

Limiting the Impact of Pressure Sensitive Adhesives on Paper Recycling by Controlling their Fragmentation during Repulping

*Jihui Guo and Steven J. Severtson**

Department of Bio-based Products, University of Minnesota, 2004 Folwell Ave., Kaufert Laboratory, St. Paul, MN 55108

Carl J. Houtman and Karen L. Scallon

USDA Forest Service, Forest Products Laboratory, One Gifford Pinchot Drive Madison, WI 53705

Mark S. Kroll and Michael J. Nowak

H.B. Fuller Company, 1200 Willow Lake Boulevard, St. Paul, MN 55110

Eliminating the problems created by the presence of pressure sensitive adhesive (PSA) in post-consumer waste is a major technical challenge to expanding the use of recycled paper. A promising approach to this contamination issue is the redesign of adhesive products to lessen their impact on paper recycling operations. Here we review our efforts to develop thermoplastic or hot-melt PSA products that form large residual adhesive particles during repulping operations. This is achieved through manipulation film properties to inhibit fragmentation and use of paper face stock designs that facilitate the release of films in aqueous environments. The larger PSA particles that develop are removed with high efficiency at the pressure screens early in the recycling process, significantly reducing or eliminating PSA contamination.

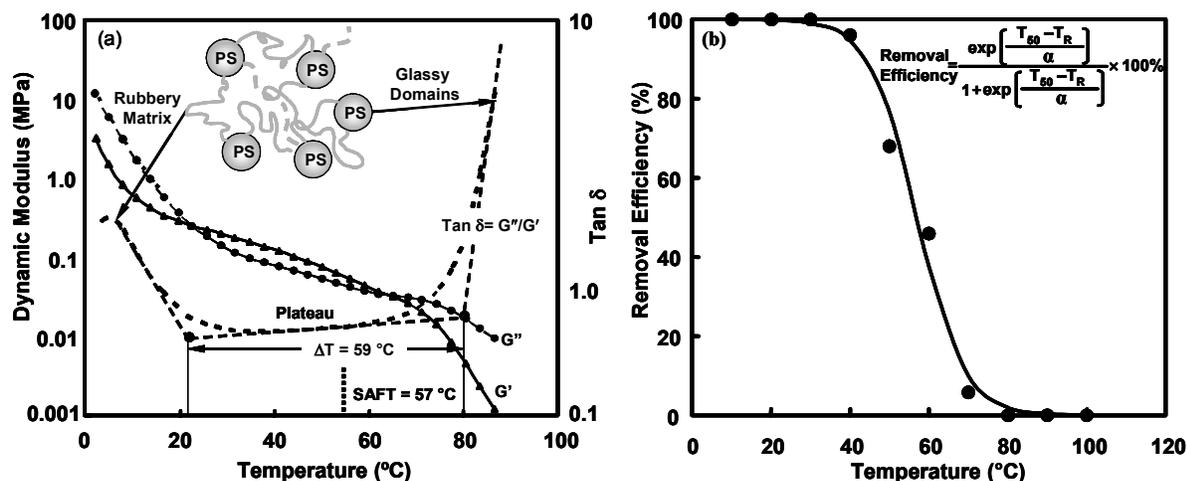


Figure 1. a.) Dynamic mechanical properties for a commercial hot-melt pressure sensitive adhesive. b.) Screening removal efficiency data for the commercial hot-melt PSA.

Thermoplastic PSAs are formulated with styrenic block copolymers and polymeric mixtures, which contain styrene segments in combination with rubbery polymeric components such as ethylene-propylene, ethylene-butene, isoprene or butadiene.^{1,2} The styrene and rubbery blocks are incompatible. This provides opportunities for manipulation of mechanical properties through the use of tackifying resins and plasticizers specific to the separated microphases. Figure 1a shows the dynamic mechanical analysis thermal scan ($\gamma = 10$ rad/s, shear mode) for a commercial hot-melt PSA. The two local maximums in the loss tangent are typical for hot-melt PSAs. As indicated by the inset structure, these transitions correspond to

the glass transition (T_g) of the tackified rubbery domains (lower temperature) and a transition related to the T_g of the polystyrene segments (higher temperature).¹ The glass transition of the tackified rubbery domains results in a significant increase in the viscoelastic properties of the PSA providing it with its adhesive properties. Subsequent to this transition, much of the remaining cohesive strength required for good adhesive performance is provided by the separated polystyrenic phase. As temperature is raised, cohesive strength decreases due to increasing fluid-like behavior of the rubbery phase and/or disruption of the styrenic phase.

The effect from this decrease of PSA film cohesive strength can be seen in the measured removal efficiencies shown in Fig. 1b for the commercial PSA. The laboratory procedure used is believed to be an excellent indicator of PSA performance in mill repulping and screening operations.³ (As part of this presentation, it demonstrated that laboratory results are consistent with pilot scale tests, specifically those used by the United States Postal Service to gauge the removal efficiencies stamp PSAs.) It can be seen that the decline in screening removal efficiency begins near the T_g of the rubbery phase, it has a sigmoidal shape and it occurs in the temperature region that separates the two transitions identified in Fig. 1a. An excellent empirical fit of removal data is found for a sigmoid function (Fig. 1b inset), where T_R is the repulping temperature, T_{50} is the inflection temperature of the curve and α determines its width. From a comparison of these fitting parameters with PSA performance data, it was found that T_{50} is similar to the film shear adhesion failure temperature or SAFT and α could be estimated from the thermal width between the rubbery and styrenic phase transitions (specifically, $0.1\Delta T$, see Fig. 1a), i.e.,⁴

$$\text{Removal Efficiency (\%)} = \frac{\exp\left[\frac{\text{SAFT}-T_R}{0.1\Delta T}\right]}{1 + \exp\left[\frac{\text{SAFT}-T_R}{0.1\Delta T}\right]} \times 100\% \quad (1)$$

As will be discussed, Eq. 1 provides an excellent prediction of removal efficiency as a function of repulping temperature for hot-melt PSAs when coated on untreated paper face stock. This is based on studies involving commercial and model thermoplastic PSA formulations. The model series consisted of a styrene-isoprene-styrene/styrene-isoprene (SIS/SI) linear triblock, diblock copolymer blend, tackifying resin, and naphthenic process oil. These were blended in combinations to produce a broad range of properties. Two types of tackifiers were used, a pentaerythritol ester of rosin and a C_5 petroleum hydrocarbon, to provide variation in the formulation composition.

As part of this study, the influence of paper face stock on PSA removal efficiency was also examined.⁵ Commercial and model PSAs were attached to paper substrates with bulk mechanical and surface properties that were manipulated using poly(amide epichlorohydrin) (PAE) and alkenyl ketene dimer (AKD) size providing variation in the strength and adhesion of PSA-substrate laminates. Water-based, reactive as well as hot-melt PSA formulations were tested. Figure 2 shows how increasing the concentration wet strength resin (a) and sizing agent (b) in the paper face stock decreases the screening removal efficiency of the commercial hot-melt PSA. The initial drop in the “V-shaped” curve of Fig. 2a indicates that increasing the wet strength of face stock that eventually fiberizes decreases removal efficiency. The influence of the wet-strength resin can be seen to be temperature dependent. Removal is not dependent on tensile loss at temperatures of approximately 30°C and lower and 70°C and

higher. In these temperature regions removal efficiencies were near 100 and 0%, respectively. The minimum in the curves at a concentration of about 0.03% corresponds to the treatment level at which the paper no longer disperses to individual fibers during repulping. As the concentration of wet strength resin is increased beyond this concentration, the degree to which the face stock fiberizes is reduced and there is an increase in the removal efficiency of the attached PSA. Increasing the concentration of sizing agent will reduce the surface energy on the paper face stock increasing PSA-face stock adhesion in aqueous environments. This is believed to be responsible for the observed decrease in removal efficiency. A linear correlation was found between removal efficiency and face stock surface energy. The sigmoid function shown in Fig. 1b provides an accurate fit of data for laminates involving various wet-strength and sizing face stock. However, the T_{50} or inflection points of these curves were shifted to values lower than that of the SAFT for the attached PSAs. T_{50} values were found to be proportional to both face stock surface energy and percent decrease in tensile strength when water saturated (i.e., the tensile loss, a measure of wet strength).

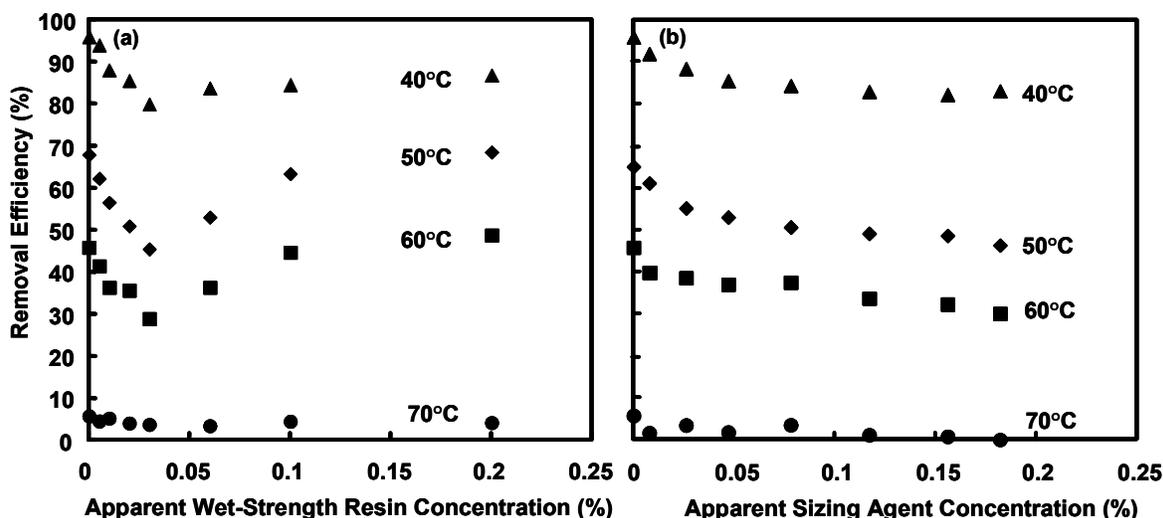


Figure 2. a.) Removal efficiency of commercial hot-melt PSA as a function of apparent PAE concentration over a range of temperatures. b.) Removal efficiency of commercial hot-melt PSA as a function of apparent AKD concentration over a range of temperatures.

Face stocks used in the production of commercial PSA products have a higher wet strength and a lower surface energy than untreated paper, and thus reduce the removal efficiencies of attached PSAs. As will be described, this influence can be significant, with reductions in removal efficiencies as high as 50% for temperatures commonly used in the processing of recycled paper. This decrease can be diminished by treating face stocks with hydrophilic polymeric coatings. Removal efficiencies actually higher than that of untreated paper were obtained using these face stock surface treatments.

Formulating commercially viable environmentally benign PSA products requires the introduction of properties that enhance their removal from recycling operations without sacrificing adhesive performance or increasing manufacturing costs. The results reviewed in this presentation show that environmentally benign PSA products can be accomplished with modifications to the PSA, paper substrates or both. It is hoped that the information presented

here will facilitate the development of laminates possessing higher screening removal efficiencies, thus limiting their impact on paper recycling operations.

Literature Cited

- (1) Tse, M. F. Studies of Triblock Copolymer-Tackifying Resin Interactions by Viscoelasticity Resins Interactions by Viscoelasticity and Adhesive Performance. *J. Adhesion Sci. Technol.* **1989**, *3*, 551.
- (2) Chu, S. G. Viscoelastic Properties of Pressure Sensitive Adhesives. In *Handbook of Pressure Sensitive Adhesive Technology*; Statas, D., Ed.; Van Nostrand-Rheinhold Co.: New York, 1988.
- (3) Houtman C. J., Scallon K. L., Guo J., Wang X. P. , Severtson, S. J., Kroll M. S., and Nowak, M. J. Factors Controlling Behavior of Adhesives in Recycling Systems. 7th Research Forum on Recycling , Quebec City, Canada, September 27-29, 2004.
- (4) Nowak, M. J.; Severtson, S. J.; Wang, X. P.; Kroll, M. S. Properties Controlling the Impact of Styrenic Block Copolymer Based Pressure Sensitive Adhesives on Paper Recycling. *Ind. Eng. Chem. Res.*, **2003**, *42*, 1681.
- (5) Guo, J., Severtson, S. J. and Kroll, M. S. Role of Paper Face Stock Properties in Determining Removal Efficiencies of Pressure Sensitive Adhesive Films from Recycling Operations. *Ind. Eng. Chem. Res.* **2004**, *43*, 1443.