Hydrogenated resin/pure monomer resin blend combinations

*as styrene-butadiene-styrene tackifiers for diaper construction pressure sensitive adhesives*
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Introduction

Styrene-butadiene-styrene (SBS) polymers are commonly used in disposable diaper construction adhesives and are typically formulated with partially hydrogenated hydrocarbon resins such as Eastman’s Regalite™ and Eastotac™ resins for compatibility, improved temperature resistance, thermal stability, and light color characteristics. However, due to recent availability concerns of hydrogenated hydrocarbon resins, Eastman has investigated alternative adhesive formulations using a combination of fully hydrogenated hydrocarbon resins with pure monomer resins (PMRs).

The major advantages of pure monomer resins compared to other resin chemistries include excellent thermal stability, temperature resistance (similar to or better than hydrogenated hydrocarbon resins), low molecular weight, and compatibility with SBS polymer systems when used in synergy with fully hydrogenated hydrocarbon resins. By controlling the amount and type of resins, adhesive formulators can effectively use PMRs in combination with fully hydrogenated hydrocarbon resins to obtain performance similar to adhesives formulated solely with partially hydrogenated resins. Specifically, our tests have shown that a combination of Eastotac™ H-100W resin with a PMR resin can be used in a diaper construction PSA formulation as a replacement for partially hydrogenated resins.

Table 1 shows typical properties of selected hydrogenated hydrocarbon resins and PMRs. Values shown are an average of typical samples and should not be interpreted as product specifications.

<table>
<thead>
<tr>
<th>Resin Type</th>
<th>Resin Type</th>
<th>Chemistry</th>
<th>Ring &amp; ball softening point (°C)</th>
<th>Mn/Mw/Mz (Daltons)</th>
<th>T_g (°C)</th>
<th>Gardner color/YID(^b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Regalite™ S5100</td>
<td>Partially hydrogenated hydrocarbon resin</td>
<td>Partially hydrogenated aromatic resin</td>
<td>97</td>
<td>600/900/1600</td>
<td>50</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Eastotac™ H-100W</td>
<td>Hydrogenated hydrocarbon resin</td>
<td>Fully hydrogenated aliphatic resin</td>
<td>100</td>
<td>450/1000/2150</td>
<td>41</td>
<td>&lt;1/8</td>
</tr>
<tr>
<td>Kristalex™ 3100</td>
<td>PMR</td>
<td>Styrenic/alkyl styrenic resin</td>
<td>100</td>
<td>700/1500/2550</td>
<td>53</td>
<td>&lt;1/5</td>
</tr>
<tr>
<td>Piccotex™ LC</td>
<td>PMR</td>
<td>Alkyl styrenic resin</td>
<td>91</td>
<td>750/1350/2200</td>
<td>46</td>
<td>&lt;1/8</td>
</tr>
<tr>
<td>Piccolastic™ A75</td>
<td>PMR</td>
<td>Styrenic resin</td>
<td>74</td>
<td>700/1300/2250</td>
<td>35</td>
<td>1/-</td>
</tr>
</tbody>
</table>

\(^a\) Obtained from Eastman publication WA-86A, Spectrum of Hydrocarbon Resins; WA133, Eastman Hydrocarbon resins
\(^b\) YID = Yellowness Index
Technical discussion

An SBS-based disposable diaper construction adhesive formulation was prepared with Regalite™ S5100 partially hydrogenated hydrocarbon resin as the control. The effects of three different PMRs (Kristalex™ 3100, Piccotex™ LC, and Piccolastic™ A75) in combination with Eastotac™ H-100W fully hydrogenated hydrocarbon resin were evaluated at two different extension amounts; 12% by weight and 18% by weight in formulation, which equates to 20% and 30% PMR with respect to total resin amount (Eastotac™ H100W:PMR, 80:20 and 70:30 ratios). Table 2 shows the formulations used in this study.

Table 2. Disposable diaper construction adhesive formulations with PMRs

<table>
<thead>
<tr>
<th>Materials wt%</th>
<th>Regalite™ S5100 control</th>
<th>Kristalex™ 3100 80:20 H100W:3100</th>
<th>Kristalex™ 3100 70:30 H100W:3100</th>
<th>Piccotex™ LC 80:20 H100W:LC</th>
<th>Piccotex™ LC 70:30 H100W:LC</th>
<th>Piccolastic™ A75 80:20 H100W:A75</th>
<th>Piccolastic™ A75 70:30 H100W:A75</th>
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</thead>
<tbody>
<tr>
<td>Kraton™ 1102a</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Regalite™ S5100b</td>
<td>59.5</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Eastotac™ H-100Wb</td>
<td>-</td>
<td>47.5</td>
<td>41.5</td>
<td>47.5</td>
<td>41.5</td>
<td>47.5</td>
<td>41.5</td>
</tr>
<tr>
<td>Kristalex™ 3085b</td>
<td>-</td>
<td>12</td>
<td>18</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Piccotex™ LCb</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>12</td>
<td>18</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Piccolastic™ A75b</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>12</td>
<td>18</td>
</tr>
<tr>
<td>Calmeol™ 5550c</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Irganox™ 1010d</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Formulations were prepared utilizing a Brabender mixer and evaluated for the following:

- Viscoelastic properties—Using Dynamic Mechanical Analysis (DMA)
- Viscosity profiles at different temperatures (before and after aging [177°C for 72 h])—Brookfield viscometer
- Adhesive property evaluation—1-mil-thick adhesive coating on Mylar™
- 180 degree peel on stainless steel substrate (peel rate @ 4 inches/min)
- Loop tack on stainless steel substrate
Figures 1a and 1b show the viscoelastic performance analysis of the Eastotac™ PMR combination formulations at two different levels, along with the partially hydrogenated hydrocarbon resin (Regalite™ S5100) control formulation.

**Figure 1.** Viscoelastic properties of formulations with Eastotac™/PMR combinations

(a) Formulations extended with 12 wt% PMRs (80:20 H100W:PMRs).

(b) Formulations extended with 18 wt% PMRs (70:30 H100W:PMRs).
As can be seen in Figure 1b, 70:30 blends of Eastotac™ H100W and Piccotex™ LC resin and 70:30 blends of Eastotac™ H100W and Kristalex™ 3100 resin formulations showed the most comparable T\(\chi\) and elastic modulus to the control formulation. The single T\(\chi\) of the Eastotac™/PMR resin combination formulation is an indication of compatibility with the SBS polymer. All of the 80:20 Eastotac™/PMR resin containing formulations showed slightly different viscoelastic characteristics than the control.

Figure 2 shows the absolute values of important parameters such as T\(\chi\), Tan \(\delta\) minimum value at Tan \(\delta\) minimum temperature (an indication of cohesive property), 3rd crossover temperature (indication of melting temperature), and Tan \(\delta\) peak value at T\(\chi\) (indication of tack). As can be seen from Figure 2, 70:30 blends of Eastotac™ H100W and Piccotex™ LC and 70:30 blends of Eastotac™ H100W and Kristalex™ 3100 formulations showed the most comparable T\(\chi\), Tan \(\delta\) minimum temperature, Tan \(\delta\) peak value and 3rd crossover temperatures. Figures 3 and 4 show the adhesive property evaluation results, which were performed with 0.9–1-mil-thick Mylar-coated films on a stainless steel substrate.
Figures 3 and 4 show the 180 degree peel adhesion loop tack results on a stainless steel substrate respectively. A combination of 20% PMR with Eastotac™ H100W resins shows lower adhesive peel and loop tack compared to the control formulation. However, a combination of 30% PMR with Eastotac™ H100W resin shows comparable adhesive peel but lower loop tack compared to the control formulation. The lower tack performance may be due to the structural differences between Regalite™ S5100 and Eastotac™ H100W/PMR combination resins. In the case of Regalite™ S5100, aliphatic and aromatic moieties are within one chain backbone compared to the fully hydrogenated/PMR resin combinations, which are a physical blend of aliphatic and aromatic moieties. Even though fully hydrogenated/PMR resin combination-based formulations exhibit similar viscoelastic characteristics compared to the control formulation, the combination of resins may have some macrophase separation limiting the total molecular mobility of the chains which results in lower loop tack. However, it should be noted that tack is not an important criterion for diaper construction adhesives, since it is mostly a one-time adhesive application process (no bonding and debonding as in PSA tapes or labels, where tack is very important).

As can be seen from the viscosity profiles (Figure 5) at five different temperatures, the viscosity characteristics for Eastotac™/PMR resin combination formulations at both PMR levels (20% and 30%) are similar or comparable to the control formulation, even after aging at 177°C for 72 h.
Figure 6 shows the pictures of the formulation before and after aging. Aging was performed in an air-drafted oven at 177°C for 72 h.

As clearly depicted in Figure 6, the before and after aging characteristics, especially in regards to color, for the Eastotac™/PMR combination resin formulations are similar or better than the control formulation.

Conclusion

The combination of fully hydrogenated hydrocarbon resins such as Eastotac with a pure monomer resin can be used as a tackifier in SBS-based adhesives used in nonwoven PSA formulations as a replacement for partially hydrogenated resins.

- 70:30 Eastotac™ H100W:Piccotex™ LC combination and 70:30 Eastotac™ H100W:Kristalex™ 3100 resin combination showed the most comparable performance.
- Similar viscoelastic characteristics and adhesive peel
- Good thermal stability on aging—both color & viscosity stability
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