Behavior of Adhesives During Paper Recycling

Steve Severtson, Jihui Guo, Haiyan Li, Helen Xu, Matt Dubay and Mark Calhoun
University of Minnesota

Carl Houtman
USDA Forest Service, Forest Products Laboratory
PSA Market Share and Composition for Label Products

- Water Based: 59%
- Hot Melt: 20%
- Solvent Based: 9%
- Reactive: 10%
- Other: 2%

Hot Melt PSA
- Tackifier
- Plasticizer
- Antioxidant
- Polymer

Water Based PSA
- Tackifier
- Wetting Agent
- Rheology Modifier
- Emulsifier
- Polymer
Project Objective

Development of new pressure sensitive adhesive (PSA) products that are engineered for enhanced PSA removal during the screening of recycled fiber

- Identification of properties that govern PSA removal and development of techniques for formulating benign PSAs
- Characterization of PSA-substrate adhesion and development of techniques for manipulating adhesion to enhance removal
- Development of wet-end recipes for facestock that promotes PSA removal

The impact on paper recycling operations should be a design parameter in the development of all PSA systems formulated for label applications.
Testing the Removal Efficiency of PSAs

PSA film is pressed onto facestock
laminates are attached to sheets in a sample of copy paper PSA content = 0.5%
removal efficiency is quantified gravimetrically

Cellulose Dissolution & Resin Oxidation (when required)

Removal \( \propto \) IA Area Efficiency \( \propto \) Fraction

Adirondack Formax Temperature Control Pulpers
- consistency = 10%
- 60 Hz (≈ 690 rpm)
- typical time = 30 min.

TAPPI Method T-205 om-88

Accepts

Rejects

Valley Flat Screen 15-cut screen (0.38 mm slots)
Research Strategy

Input
Commercial Assessment of New Product Approaches

Identification of Key Characteristics for Benign Materials

Synthesis and Formulation of Model and Commercial Systems

Database Generation & Analysis

Laboratory and Pilot Testing of Screening Removal Efficiencies

Characterization of Bulk Mechanical and Surface Properties

Output
Commercially Feasible Benign Products

Input
Access to Existing Product Lines
Hot-melt PSA’s

20-50 wt. % Base Polymer

30-60 wt. % Tackifier

0-25 wt. % Plasticizer

Easily Shaped for Characterization

G’, G”, Tan δ

Melt Processing
Predicting Hot-melt PSA Recycling Performance

Commercial PSA 1
SAFT = 57 °C
ΔT = 59 °C

Commercial PSA 2
SAFT = 80 °C
ΔT = 70 °C

Removal Efficiency (%)

Temperature (°C)

Tan δ

Commercial PSA 1
T_{50} = 57 °C
α = 7.3 °C

Commercial PSA 2
T_{50} = 80 °C
α = 6.2 °C

Predicted Removal
Water-based PSA’s

Polymerization

- Monomers
- Emulsifier(s)
- Initiator(s)
- Crosslinking Agents
- Biocide(s)
- Buffer(s)

Adhesive Emulsion

Tackifying Dispersions

- Rheology Modifiers
- Wetting Agent(s)
- Defoamer(s)

Formulation

- Characterization samples restricted to thin films with properties dependent on formulation

Formulated PSA

- G', G'', Tan δ?

Processing requires coating of low energy substrate and drying
COO$^-$-SO$_4$$^2$$^-$

Carboxylate anions on latex surface

Positive counterions (Na$^+$, K$^+$, or NH$_4^+$)

Persulfate residues from initiator

Anionic and/or nonionic surfactants

Coiled, HMW polymer chains

1 μm
Estimated compositions of water-based PSA latex and film

<table>
<thead>
<tr>
<th>Components</th>
<th>Coating ready emulsion dispersion (Mass%)</th>
<th>Dried PSA film(Mass%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>33.06-48.45</td>
<td>0</td>
</tr>
<tr>
<td>Monomers</td>
<td>50.21-61.46</td>
<td>88.93-97.79</td>
</tr>
<tr>
<td>Emulsifier(s)</td>
<td>1.21-6.58</td>
<td>2.13-9.86</td>
</tr>
<tr>
<td>Initiator(s)</td>
<td>0.08-0.31</td>
<td>0.12-0.55</td>
</tr>
<tr>
<td>Buffer</td>
<td>0.08-0.59</td>
<td>0.32-1.03</td>
</tr>
<tr>
<td>Biocide(s)</td>
<td>0.04-0.25</td>
<td>0.065-0.54</td>
</tr>
<tr>
<td>Reducer &amp; oxidizer</td>
<td>0.08-0.93</td>
<td>0.05-0.68</td>
</tr>
</tbody>
</table>

**Note:** The components of PSA latex film listed above refer to those in adhesive emulsion. Formulated PSAs also contain coating package comprised of tackifying dispersion(s), rheology modifier(s), wetting agent(s) and defoamer(s).
Fate of Latex Additives

TRANSFER COATING

“cold flow” in addition to additive migration

Temperature/Relative Humidity

HIGH

LOW
Removal Efficiency NOT Controlled by Performance Properties
### Composition of Model Systems

<table>
<thead>
<tr>
<th>Model System</th>
<th>RE (%)</th>
<th>Soft Monomer</th>
<th>Hard Monomer</th>
<th>Functional Monomer</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>n-BA</td>
<td>EHA</td>
<td>MMA</td>
</tr>
<tr>
<td>M1</td>
<td>2</td>
<td>70.8</td>
<td>10.0</td>
<td></td>
</tr>
<tr>
<td>M2</td>
<td>57</td>
<td>70.8</td>
<td>10.0</td>
<td>16.0</td>
</tr>
<tr>
<td>M3</td>
<td>79</td>
<td>70.8</td>
<td>10.0</td>
<td>16.0</td>
</tr>
<tr>
<td>M4</td>
<td>10</td>
<td>70.8</td>
<td>10.0</td>
<td>3.2</td>
</tr>
<tr>
<td>M5</td>
<td>77</td>
<td>70.8</td>
<td>10.0</td>
<td></td>
</tr>
<tr>
<td>M6</td>
<td>70</td>
<td>70.8</td>
<td>10.0</td>
<td></td>
</tr>
<tr>
<td>M7</td>
<td>75</td>
<td>70.8</td>
<td>10.0</td>
<td>16.0</td>
</tr>
<tr>
<td>M8</td>
<td>90</td>
<td>80.8</td>
<td>16.0</td>
<td>3.2</td>
</tr>
<tr>
<td>M9</td>
<td>84</td>
<td>80.8</td>
<td>8.0</td>
<td>3.2</td>
</tr>
<tr>
<td>M10</td>
<td>90</td>
<td>80.8</td>
<td>16.0</td>
<td>3.2</td>
</tr>
<tr>
<td>M11</td>
<td>81</td>
<td>80.8</td>
<td>16.0</td>
<td>3.2</td>
</tr>
</tbody>
</table>
Tensile Strength vs. Removal

Dry Tensile Force at 22°C

Wet Tensile Force at 22 and 50°C

Jihui Guo, Steven J. Severtson and Larry G. Gwin *INDUSTRIAL & ENGINEERING CHEMISTRY RESEARCH* 49(9), 2753-2759, 2007
Microstructure Characterization of PSA film

Cryo SEM image

AFM image
AFM of Adhesive Films

**Tapping Mode**

- **Cantilever**
- **Tip**
- **Sample**
- **Piezoelectric (z)**
- **Stage (x,y)**
Surface morphologies of water-based PSA films

AFM image of PSA film

Particle size distribution
Surfactant Removal

Top surface of PSA latex film

Solvent Soaking for 1 minute

Solvent soaking for 5 minutes
Effect of Moisture Cycles

Initial cycle 3% RH

Increase moisture to 90% RH

End cycle 3% RH
Results

• Water uptake of PSA films likely controls behavior during paper recycling

• Surfactants used to synthesize the original polymers can migrate during processing

• Physical properties of wet films can be used to predict removal efficiency during screening
Recycling-compatible Adhesives (RCA)

- A committee associated with the Tag and Label Manufacturers Institute (TLMI) have developed test methods and specification for certifying an adhesive is RCA.

- Several label suppliers are now marketing labels with RCA’s.

- State of Wisconsin is using this specification for Governmental purchases.
Recycling-compatible Adhesive Testing

PSA label is pressed onto envelope paper at 5% label stock by weight

Adirondack Pulper
Temperature = 46°C
Consistency = 15%
40 Hz (≈ 500 rpm)
Time = 8 min.

Paper Shredder
1/4" strips

Valley Flat Screen
6-cut screen (0.15 mm slots)

Accepts
Rejects

Accepts

Handsheets

Handsheets

Handsheets

Denver Flotation Cell

Full description found at http://www.tlmi.com/recycling-standards.php
Summary

• Removal efficiency of hot-melt PSAs can be predicted from its dynamic mechanical and performance properties.

• Water-based PSAs involve more complex formulations and processing. Removal efficiency is determined by its strength when saturated with water.

• The removal efficiency of a PSA is strongly influenced by the overall laminate design and its processing, e.g., the wet-strength of facestock properties can vary the removal efficiency of an attached PSA film by as much as 60%.

• Test methods have been developed to certify Recycling-compatible adhesives, and RCAs are now commercially available.
Acknowledgements

• Department of Energy
• United States Postal Service
• Mark Kroll (H.B. Fuller)
• Larry Gwin (Franklin International)
• Jennifer Lien (Boise Solution)
• Karen Scallon (FPL)
• Mike Nowak (H.B. Fuller)