CONTROLLING ADHESIVE BEHAVIOR DURING RECYCLING

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ABSTRACT

Adhesives can be formulated to facilitate their removal by typical paper recycling unit operations. The investigations described in this paper are focused on determining fundamental properties that control particle size during pulping. While pressure-sensitive adhesives (PSAs) with high elastic moduli tend to survive pulping with larger particles, facestock and adhesive surface properties also play critical roles. We investigated both the effect of wet-strength additives and sizing agents on screen removal efficiency and found that one can shift a recycling-benign adhesive to one that will probably cause a problem in a mill just by changing the facestock properties. A conclusion from this work is that all the components of a PSA label, laminate, facestock, primers, and adhesive, play a role in determining the behavior of adhesives during pulping and screening.

INTRODUCTION

Pressure-sensitive adhesives (PSAs) continue to cost the recycling industry downtime, product quality, and money. Technical solutions in recycling mills, better screens, reduced pulper temperature, chemical additions, and passivation, have provided some relief. Preventing problematic materials from entering the recovered paper stream will be a more effective long-term solution to the stickies problem. Efforts to first identify troublesome components of adhesives and then modify adhesive formulations to make them more compatible with recycling are beginning to be effective.

The amount of PSAs on labels, envelopes, and stamps has rapidly increased over the last 10 years. For example, in the United States, 1% of stamps had PSA in 1989. In 2004, more than 96% of stamps have PSA. Although stamps are a small contributor to the PSA currently in the recovered paper stream, this rapid rise in use illustrates the increase in consumer acceptance of PSA products.

Soon after large-scale introduction of PSAs into the recovered paper stream began, observations that PSAs were causing “stickies” problems in recycling mills appeared in the trade literature [1]. Some adhesive suppliers soon began work on developing products that were compatible with paper recycling [2], but market acceptance was slow. In 1994, the United States Postal Service (USPS) initiated the Environmentally Benign Pressure Sensitive Adhesives for Postal Applications program [3]. Resulting specification for stamp adhesives were released on July 5, 2001. The USPS recycling-benign specification requires that adhesives used on stamps be removed by fine screening (usps-stamp-technology.com). Also, two efforts to develop certifications of recycling-benign adhesives have been initiated [4] (www.tlmi.com/data/b9/index.html). Unfortunately, the market penetration of reformulated adhesives has largely been driven by governmental regulation, and thus these materials are only slowly moving into general use.

Fundamental studies of adhesive-containing products have been conducted to determine properties that control adhesive particle size during repulping. Lucas et al. [5] have shown that pulper particle size is positively correlated with yield strain and elastic modulus of various adhesive formulations. In fact, it has been shown that laboratory-measured physical properties of hot-melt PSAs can be used to accurately predict adhesive removal during slotted screening [6]. Beyond bulk adhesive properties, surface energy and wet strength of the paper facestock has also been shown to play a role in determining pulper particle size [7].

The purpose of this paper is to describe factors that control adhesive particle size during pulping. Because particle size developed in the pulper largely determines the ultimate removal efficiency by the whole recycling process, results discussed here will lead to adhesives that are easier for a typical paper recycling mill to remove.

MATERIALS AND METHODS

Chemicals and Materials

All experiments used a laminate structure consisting of a thermoplastic or hot-melt PSA sandwiched between various facestocks and envelope paper. Table I lists tensile loss and surface energies of tested facestocks and substrates. Hercules Corporation (Wilmington, Delaware) provided the alkenyl ketene dimer size and polyamide wet-strength resin for producing the laboratory papers. Boise-Cascade Corporation (International Falls, Minnesota) provided papermaking fiber and commercial facestocks. This study presents data for a thermoplastic or hot-melt PSA formulation, which was supplied by H.B. Fuller Company (St. Paul, Minnesota). The PSAs consist of styrenic block copolymers, tackifying resins, and plasticizers. Techniques used in the characterization of bulk mechanical and surface properties of paper and adhesives are described in detail elsewhere [6, 7, 8].

1. The use of trade or firm names in this publication is for reader information and does not imply endorsement by the U.S. Department of Agriculture of any product or service.

TABLE I. PROPERTIES OF FACESTOCKS

Facestock	Basis weight (g/m^2)	Tensile loss (%)	Surface energy (mJ/m^2)
Untreated handsheet	60	98 ± 2	71.1
Envelope paper	90	97 ± 6	43.4
Electronic data processing (EDP)	77	96 ± 4	39.7
Coated one side (C1S)	77	94 ± 4	39.7

Laboratory-Scale Test Methods

Adhesives were tested by Forest Products Laboratory (FPL) and University of Minnesota (U of M). The methods were similar to that described in the USPS specification (usps-stamp-technology.com) consisting of high-consistency, 15%, pulping of 360 g o.d. pulp which contains 1% adhesive by weight in an Adirondack 450H pulper (Queensbury, New York) with a water jacket to maintain the desired temperature. At FPL, 0.15-mm slotted screening, with a Sommerville (Wauconda, Illinois) automated flat screen, was followed by dewatering on a 200-mesh screen. Samples taken after pulping and after 0.15-mm screening were analyzed for residual adhesive levels using a dyeing and image analysis test method [9]. At the U of M, 0.38-mm slotted screening was used and removal of PSA was quantified gravimetrically using a cellulose dissolution technique [8].

Pilot-Scale Test Method

The pilot plant test method provides six major unit operations: pulping, slotted screening, forward cleaning, flow-through cleaning, flotation, and washing and is similar to the method described in the USPS specification (usps-stamp-technology.com). Pulping was conducted at 12.5% consistency with a 50/50 mix of virgin copy and virgin envelope paper and approximately 1% PSA material. The total pulper load was 112.5 kg o.d. The tank sizes and flow rates in the test method were set to give approximately 1 hour of screen operation. The total fiber yield ranged from 50% to 67%, which is lower than typical mill-scale operation and largely due to the lack of adequate secondary screening and cleaning unit operations.

The physical properties of adhesives are very temperature sensitive. Thus, temperature was 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 repeatable within 2°C. Pulp samples were taken after each unit operation. These samples were analyzed for residual adhesive levels using a dyeing and image analysis test method [9].

RESULTS AND CONCLUSIONS

Validity of Laboratory Results

Previous work at FPL has shown that results of pilot-scale trials of adhesive materials can be used to predict whether an adhesive will cause either runability or quality problems in recycling mills [10]. The data also show that laboratory trials are less correlated with adhesive behavior in mills. Closer inspection of the data revealed that laboratory flotation was too effective and that laboratory pulping and screening are useful indicators of adhesive removal during recycling. The conclusion that laboratory pulping and screening are representative of results generated during pilot-scale, and by extension during mill-scale, trials is supported by the data shown in Figure 1. For the hot-melt PSA used in these trials, the elastic modulus is very temperature-dependent — as temperature goes up, particles size goes down. The particle size versus temperature curves are very similar in the laboratory and pilot-scale pulping. Thus, one can conclude that the forces generated in the lab pulper are similar to those in the pilot pulper. Particle size in Figure 1 is the number-averaged projected area of the dyed particles in handsheets. While the measured size is related to particle sizes generated in the pulper, the relationship changes for different particle morphologies.

When the same material is subsequently screened using the respective laboratory and pilot-scale screening systems, one can conclude that the laboratory and pilot system behave similarly (Fig. 2). The laboratory system exhibits slightly better screening efficiency. The pilot screening efficiency in Figure 2 is for the first primary screen, which has 0.3-mm slots. Since the laboratory screen had 0.15-mm slots, the slight improvement in lab efficiency is to be expected.

At U of M, lower PSA removal efficiencies for the same laminates were observed. Figure 3 shows the PSA screening efficiency versus temperature for three different sets of experiments. Typical 95% confidence intervals for removal efficiency curves are less than 5% and for temperatures are

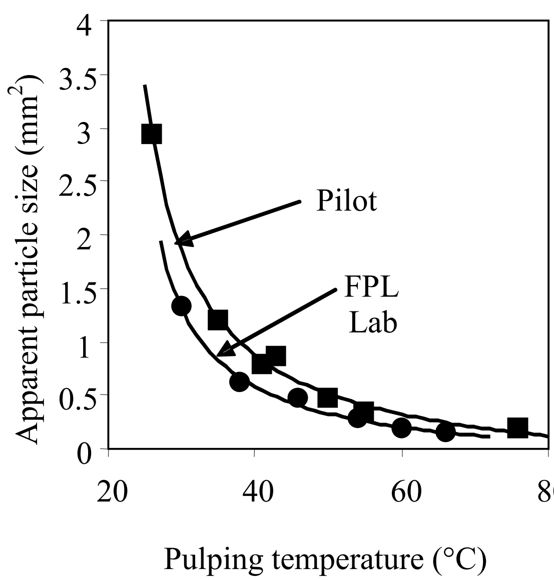

Figure 1. Pulper particle size versus pulping temperature for a representative hot-melt PSA.

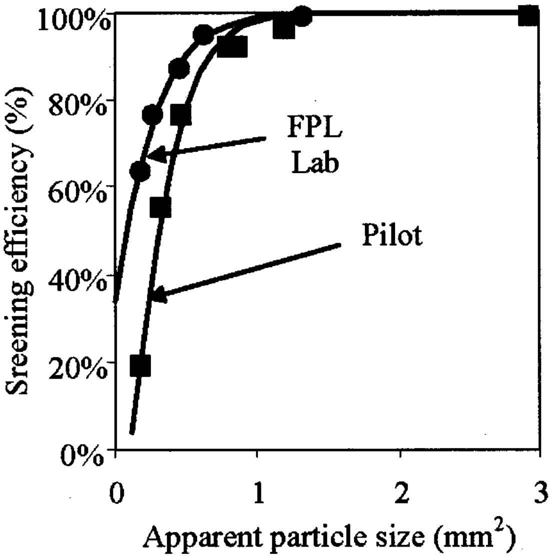

Figure 2. Screening efficiency versus apparent particle size as measured by image analysis.

2°e. Although two different methods are used for determining PSA content (image analysis of accepts versus gravimetric measures of isolated particles rejects), the removal curves have a similar sigmoidal shape. For most adhesives that have a pilot screening efficiency greater than 90% pass the USPS specification (usps-stamp-technology.com). Thus, for the adhesive used in these trials, a pulper temperature below 40°C will be required for it to be removed efficiently in a recycling mill.

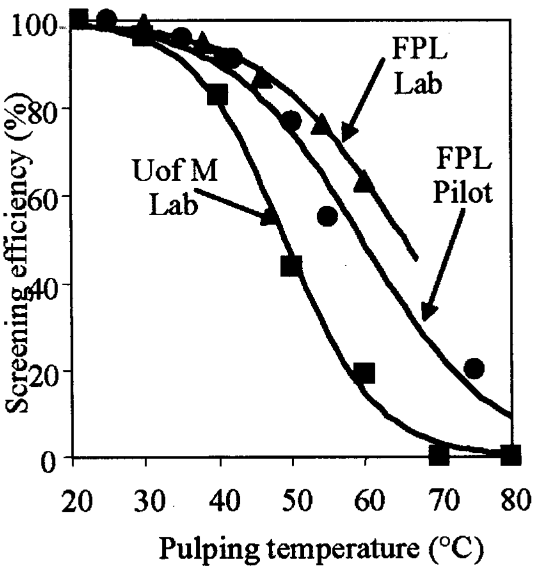

Figure 3. Comparison of PSA screening efficiency determined by pilot testing and laboratory testing at both FPL and U of M.

The correspondence of laboratory pulping and screening results to pilot results indicate that laboratory studies should be sufficient to explore the effect of various adhesive properties on pulper particle size and screening efficiency.

Review of Thermo-Mechanical Model

.As.was introduced elsewhere [6], removal efficiency versus temperature data for hot-melt PSAs can be fit with a sigmoidal function of the form

$$\text{Removal efficiency} = \frac{\exp\left(\frac{T_{50}-T_R}{\alpha}\right)}{1+\exp\left(\frac{T_{50}-T_R}{\alpha}\right)} \times 100\% \quad (1)$$

where T_R is the repulping temperature and T_{50} and a are fitting parameters. It was previously shown that the shear adhesion failure temperature (SAFT) is an accurate predictor of *T50* values, and *a* was found to correlate with the thermal separation between the glass transition *(Tg)* of the rubbery domains for the base copolymer and the transition related to the *Tg* for the polystyrenic domains [6,7]. Measurements used to develop this model involved facestocks that have little wet strength and weak adhesion to PSA ftlms in aqueous environments.

Adhesive Removal for Various Facestocks

.Aswe have previously demonstrated [7], the properties of commercial paper facestocks have a significant impact on the removal of attached PS.As.(Fig. 4). For example, at 50°C, the removal efficiency of CIS measured using the laboratory procedure was 37% compared with 68% and 82% for untreated paper and no facestock, respectively. A least squares fitting of th~ parameters in Equation (1) to the data was conducted. The *T50* values for the CIS (coated one side), EDP (electronic data processing), untreated paper (handsheets), and no facestock are 47°C, 49°C, 57°C, and 61DC,respectively. In contrast to the shift in *T50* values, the estimated *a* values were all similar, 6.5°C, 6.2°C, 6.4°C, and 6.2°C, respectively.

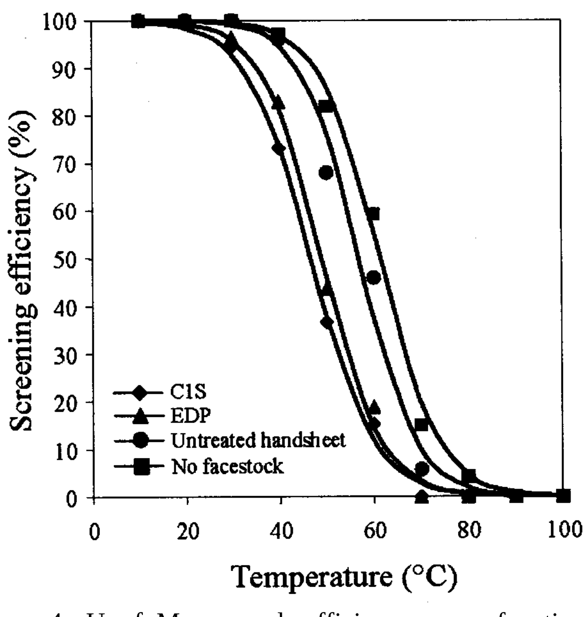

Figure 4: U of M removal efficiency as a function of temperature for hot-melt PSA for laminates produced with CIS, ED~ untreated handsheet, and no facestock.

It appears that the PSA system without facestock, i.e., the PSA film and envelope substrate only, provide for the formation of the largest residual PSA particles subsequent to repulping and thus the highest removal efficiencies. It has been proposed that the facestock properties most responsible for reducing PSA particle size are wet-strength and surface energy [7]. These properties appear to limit the ability of a PSA to release from the facestock and assume a more collapsed configuration.

Effect of Wet-Strength and Sizing Agents

Figure 5 shows the influence of increasing wet-strength resin concentrations. Here, the removal efficiency is plotted versus the tensile loss for the paper facestock. Tensile loss is the percentage decrease in tensile strength of the paper after being saturated with water, i.e., the wet-strength of the paper increases with decreasing tensile loss. Following the curve from right to left, it can be seen that increasing the wet strength of the facestock decreases the removal of the attached PSA. The minimum at 95% tensile loss corresponds to the wet-strength level required to begin inhibiting fiberization during repulping. As tensile loss decreases below this value, the facestock is fiberized to a lesser extent. This strengthened paper is believed to act as reinforcement for the PSA, reducing its fragmentation [7]. Indeed, if this effect is accompanied by an increase in PSA–facestock adhesion, PSA removal efficiencies can be increased, often dramatically, at the expense of fiber yield [8].

Figure 6 shows the T_{50} parameters extracted from fits of PSA removal efficiency versus temperature data for laminates produced with facestocks treated with increasing levels of sizing agent. The inset of Figure 6 shows facestock surface energy calculated from contact angle measurements versus apparent sizing concentration.

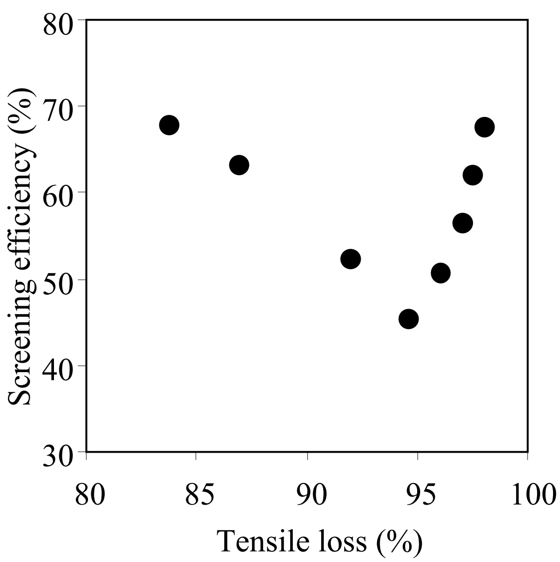

Figure 5. Removal efficiency of hot-melt PSA as a function of percentage decrease in tensile strength of facestock subsequent to saturation with water (i.e., tensile loss) at 50°C.

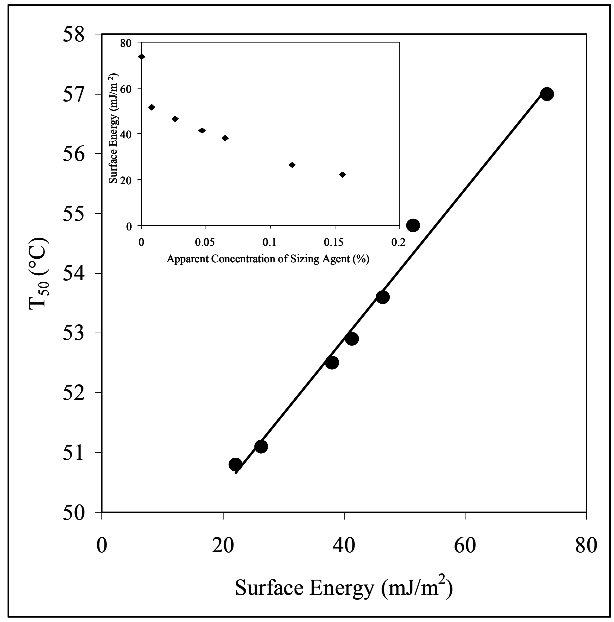

Figure 6. T_{50} values obtained from fits of removal efficiency using Eq. (1) as a function of facestock surface energy.

The results indicate that increasing the surface energy of the facestock shifts the removal curves for the attached PSAs to higher temperatures. As indicated by the inset curve, these changes occur at a relatively low degree of sizing, levels commonly found in commercially produced papers. The influence of surface energy can be described to a certain extent through the thermodynamic work of adhesion for the removal of PSA from paper facestock. It can be shown that increasing the surface energy of cellulose fiber reduces the thermodynamic work of adhesion for the removal of attached PSA in an aqueous medium [8]. This effect reduces the amount of stress required to remove the PSA film, promoting the formation of larger adhesive particles during repulping.

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In: 7th Research Forum on Recycling, Sept. 27-29, 2004, Quebec City, QC. Montreal, Quebec : Pulp and Paper Technical Association of Canada, 2004. Book or CD-ROM. ISBN Book 1-897023-05-7 ; ISBN CD 1-897023-06-5. pp. 11-16