Stamp Program, a laboratory-scale test method and a specification were developed and validated for pressure-sensitive adhesive labels, By comparing results from this new test method and pilot-scale tests, which have been previously validated with mill trials, we showed that the new test could be used to qualify label products as recycling-compatible

KEYWORDS
ADHESIVE, IMAGE ANALYSIS, HOT MELT ADHESIVE, PRESSURE SENSITIVE ADHESIVE (PSA), RECYCLE COMPATIBLE ADHESIVE (RCA), RECYCLING, SCREENING

INTRODUCTION

The U.S. Postal Service (USPS) Environmentally Benign Pressure Sensitive Adhesive Postage Stamp Program resulted in the qualification of 12 pressure-sensitive adhesives (PSAs) as environmentally benign. This effort utilized a vertical team consisting of representatives from the paper, recycled pulp, adhesive, testing, printing, and converting industries and the USPS. During the 7-year span of the USPS effort, laboratory- (360 g OD pulp), pilot- (112 kg OD pulp), and mill-scale (40–600 ton OD pulp/day) test methods were developed and applied to 15 prospective adhesives [1,2]. Results of mill-scale testing of these adhesives suggested two caused either runability or product quality difficulties. Specifications for the pulper particle area and final parts per million (PPM) values for pilot-scale test results were set to exclude the two adhesives that caused problems for the mills. Upon review of the data, representatives of six paper companies endorsed the proposed limits and agreed that pilot-scale data were likely sufficient to predict if an adhesive will cause problems in a mill. These data and endorsement were used as the basis for USPS P-1238-F specification for pressure-sensitive postage stamps. Since that time, all USPS postage stamps have been made with RCAs, and the lab and pilot-scale test methods developed by the USPS have become generally accepted as valid.

Concurrently, the President of the United States endorsed the utilization of qualified adhesives throughout the Federal Government by issuing Executive Order 131 48, Greening the Government Through Leadership in Environmental Management [3]. Specifically, E.O. 131 48 Section 702 states,

Section 702 Environmentally Benign Adhesives. Within 12 months after environmentally benign pressure sensitive adhesives for paper products become commercially available, each agency shall revise its specifications for paper products using adhesives and direct the purchase of paper products using those adhesives, whenever technically practicable and cost effective. Each agency should consider products using the environmentally benign pressure sensitive adhesives approved by the U.S. Postal Service (USPS) and listed on the USPS Qualified Products List for pressure sensitive recyclable adhesives.

In 2003, an independent industry consortium, Initiative to Promote Environmentally Benign Adhesives (IPEBA), followed the lead of the USPS to develop test methods and a specification for PSA paper labels. In February 2005, IPEBA dissolved after having accomplished its goal and became a subcommittee of the Tag and Label Manufacturers Institute's (TLMI) Environmental Committee. Furthermore, TLMI agreed to act as custodian for these test methods [4].

Similarly, an industry association International Association of the Deinking Industry (INGEDE) also has developed a method for determining the fragmentation and subsequent screenablity of adhesive formulations, INGEDE Method 12 [5]. Method 12 follows a similar process scheme to the USPS test methods, except that reject retained on the flat screen are analyzed and results are expressed as a percentage of particles that have a projected area greater than 2 mm^2. By testing a range of adhesive materials using both the USPS test methods and INGEDE Method 12 [6], a strong correlation was observed. Materials that pass the USPS test methods also produce a large number of adhesive fragments larger than 2 mm^2.

Although USPS coined the term "environmentally benign" to describe the adhesives developed under their program, one can question the validity of this broad claim. Environmentally benign likely implies no negative environmental impacts throughout the life cycle of the material. Clearly this claim was not in the scope of the USPS project or the current effort; thus a more accurate term would be "recycling-compatible adhesive" (RCA), which will be used throughout the remainder of this paper.

MOTIVATION

Currently, the USPS switch to RCAs and implementation of Executive Order 131 48, Sec. 702, is slowly shifting the types of adhesives arriving at recycling mills in their recovered paper. However, noticeable reductions of the "stickies" problem in paper mills will not be achieved until RCAs are utilized in the vast majority of pressure-sensitive paper-to-paper labeling applications, in both the governmental and private sectors.

Thus, a simple, cost-effective, and reliable lab-scale method is desired for testing adhesive behavior during paper recycling. Previous work indicated that pilot-scale testing was sufficient to predict whether or not an adhesive material would cause operational or product-quality problems for recycling mills. Ideally, similar mill validation efforts should be used to support this new method. Unfortunately, the time, expense, and effort are not available. Thus, we will use previous correlations between pilot and mill to predict mill results from pilot-scale tests alone. The purpose of this paper is to show the laboratory-scale test method (LRP-2) and associated image analysis method (IAP-2) can be used to qualifying PSA paper label products as recycling-compatible, or RCA.

METHODS

Laboratory-Scale Test Method (LRP-2)

The laboratory-scale test method consists of five unit operations. The complete details are available on the Tag and Label Manufacturer's Institute (TLMI) website [4]. The first operation is high-consistency, 15% pulping of 360 g OD pulp, which contains 1% adhesive by weight, for 8 min. After 0.15-mm slotted screening with a Sommerville Flat Screen, the pulp is dewatered with a 200 mesh screen. The pulp slurry is further cleaned in a Denver flotation cell and finally dewatered with a 200 mesh screen. Three pulp samples were taken for image analysis (1) after pulping, (2) after 0.15-mm screening, and (3) after flotation. INGEDE method 12 includes an accelerated aging step, i.e., 60° C for 72 hours. LRP-2 only uses room temperature aging for 72 hours. At this time, no effort has been made to quantify the impact of aging, but it is the subject of ongoing research.

Pilot-Scale Test Method

The pilot plant test method is described in the USPS specification [1] and 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, because pressure screening will be essential to removing most adhesive formulations, the pilot test method was developed around commercial pressure screens. Sample sizes and flow rates in the test method were set to give approximately 1 h 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, passing speeds for the pilot plant trials are 0.35 to 0.4 m/s. Lower passing speeds are often associated with higher screening efficiencies, at the cost of fiber fractionation.

Table 1: Pressure screen operating parameters

Slot size (mm)	Inlet consistency (%)	Open area (%)	Passing speed (m/s)
0.3	1.10	4.3	0.41
0.1	0.94	2.3	0.35

Total fiber yield is 67%, which is lower than a typical mill-scale operation. Significant yield improvements could be achieved by adding secondary pressure screens and forward cleaners.

During a trial, temperatures and flowrates are recorded using a computer data acquisition system and used to monitor operating parameters. Consistencies are also determined on all the process streams. Pulping temperatures are all between 44°C and 48°C.

Image Analysis Method (IAP-1)

Image analysis can measure the size and count the number of particles in handsheets. Development and testing of the image analysis method used in this study was described previously [7,8] and the complete details can be found on the TLMI website [4]. It involves making TAPPI standard handsheets, speed drying them, and staining any hydrophobic adhesive particles with an anthraquinone dye in heptane. After evaporation of the heptane, stained particles are counted and the area measured using a flatbed scanner and image analysis software.

Statistical Analysis

Theoretically, random sampling of rare events leads to Poisson distributions [9]. For many of the adhesives in this study, the number of adhesive particles in the final pulp was less than 2 per handsheet, so analysis showed that a Poisson distribution is a better approximation to the number of particles per handsheet than a normal distribution. An example of such a set of handsheets is shown in **Figure 1.** Calculation of averages and 95% confidence intervals are often based on the assumption of normal distributions. In this case, the observation of Poisson distributions implies that alternative analyses are required. For the example shown in Figure 1, the minimum Chi square is found at 2.5 particles/sheet, and the 95% confidence interval is 1.8 to 3.9 particles.

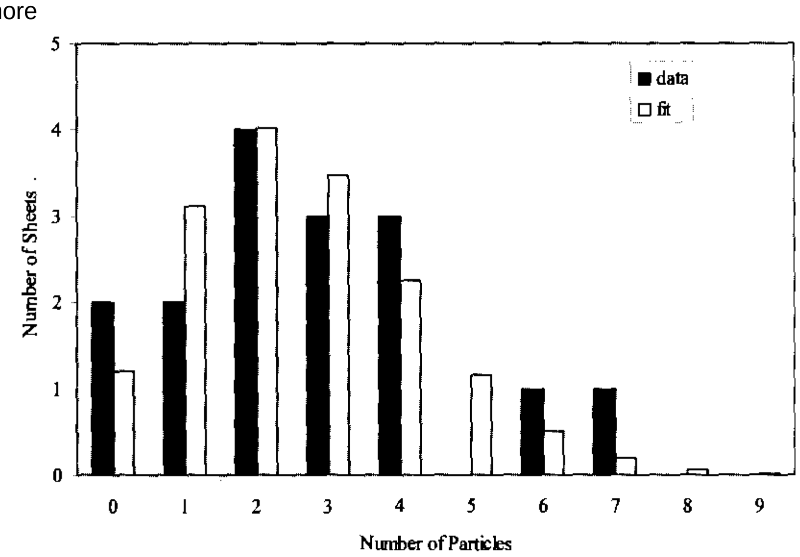

Figure 1: Typical distribution of number of particles per handsheet. The fit distribution is a Poisson distribution with an average of 2.5 particles/handsheet.

For our analysis, a minimum Chi square method was implemented using the Solver function of Microsoft Excel (Microsoft Corp., Redmond, Washington). The Chi square parameter was calculated between the data and a model Poisson distribution and minimized by changing the average number of particles per handsheet Furthermore, the upper 95% confidence limit was determined by adjusting the average number of particles in the model up to the point where the probability that the real and approximate distributions were the same dropped below 5%. Similarly the lower limit was determined by moving the model average down. Since Poisson distributions may be approximated by normal distributions as the number of particles increases, these alternative methods need to be applied only to samples that exhibited low number of particles per sheet By trial and error, it was determined that when the number of particles per handsheet exceeded 5, an analysis assuming a normal distribution was a reasonable approximation; below this limit the alternative method assuming a Poisson distribution was required. The spreadsheet used for these calculations can be obtained from the authors.

GENERATION OF TEST MATERIALS

Three adhesives produced by Dyna-Tech (Grafton, West Virginia), Techcryl 2077, Techcryl 2201, and Techcryl 2233, were used for validation of LRP-2. All three are water-based tackified acrylic PSAs. Actual adhesive samples used to prepare the labels for testing were taken from commercial production batches of each adhesive. Although these labels were prepared in the laboratory, they were in conformance with Section 3.1.1 of LRP-2, which requires that the adhesive formulations be representative of the finished commercial product All three adhesives contained coating modifiers, foam control agents, and wetting agents necessary for commercial coating processes.

Label samples of Techcryl 2077 and Techcryl 2201 were prepared on production coating lines for the pilot-scale testing. This was necessary in order to obtain sufficient quantities of each adhesive coated onto the three specified label stocks.

The unprinted benchmark material is an acrylic-based adhesive coated on a standard stamp facestock at approximately 24 g/m2 and is the same laminate from the same lot that was used as the benchmark for the USPS study [1,2]. Tests with this material since its conversion have suggested that its behavior during recycling has not changed significantly. This facestock contained the USPS water-soluble primer beneath the adhesive layer.

The stamp laminates had a similar adhesive coating weight as the benchmark and were used with no characterization of their chemical composition but likely included both acrylic and rubber-based formulations. They were all printed with a standard test pattern using a rotogravure process approximately seven years prior to the start of the current study.

RESULTS

Behavior in Pulpers

Behavior of an adhesive in a paper recycling system is largely determined in the pulper. Adhesives that are reduced to numerous small particles are likely to overwhelm even the most modern screening, cleaning, and flotation systems. Thus, in designing a test method to determine adhesive behavior during recycling, the laboratory- and pilot-scale pulpers should impart shear fields and energy intensities similar to those of commercial pulpers. The pilot-scale pulper used in this study is a 1/5 scale commercial helical-rotor Voith high-consistency pulper, Model HC-1.5. At 12.5% consistency and with a 20-min pulping time, this pulper consumes approximately 52 kW h/OD ton of pulp. The Adirondack Formax Model 450H laboratory pulper was operated at 15% consistency for 8 min. Estimates of energy input into the pulp on the laboratory pulper were determined to be extremely inaccurate because of mechanical and electrical losses.

Inspection of particle area distributions for several adhesives shows that the number fraction distributions on the laboratory and pilot-scale pulpers are similar. A comparison of the laboratory and pilot distributions for Techcryl 2077 is shown in **Figure 2** (a log-log plot). This adhesive fragments to produce the majority of particles in small area classes, with 80% of the particles less than 0.1 mm^2. The laboratory pulper produced fewer particles in large area classes, suggesting that it is reducing the area of large particles for this adhesive. However, because the number of particles in these area classes is very low, the data may have significant statistical errors. Furthermore, **Figure 3** shows that the particle area distributions for the benchmark adhesive are virtually identical.

The distributions in Figures 2 and 3 appear to be approximately linear in the log-log plots. This observation suggests that a power-law model exists for particle area distribution. Fitting the equation, $\text{Number} = a(\text{area})^n$ (1) to these data, exponent n was found to be near -1.0 for a wide range of materials. If one assumes that energy absorption is linearly related to surface area generation, then this model suggests that both the laboratory and pilot-scale pulpers are putting a uniform amount of energy into each area class. In summary, the particle area distributions generated by both laboratory and pilot-scale pulpers are very similar.

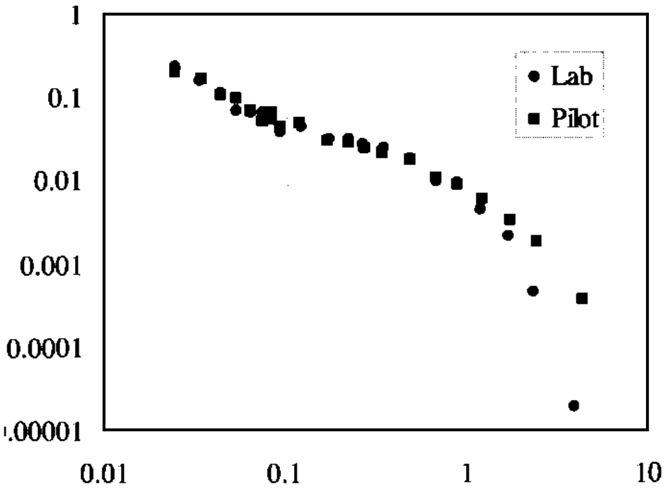

Figure 2: Log-log plot of number distribution of particles for Techcryl 2077

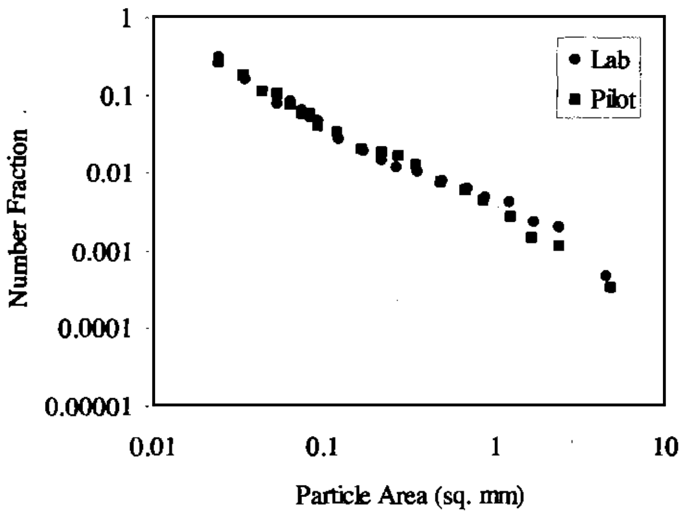

Figure 3: Log-log plot of number distribution of particles for benchmark adhesive.

Behavior during Screening

Adhesive fragmentation during pulping largely determines removal efficiency during slotted screening. Adhesives that generate large numbers of particles in the small area classes are not well removed. While an arithmetic average area does not describe the skewed distribution for particle area, it does provide a measure fo the relative number of small particles. Figure 4 shows that, as expected, more adhesive passes through slotted screens when the adhesive fragments into smaller particles in the pulper. The data in Figure 4 are for the laboratory-scale screening, but a similar trend is found for pilot-scale tests, as is illustrated by the curve. This curve was generated by a least squares fitting procedure using the data from the three plot tirals from this study as well as data for 10 samples of stamp laminates. These data may suggest that the pilot screens are less efficient than the laboratory screen when particle area is less than 0.5 mm^2 but similar for larger particles.

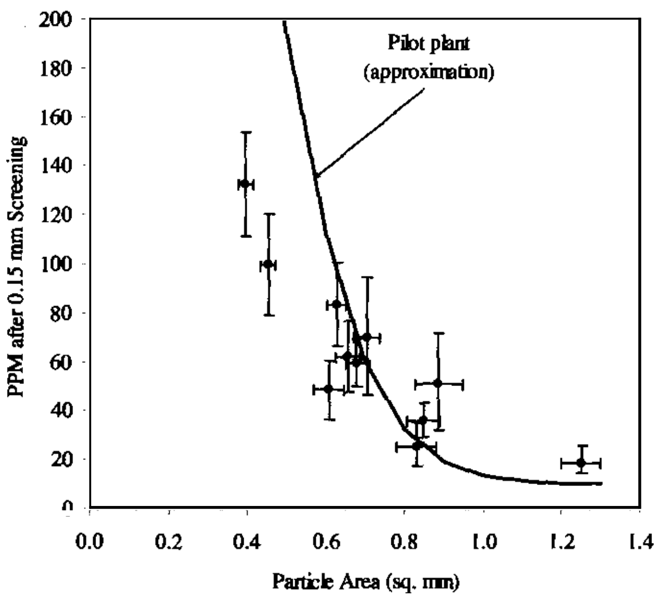

Figure 4: Adhesive level after laboratory-scale 0.15-mm screening versus arithmetic average particle area in pulper samples. Error bars represent the 95% confidence interval for the measurements. The pilot plant results are shown as a curve, which was fit to data from this study and 10 other stamp adhesives.

screen, removal during flotation is the ultimate determinant of success. Figure 5 shows pilot-scale final pulp PPM values for three different adhesives and the average value of successful adhesives in the USPS stamp project.

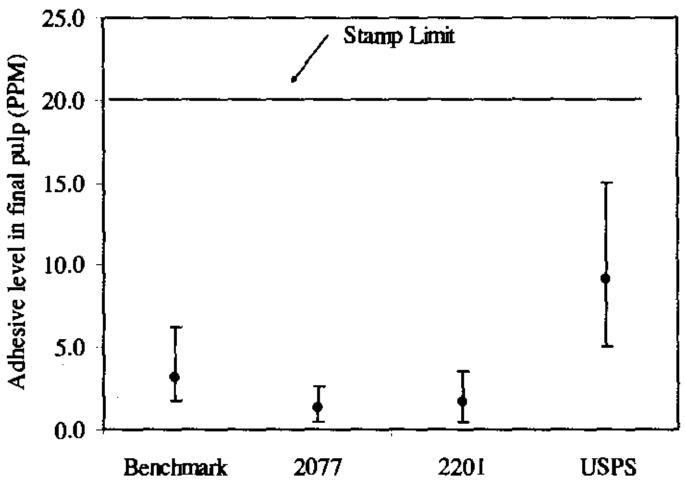

Figure 5: Pilot-scale final pulp adhesive values for three different adhesives and the average of the 12 successful adhesives in the USPS environmentally benign stamp project. The error bars represent 95% confidence interval for the three adhesives and the range of average values for the USPS project.

Inspection of Figure 5 suggests that all three adhesives in this study give pilot-scale final pulp values below the limit set in the USPS stamp specification. In fact, they appear to be on the lower end of the range of values observed for other successful stamp adhesives. Because the pilot limit was set based on mill trials and was approved by representatives of six paper recycling companies, the materials in this study would not likely cause either operational or product quality difficulties in commercial paper recycling mills.

Values for laboratory measurements are shown in Figure 6. The benchmark is shown three times, twice using the proposed test method and once using an adhesive that was pre-dyed before recycling by flooding the adhesive surface with a heptane solution of dye. Comparison of the benchmark samples indicates that pre-dyeing the adhesive changes its properties and should not be used in the testing. In data not shown here, the pre-dyed adhesive was changed in a way to produce larger particles in the pulper, which make it easier to screen, and ultimately presented less adhesive to be removed by flotation. Also shown in Figure 6, are results from retests of five qualified stamp adhesives that were coated and printed in the fall of 1998. All of the materials were below 10 PPM in the original tests. The performance of three of the samples does not appear to have been affected by aging, but two of the stamp adhesives have become much harder to remove. Loss of plasticizer and migration of adhesive into the facestock are two affects of aging that could result in this poor performance.

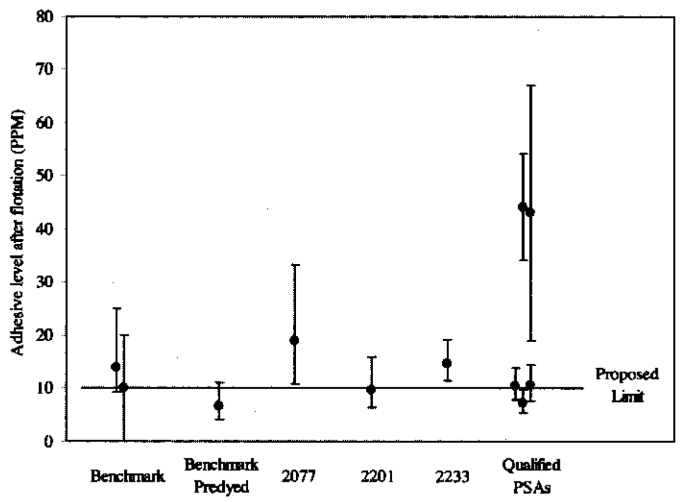

Figure 6: Laboratory-scale adhesive levels in the pulp after flotation. The error bars represent the 95% confidence interval for the measurements.

Also shown in Figure 6 is the proposed limit on final PPM values for an adhesive to be declared a RCA. This limit was chosen to be the same as the limit chosen by the USPS for the laboratory-scale tests. Comparing the results in Figures 5 and 6, Techcryl 2077 passes the pilot-scale test but does not pass the laboratory-scale test. Its final value of 19 PPM is well above the value of 10 PPM, which has been proposed as the laboratory-scale final pulp limit based on previous USPS work, with a confidence level greater than 95% that the value is greater than 10 PPM. Similarly, Techcryl 2233 does not pass the laboratory-scale test. In contrast, Techcryl 2201 passes, with a final value of 9.6 PPM and a 95% confidence interval of 6 to 16 PPM.

Although the data presented here indicate that materials that pass the laboratory-scale test will also pass the pilot scale-test, many samples tested failed both tests. For example, a hot-melt material that produced an average pulper particle area below 0.2 gave final pulp PPM values greater than 1,000 for both laboratory- and pilot-scale tests [10].

In summary, the value of 10 PPM as a limit appears to be a conservative choice, because the laboratory-scale test method will likely exclude some materials that could pass the pilot-scale or mill-scale tests.

EFFECT OF PAPER SUBSTRATE

Previous work has shown that the label facestock paper to which an adhesive is attached has an impact on adhesive behavior during paper recycling [10]. As part of LRP-2, one can qualify either a specific product or an adhesive for general use. For general adhesive qualification, the adhesive needs to demonstrate good performance on a wide variety of facestocks; thus, a single test of a facestock blend is used. The facestock blend-chosen to represent the weight distribution of materials coated in the label industry-contained 80% electronic data processing paper (EDP), 10% coated one side paper (C1S), and 10% thermal transfer paper (TT). Figure 7 shows results of tests of two adhesives on each of these three label facestocks and the blend. For both of these adhesives, the C1S face paper gives the best performance. The blend and EDP results are not statistically different from each other.

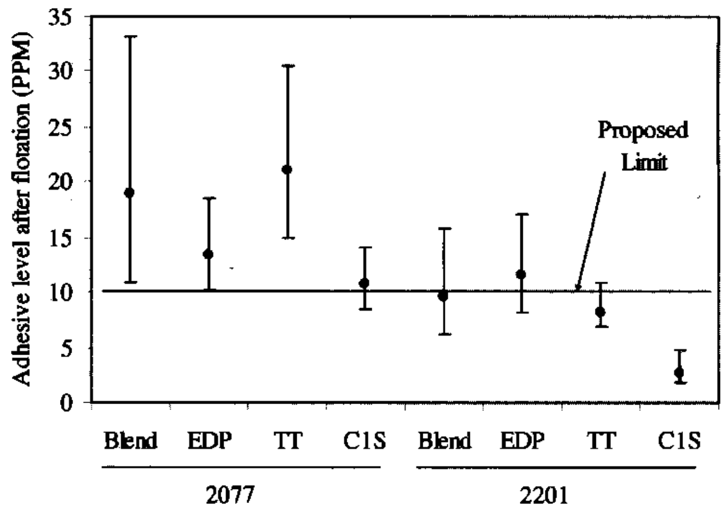

Figure 7: Laboratory-scale adhesive levels in the pulp after flotation for two adhesives on three different substrates. The error bars represent the 95% confidence interval for the measurements.

These data show that blending laminates with the same adhesive on different facestocks does not introduce any unexpected behavior. The use of a blend to represent all facestocks, as specified in LRP-2, seems to be supported by these data.

CONCLUSION

The particle area distributions generated by the laboratory- and pilot-scale pulpers are similar. Furthermore, the relationship between particle area and PPM after screening also appears to be similar for the laboratory- and pilot-scale tests. The fact that laboratory-scale pulping and screening can mimic pilot-scale unit operations suggests that a laboratory-scale test may be sufficient to determine the recycling compatibility of an adhesive.

All three adhesives used in this study passed the pilot-scale test by having a final pulp PPM value of less than 20 PPM, but only one adhesive passed the laboratory-scale test by having an average final pulp PPM value below 10 PPM. Thus, if an adhesive passes the laboratory-scale test, one can be assured that it will pass the pilot-scale test Furthermore, because the pilot-scale test has been previously shown to predict performance at a recycling mill, an adhesive that passes the laboratory-scale test is also likely a RCA.

Finally, the results shown here indicate that adhesives should be tested in a form as close as possible to the form that will be presented to the paper recycling stream. Paper substrate, coating weight, sample age, and adhesive additives can all significantly affect adhesive behavior under typical recycling conditions,

REFERENCES

1. Houtman, C., Sutherland, N.R., Bormet D., and Donermeyer, D., 2000 TAPPI Recycling Symposium, TAPPI Press, Atlanta, 2000, vol. 2, p. 403.

2. Houtman, C., Seiter, D.F., Donermeyer, D., and Ross Sutherland, N., 2002 TAPPI Pulping/Process and Product Quality Conference, San Diego, CA, USA, 8-10 Sept. 2002, 5pp

3. Clinton, W, Federal Register Vol. 65, No. 81, p.24595, April 26, 2000, http://www.gpoaccess.gov/fr/search.html.

4. http://www.tlmi.com/recycling-standards.php

5. http://www.ingede.com/ingindxe/methods/meth12pe.pdf

6. Putz, H.J., Ackermann, C., and Gottsching, L., Adhesives Less Detrimental to Paper Recycling Workshop, Genoble, France, 19-20 June 2002.

7. Houtman, C., and Tan, F., 2002 Pulping/process and Product Quality Conference, San Diego, CA, USA 8-10 Sept. 2002, 5pp

8. Rosenberger, R.R., and Houtman, C.J., 2000 TAPPI Recycling Symposium, TAPPI Press, Atlanta, 2000, vol. 2, p. 713.

9. Box, G.E.P., Hunter, W.G., and Hunter, J.S., Statistics for Experimenters: An Introductions to Design, Data Analysis and Model Building, John Wiley and Sons, New York, 1978, p. 137.

10. Houtman, C., Scallon, K., Guo, J., Wang, X., Severtson, S., Kroll, M., and Nowak, M., 7th Research Forum on Recycling, Pulp and Paper Technical Association of Canada, 2004, p, 11,