The Investigation of Adhesion of Resins Used as Tissue Creping Adhesives for Yankee Dryer Surface Coating

F. Rezaei-Arjomand1∗, A. A. Enayati1, A. H. Lohrasebi2, S. R. Ghaffarian Anbaran3, and S. J. Hashemi4

ABSTRACT

In production of tissue paper, the adhesion of a liquid adhesive creping aid, compound polymer, as creping aids on the Yankee dryer surface was investigated. A compound polymer was prepared based on mixing ratio of poly (amideamine)-epichlorohydrin resin (PAE) as an adhesive agent, polyvinyl alcohol (PVA) as a modifier agent, and cationic fatty acid (CFA) and ethylene glycol (EG) as release agents. In order to simulate the Yankee dryer cylinder, the adherence of paper to the substrate that was coated by the compound polymer solution was measured using a peel strength test. Results indicated that increasing the percent of total solid content of compound polymer would increase peel strength of paper. Peel strength of paper increases if EG as a release agent is used. The amount of PVA/PAE resin in the formulation of creping aid was critical for building a flexible and tough coating layer on to the substrate and to provide adequate adhesion of paper in peel strength test.

Keywords: Dry crepe, Peel strength, Polyvinyl alcohol, Tissue, Wet strength resin.

INTRODUCTION

Competition in tissue market leads to continued efforts to improve the quality of tissue products. Among tissue properties, softness is considered as the most consumer-desired quality-related property (Liu and Hsieh, 2004).

Softness is the tactile sensation a user perceives as the user holds, rubs or crumples a particular paper product. This tactile sensation is provided by a combination of several physical properties including the bulk, stiffness and stretchability of the paper. Creping, a process which is well-known in the art, breaks a large number of fiber-to-fiber bonds in the sheet, in order to enhance the softness, bulk and stretchability of the paper. The amount of adhesion provided by the coating adhesive plays a significant role in the development of these tissue properties (Allen et al., 1998; Neal et al., 2001; Sommese et al., 1994).

Various chemical additives such as poly (amideamine)-epichlorohydrin (PAE) and polyvinyl alcohol (PVA) are utilized to provide thin adhesive coatings on Yankee dryers during the production of creped tissue papers. The ability to crepe the sheet in the desired manner is strongly influenced by the chemistry (types of chemical additives) and

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properties of the coating which is present on the Yankee surface. The coating is a thin layer of organic and inorganic material deposited on the dryer as the process water flushes off. There are two kinds of coating layer: natural coating layer and synthetic coating layer. A natural coating layer contains fibers and fines whose major parts are hemicelluloses (Sloan, 1991, Oliver, 1980). A synthetic coating layer contains polymers (such as PAE and PVA) that are used for completing the natural adhesive.

The occurrence of the natural coating may be supplemented by the addition of synthetic materials, creping aids, sprayed directly on the Yankee surface. Besides the important function of providing the proper amount of adhesion, the coating also serves as a protective role shielding the dryer and creping blade surfaces from excessive wear (Furman et al., 1993).

This creping aid is a compound polymer that includes a mixture of an adhesive agent, a modifier agent and a release agent creating a uniform coating layer on the Yankee surface and to improve the adhesion between paper and Yankee surface (Luu et al., 2001; Neal et al., 2000; Sekiya et al., 2002; Soerens et al., 1985; Sommese et al., 1994).

Considering the importance of creping process in the tissue manufacturing and the role of the coating layer adhesion in crepe quality, this work is aimed at investigating the formulation of creping aid. This formulation is based on PAE resin, the chief component in creping adhesive, as an adhesive agent. The performance of each used agents (adhesive, modifier and release agents) in compound polymer solution is explored by means of a peel strength test. Knowledge of the creping adhesive may help improve the control of crepe in tissue paper.

**MATERIALS AND METHODS**

A compound polymer was prepared based on mixing ratio of poly (amideamine)-epicholorohydrin (Luresin KNU) (PAE) as an adhesive agent, polyvinyl alcohol(GH-17R) (PVA) as a modifier agent, cationic fatty acid (Cartaflex® liquid) (CFA) and ethylene glycol(Merk) (EG) as release agents as follows: PVA (40% to 90%), CFA (2%), EG (5%) at three levels of solid content: 3, 4 and 5% (Table 1).

In order to measure the adherence of paper to substrate, the 90° peel strength test (Furman et al., 1990) by means of Adamel tensile apparatus was carried out according to ISO-5810-1 standard test method (ISO, 1990). Some modifications were made to simulate real conditions in papermaking process. The substrate was a cast iron plate, 5x70x170 mm in dimensions, and 0.2-0.3 micron roughness with the same composition of Yankee cylinders casting. ALBET® filter paper was used as tissue paper.

The substrate surfaces were degreased in liquid degreasing bath of Meta sodium silicate solution. After drying, the plates were heated to 70°C by means of an oven-dryer. There was no change in the color of the plate surface.

A 90 micron thickness of coating layer was applied on the plate by means of an applicator. A wet strip paper (25x185 mm)

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**Table 1.** Mixing ratios of resins constituents in compound polymers (%).

<table>
<thead>
<tr>
<th>Treatments</th>
<th>Resins</th>
<th>Sample</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>T1</td>
</tr>
<tr>
<td>Acid</td>
<td>PVA</td>
<td>90</td>
</tr>
<tr>
<td></td>
<td>PAE</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>CFA</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>PVA</td>
<td>90</td>
</tr>
<tr>
<td>Alcoholic</td>
<td>PAE</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>EG</td>
<td>5</td>
</tr>
</tbody>
</table>
with 60% moisture was placed on the plate which was coated by compound polymer solution. After that, it was rolled with a 5 kg roller to adhere the paper to the plate surface.

According to Yankee surface temperature, the temperature of paper surface adhered to the substrate was increased to 100°C in order to Cross-linking coating layer on the substrate. Then the 90° peel strength test was conducted with a peel speed rate of 430 mm min\(^{-1}\) (Furman et al., 1990).

The hardness of coating layers was measured using Pendulum hardness tester according to ISO 1522, ASTM D 4366-87 standard test method based on the oscillation number and oscillation time.

The thickness of coating layers was measured using a micrometer at five locations, and the mean thickness was calculated.

The treatments were six acid treatments with different blending ratios of PVA/PAE/CFA and six alcoholic treatments with different blending ratios of PVA/PAE/EG (Table 1). Four measurements were performed for each treatment at three levels of solid content. The results obtained from the peel strength, hardness and thickness were analyzed statistically using SAS analyzer software version 7, 1995 (ANOVA method) and Duncan’s Test was used in order to compare mean values. Each result was the mean value of measurements reported in Table 2.

### RESULTS AND DISCUSSION

According to Table 2, the results suggest that by increasing the amount of PAE resin in compound polymer solution along with decreasing the PVA content (from T1 sample to T6 sample in both acid and alcoholic treatments), the peel strength of paper decreases. It could be that blending of two polymers probably leads to an intermolecular interaction based on adsorption theory of adhesion (Kin Loch, 1987) between the two polymers. The molecules in films of high-polymeric materials are held together by secondary valence forces. These forces are weaker than primary valence forces, or direct chemical bonds, and they may be neutralized or blocked fairly easily.

PAE is a thermoset resin which crosslinks upon heating and makes a rigid film. PAE is formed by the reaction between adipic acid and diethylene triamine, and subsequent derivation of the resulting copolymer with epichlorohydrin. The initial polymerization

### Table 2. The average peel strength of paper at three levels of solid content (% of compound polymer solution).

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Sample</th>
<th>Sc= 3%</th>
<th>Sc= 4%</th>
<th>Sc= 5%</th>
<th>Hardness (Oscillation No.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acrylic</td>
<td>T1</td>
<td>60.26</td>
<td>87.89</td>
<td>94.62</td>
<td>140.5</td>
</tr>
<tr>
<td></td>
<td>T2</td>
<td>67.29</td>
<td>77.39</td>
<td>138.5</td>
<td>150.75</td>
</tr>
<tr>
<td></td>
<td>T3</td>
<td>50.67</td>
<td>74.53</td>
<td>150.75</td>
<td>177.25</td>
</tr>
<tr>
<td></td>
<td>T4</td>
<td>50.57</td>
<td>61.87</td>
<td>71.17</td>
<td>141.25</td>
</tr>
<tr>
<td></td>
<td>T5</td>
<td>32.22</td>
<td>57.71</td>
<td>146.25</td>
<td>180.25</td>
</tr>
<tr>
<td></td>
<td>T6</td>
<td>19.98</td>
<td>44.45</td>
<td>158.75</td>
<td>181.25</td>
</tr>
<tr>
<td>Alcoholic</td>
<td>T1</td>
<td>79.63</td>
<td>104.41</td>
<td>115.83</td>
<td>121.25</td>
</tr>
<tr>
<td></td>
<td>T2</td>
<td>71.88</td>
<td>88.81</td>
<td>98.09</td>
<td>112.5</td>
</tr>
<tr>
<td></td>
<td>T3</td>
<td>63.32</td>
<td>67.29</td>
<td>69.84</td>
<td>128.25</td>
</tr>
<tr>
<td></td>
<td>T4</td>
<td>65.15</td>
<td>67.91</td>
<td>73.01</td>
<td>124.5</td>
</tr>
<tr>
<td></td>
<td>T5</td>
<td>55.16</td>
<td>54.24</td>
<td>64.44</td>
<td>124.5</td>
</tr>
<tr>
<td></td>
<td>T6</td>
<td>38.84</td>
<td>43.03</td>
<td>62.91</td>
<td>130.25</td>
</tr>
</tbody>
</table>
during the production of PAE creates a chain very similar to that of Nylon. What's different is that the chain contains secondary amine groups. Some of these react with the epichlorohydrin, and some remain in the product, helping to make it highly cationic and soluble under the conditions of use. The reaction with epichlorohydrin creates highly reactive aminochlorohydrin and azetidinium chloride (ring) groups along the chain. These groups appear to be capable of reacting with either the carboxyl groups at a cellulose surface or amine groups on adjacent molecules of the additive during drying of paper. The groups added to the molecular backbone by reaction with epichlorohydrin are also highly cationic.

PVA is a thermoplastic resin which is applied to compound polymer solution as a plasticizer. Plasticizers are materials which are added to high-polymeric film-formers (such as PAE) to increase the flexibility of their films. It appears that plasticizers neutralize some of the secondary valence forces which bind polymeric molecules together.

On the other hand PVA acts only physically as a molecular spacer between PAE chains. This permits greater freedom of movement of the molecules under stress, which means greater flexibility in the coating (Payne, 1954). According to Hardy, a flexible material is able to better distribute the stresses associated with the peeling action leading to higher peel strength values (Hardy, 1965). Thus, by decreasing the PVA content, the flexibility of the coating layer decreases and lower peel strength is resulted. According to Soerens, the use of PVA and PAE together in a compound polymer solution causes better adhesiveness compared to 100% PVA or 100% PAE (Soerens, 1985).

The measurements of the hardness of coating layers (Table 2) indicated that by decreasing the peel strength, the hardness of coating layer was increased in both acid and alcoholic treatments. Decrease in flexibility of coating layer resulted from reducing PVA content was a reason to increase the hardness of coating layer. Therefore, the hardness of coating layers decrease by increasing PVA content. Allen and other research workers (Allen et al., 1997) have also supported these results.

Upon increasing the PVA content in compound polymer solution, the increased flexibility of the coating layer lowered the hardness and improved adhesive property.

As Figure 1 shows, the use of EG instead of CFA in compound polymer solution as a release agent increased the peel strength. Also the hardness of coating layer was decreased in alcoholic treatments containing

![Figure 1](image.png)

**Figure 1.** The effect of CFA and EG as release agents on peel strength of paper and hardness of coating layer in different treatments.
Figure 2. The effect of solid content on peel strength of paper in different (a) alcoholic treatments, (b) acid treatments.

EG. Another material used in this study was the release agent. Mainly, release agents retard coating sets and soften the coating thereby increasing intimacy. The intimacy factor is described for achieving high uniformity of attachment of the sheet to the substrate surface. As the release agent is added to the compound polymer solution, the coating becomes softer and more pliable, filling the sheet valleys (Stitt, 2002). According to Donnelly, the use of release agents causes an increase in paper softness (Donnelly, 1961). Therefore, EG as a release agent improves the level of adhesion of the coating layer.

Results also suggest that the effect of CFA and EG on peel strength is more significant for samples with more flexibility (more PVA content) or more rigidity (more PAE content).

The effect of solid content on peel strength in both acid and alcoholic treatments is shown in Figures 2-a and b, respectively. Increasing the solid content of the compound polymer solution in two cases increased peel strength of paper through increasing the amount of resin per unit surface. Increasing the amount of solid content with constant thickness of wet coating layer, increases the dry thickness of coating layer. According to Hardy, increasing the thickness of coating layer causes an increase in peel strength (Hardy, 1965).

As it can be seen, the effect of solid content on peel strength is more significant for samples with more flexibility (more
PVA content) or more rigidity (more PAE content). Also the effect of EG with the solid content of 3% and CFA with the solid content of 5% on reducing peel strength by decreasing PVA content is more uniform. Since the use of EG instead of CFA increases the peel strength and, also increase of the solid content holds the same result, it seems that the use of EG with lower solid content or CFA with more solid content can result the same adhesion.

CONCLUSIONS

The results indicated that decreasing the percent of PVA decreases peel strength. Also, increasing the percent of total solid content of compound polymer would increase peel strength of paper. Peel strength of paper increases if EG as a release agent is used. The amount of PVA/PAE resin in the formulation of creping aid was critical for building a flexible and tough coating layer on to the substrate and to provide adequate adhesion of paper in peel strength test.

Considering that PVA is a thermoplastic resin and produces a flexible layer whereas PAE is a thermost resin and produces a glassy layer, it could be concluded that for a given level of adhesion, a compound polymer having comparatively flexible layers and yet sufficient toughness will yield a more uniform adhesion. Thus, a PVA/PAE compound polymer solution showed a better performance as a creping agent than a one-component agent.

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چکیده

در تولید دستمال کاغذی چسبندگی مانع چسبندگی کرب نمی‌تواند مورد بررسی قرار گرفد. پیامدهای ترکیب نسبی از رزین‌پلی‌اکسید و آمین‌اکسید کلرید (PAE) به عنوان عامل چسبندگی، پلی وینیل الکل (PVA) به عنوان عامل اصلی کننده، استردی کاتونی (CA) و اتیل‌گلیکول (EG) به عنوان عامل جداینده تهیه شده‌اند. سپس به منظور شیب‌سازی سیلندر خشک کن یانکی، نیازی به اضافه‌کردن پلیمر ترکیبی بر روی صفحات از جنس سیلیکون مذکور قرار داده شد. با چسبندگی کاغذی بر روی سطح مذکور و با استفاده از آزمون مقاومت به کندن، چسبندگی گمک کننده کرب اتاق‌زد و چسبندگی که افزایش دیده شده و نیاز به افزایش مقاومت به کندن می‌باشد. میزان رزین EG به عنوان عامل جداینده شیب افزایش مقاومت به کندن می‌باشد. میزان رزین EG در فرمول‌سازی گمک کننده کرب در تشکیل لاپ پویشی نرم و مقاوم بر روی سطح صفحه مورد آزمایش به‌طوری که منجر به چسبندگی کافی کاغذی در آزمون مقاومت به کندن شود، از اهمیت ویژه‌ای برخوردار است.