

Evaluation of Disposable Diaper Construction Hot Melt Pressure-Sensitive Adhesives Using Blends of Olefinic Block Copolymer and Amorphous Polyolefin Polymers—Adhesive Performance, Sprayability, and Viscoelastic Characteristics

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ABSTRACT: Pressure-sensitive adhesives (PSAs) used in disposable diaper construction have been formulated using blends of olefinic block copolymer (OBC) and an ethylene-propylene (PE-PP) amorphous polyolefin (APO) polymer, with three different unsaturated hydrocarbon resins (with varying aromatic content), and also with two different saturated aliphatic hydrocarbon resin (with varying cycloaliphaticity). The viscoelastic properties of these PSA formulations were studied using dynamic mechanical analysis (DMA). Viscosity profiles at five different temperatures were generated to better understand the application window for the resulting adhesive formulation. Rheology master curves were generated using time-temperature-superposition analysis and correlated with the processability characteristics. Adhesives used in disposable diaper construction were applied between a polyethylene backing and a nonwoven substrate with an air-assisted spiral spray application technique on an Acumeter Spray Coater. After the adhesive was applied, peel adhesion testing on the samples was performed. It has been observed that the OBC/PE-PP-based disposable diaper construction PSA has a lower application temperature along with wider tolerance for hydrocarbon resin chemistries, especially for the saturated aliphatic resin-based PSA formulations. Based on the coating parameters used, it has been learned that the adhesive formulations seem to show a higher shear rate at the nozzle, but Reynolds number calculated indicated no major turbulence occurring at the nozzle during spraying. Very good spray patterns were obtained for the olefinic polymer-based PSA formulations. Disposable diaper construction article showed good adhesive peel properties, especially for the adhesive formulations containing saturated aliphatic hydrocarbon resin, which were comparable to the SBS-based control. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 130: 3311–3318, 2013

KEYWORDS: adhesives; blends; polyolefins; resins; viscosity; viscoelasticity

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INTRODUCTION

Since the advent and commercialization of Pampers® by P&G in the late 1950s, baby diapers, feminine hygiene care products, and adult incontinent undergarments have been an indispensable part of human life. Availability, cost, usefulness, and affordability of the hygiene care products for the global human population has also created lots of challenges for the hygiene suppliers, especially in terms of cost, performance improvements, product design, and assembly processes.^{1,2} Diaper products are the largest volume of the disposable hygiene market.² Even though a baby diaper looks very simple in appearance, the components and assembly is rather complicated and can be seen in Figure 1. Hot melt adhesives are a major component in the diaper assembly and are used to bond different parts of the diaper together. Two kinds of hot melt adhesives are usually

employed in a diaper assembly, namely, (1) a hot melt pressure-sensitive construction adhesive used to bond the plastic and nonwoven fabric and (2) an elastic attachment adhesive that has higher elasticity and bonding strength to bond the legs and waist foam elastics with the plastic and nonwoven fabric.

Even though the early hot melt adhesive assembly technologies were based on ethylene vinyl acetate-based hot melt adhesives typically applied with slot-die coaters, the advent of styrenic block copolymer-based hot melt adhesive formulations³ emerged as the prominent hot melt adhesive of choice for the disposable hygiene articles since 1980s because of the versatility in product assembly and performance advantages.^{2,4–7} The next notable breakthrough in the pressure-sensitive adhesives (PSAs) for disposable hygiene article technology came through a newly developed hot melt application technology in the 1980s by

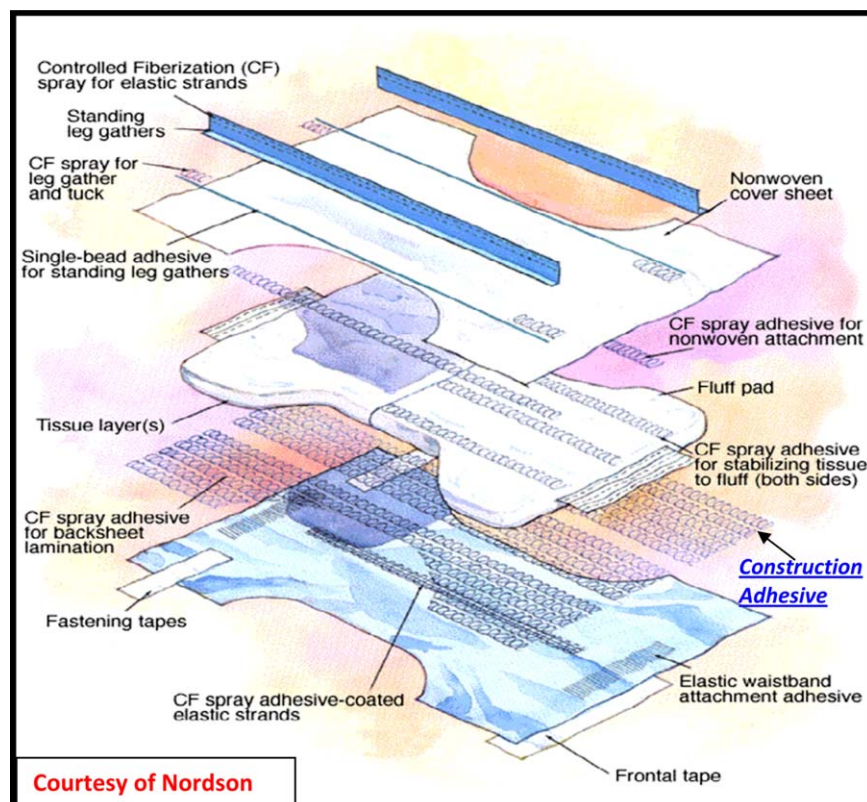


Figure 1. Different components of a disposable baby diaper. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Nordson known as hot melt PSA air-assisted spray technology.⁸ As Raterman of Nordson reported,⁸ this air-assisted spray technology drastically enhanced the refinement of controlled fibrillation spray that resulted in more control over adhesive application amount, faster line speeds, and use of thinner substrate materials (polyethylene and nonwoven fabric) resulting in superior economic advantages. Since then styrene block copolymer-based PSA systems with air-assisted spray technology configurations (especially spiral spray patterns) has emerged as the workhorse adhesive system and application method of choice for disposable hygiene articles. Unfortunately, there is only limited published literature available on the art of formulation and performance characteristics of disposable diaper construction adhesives. Much of the art of formulating diaper construction PSAs were considered a trade secret to some of the global adhesive manufacturers and was only available through some patents.

The raw material availability dynamics, cost, in conjunction with performance improvements has led to significant advancement in the design, shape, assembly, and process of disposable hygiene products.¹ As Fornes¹ pointed out, PSAs used in the disposable hygiene articles are one of the major means to achieve the cost savings, and process and performance enhancements. Styrenic block copolymer [especially styrene-butadiene-styrene (SBS) and styrene-isoprene-styrene (SIS)]-based PSAs are still the workhorse PSA formulations for disposable hygiene applications (especially diaper). Styrenic block copolymers have

a glass-transition temperature (T_g) that is too low and a shear modulus that is too high at room temperature, and thus the properties of styrenic block copolymers as such is not suitable for most PSA applications. Previous studies have shown that viscoelastic processes and surface wetting characteristics of a PSA can only be activated if the T_g is close to the application temperature. Therefore, for PSA applications, styrenic block copolymers are always formulated with tackifier resins and plasticizers to achieve the surface wetting characteristics, coating rheology/viscosity requirements, and final adhesive properties.⁹ Figure 2 shows the typical viscoelastic window [T_g and storage modulus (G')] for different PSA applications (tapes, labels, and hygiene PSAs).

Sheriff et al.,¹⁰ Kraus et al.,^{11,12} and Class and Chu^{13,14} correlated the viscoelastic properties of PSAs containing tackifier resin with adhesive properties and reported that tackifier resin structure, molecular weight, and concentration have significant influence in determining the viscoelastic characteristics of the PSA and thus, the adhesive properties.¹⁰⁻¹⁴ The surface energy characteristics and viscoelastic behavior of a PSA formulation determines the coating application parameters and final adhesive performance characteristics.¹⁵ Although there is some reported literature on hot melt PSAs based on styrenic block copolymers, recent styrenic block copolymer availability issues are forcing more and more formulators to look for alternate technologies, especially olefinic polymer-based PSAs. In recent years, polyolefin-based PSA technology has been getting a lot of

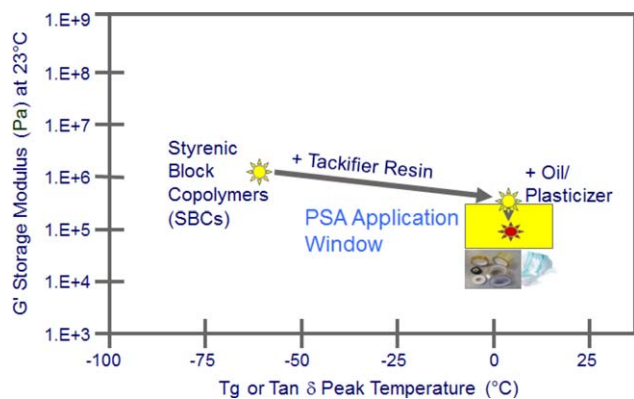


Figure 2. T_g and storage modulus (G') window for typical PSA applications. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

attention because of cost and availability.²⁴ Yuan et al.¹⁶ studied the effect of hydrogenated tackifier resins on amorphous polyolefin (APO)-based PSAs and revealed that tackifiers can increase the T_g and reduce the shear modulus into PSA region. However, one of the biggest disadvantages of most of the APO (atactic or random copolymers)-based PSAs is the lack of elastomeric character resulting in lower peel and cohesive properties of the final adhesive compared to the styrenic block copolymer-based PSAs. Thus, since 1974, most of the commercially available APO-based PSAs are blends of APOs with styrenic block copolymers^{17,18} or blends with polyisobutylene.^{17,19,20} In 2006, Arriola et al.²¹ reported an olefinic block copolymer (OBC) based on copolymerization of ethylene with octene, using a novel chain-shuttling polymerization process. The chain-shuttling catalyst agent promotes a blocky polymer structure that combines the attributes of high-density polyethylene with an olefin elastomer. Even though there have been several polyolefin-based polymers, this polymer is of particular interest because of its similarity in rheological performance characteristics to some of the styrenic block copolymers used in hot melt PSA applications.^{22–24}

In 2007, Shan et al.²⁴ first reported PSAs using high-melt index (15 and 21 at 190°C, 2.16 kg) developmental OBCs based on ethylene and octene. Even though these developmental OBCs had similar T_g to SIS block copolymer (15 wt % styrene), they had comparably high G' with higher slope (temperature ramp) than SIS block copolymer (15 wt % styrene). Thus, the resulting PSA formulation with developmental OBCs showed high stiffness and required significant amount of oil and tackifier, resulting in a soft adhesive with inferior adhesive performance compared to the SIS block copolymer (15 wt % styrene)-based formulation.²⁴ Recently, Dow has commercialized a five melt index (190°C, 2.16 Kg), 0.866 g/cm³ density ethylene–octane-based OBC. The G' for this OBC is still higher than the typical styrenic block copolymer and correlates well with reported developmental OBCs.²⁴ We believe that blending this polymer with APO polymers will increase the T_g and reduce the G' , resulting in similar G' to a styrenic block copolymer. Further, modifying this blend with tackifier resins and oil will make a PSA with comparable adhesive properties to styrenic block copolymers for use in disposable diaper construction adhesive.

In this study, a disposable diaper construction PSA was formulated using OBC/APO (especially PE–PP) olefinic copolymer blend polymer system containing an unsaturated or saturated hydrocarbon resin. This may provide adhesive raw material sustainability, lower adhesive application temperatures with improved bond strength for the current disposable hygiene industry compared to the standard SBS-based disposable diaper construction adhesive, which Fornes¹ points out as one of the major focus areas. Blends of OBC/PE–PP containing three different unsaturated hydrocarbon resins with varying aromatic content and two different saturated aliphatic hydrocarbon resins with varying cycloaliphaticity were evaluated in a typical disposable diaper construction adhesive formulation. Viscoelastic characterization of the adhesive has been performed using dynamic mechanical analysis (DMA). Viscosity and rheology characteristics has also been studied and correlated to coating/sprayability performance. Adhesive peel has been evaluated on final coated samples and compared against current styrenic block copolymer-based control adhesive (SBS control). To our knowledge, there has not been any reported literature regarding the use of OBC/APO blends for disposable diaper construction adhesives.

MATERIALS AND METHODS

A commercially available (INFUSE 9507[®]) five melt index (190°C, 2.16 Kg), 0.866 g/cm³ density ethylene–octane-based OBC was obtained from Dow Chemical Company. Atactic ethylene–propylene APO copolymer was obtained from Eastman Chemical Company. Properties of the APO (atactic ethylene–propylene copolymer) are given in Table I.

Properties of the hydrocarbon resins selected for this study are given in Table II. Ring & Ball Softening Point of hydrocarbon resin was measured using the Herzog Ring & Ball Tester. The softening point is defined as the temperature at which a disk of the sample held within a horizontal ring is forced downward a distance of 25.4 mm (1 in) under the weight of a steel ball as the sample is heated at 5°C/min in a silicon bath (400 mL). The temperature is recorded, when the resin sample passes through the sensors of the unit (ASTM D-6493-99).

To determine the aromatic hydrogen content of each hydrocarbon resin, the ratio of the integration area of aromatic hydrogen relative to the total integration area of hydrogen on the resin's nuclear magnetic resonance (NMR) spectrum was determined via ¹H NMR analysis. The NMR analysis was performed using a JEOL 600 MHz Eclipse NMR system with a pulse interval of 15 s, acquisition time of 3.6 s, pulse angle of 90°, X resolution of

Table I. Properties of Polymers

Name	Penetration hardness (ASTM D5)	Viscosity (190°C) mPa.S (ASTM D3236)	T_g (°C)
PE-PP (atactic ethylene-propylene copolymer)	35	5700	–20

Table II. Properties of Hydrocarbon Resins

Resin	Type	Ring and ball softening point (°C)	Percent aromatic content (NMR)	Molecular weight Mn/Mw/Mz (Da)
Resin 1 (R1)	Aliphatic	95	0.5	800/1700/3500
Resin 2 (R2)	Aliphatic/aromatic	95	5	850/2200/5500
Resin 3 (R3)	Aliphatic/aromatic	95	14	800/1700/4000
Resin 4 (R4)	Cycloaliphatic	92	<0.1	500/700/1100
Resin 5 (R5)	Linear aliphatic–cycloaliphatic	100	<0.1	450/1000/2300

0.27 Hz, and number of scans set at 16. The resin NMR samples were prepared by dissolving a known amount of each of hydrocarbon resins in methylene chloride-d₂. The total integration value was normalized to 100. The results were reported in area percent.

Molecular weights (Mn, Mw, and Mz) of hydrocarbon resins were determined via gel-permeation chromatography (GPC) with tetrahydrofuran (THF) as a solvent. Each resin was analyzed at ambient temperature in Burdick and Jackson GPC-grade THF stabilized with butylated hydroxytoluene (BHT), at a flow rate of 1 mL/min. Sample solutions were prepared by dissolving about 50 mg of each resin in 10 mL of THF and adding 10 μ L of toluene thereto as a flow-rate marker. An auto sampler was used to inject 50 μ L of each solution onto a Polymer Laboratories PLgelTM column set consisting of a 5 μ m Guard, a Mixed-CTM, and an OligoporeTM column in series. The eluting polymer was detected by differential refractometry, with the detector cell held at 30°C. The detector signal was recorded by a Polymer Laboratories CaliberTM data acquisition system, and the chromatograms were integrated with software developed at Eastman Chemical Company. A calibration curve was determined with a set of 18 nearly monodisperse polystyrene standards with molecular weight from 266 to 3,200,000 g/mole and 1-phenylhexane at 162 g/mole. The molecular weight distributions and averages were reported as equivalent polystyrene values.

A typical PSA formulation for disposable diaper construction based on SBS contains 20–25 wt % SBS polymer (20–40% styrene content), 55–60 wt % hydrocarbon resin, and 20–25 wt % oil [2]. The higher resin content is required to obtain the necessary adhesive properties such as tack and peel. Typically, 20–25 wt % oil is added to control the viscosity of the total formulation so that it can be sprayed using air-assisted hot melt

spraying techniques. An SBS-based PSA formulation for disposable diaper applications containing 20/60/20 Kraton[®] D1102 (SBS)/100°C softening point cycloaliphatic–aromatic hydrocarbon resin/naphthenic oil was evaluated as a control. The control adhesive formulation was blended with a Plasticorder Brabender at 150°C using sigma blades. Polymer was initially masticated for 10 min with antioxidant before adding resin and oil. Solid resins and oils were then added to the masticated polymer and blended for 20–45 min until the torque became constant.

Calsol[®] 5550 naphthenic oil was obtained from R.E. Carroll industries and antioxidant Irganox[®] 1010 was obtained from BASF. The OBC/PE–PP-based disposable diaper construction PSA formulations as shown in Table III were produced using mechanical agitation (Paddle type agitator controlled by a variable speed motor) in pint-sized cans with a heat block set at 177°C. Polymer and antioxidant were introduced into the can and heated up to 177°C under a nitrogen blanket. Resin followed by oil was then introduced into the can after the polymer was melted. This was agitated for 30 min until the mixture was completely homogeneous. After thorough mixing, the adhesive was poured into a silicone-lined cardboard box and allowed to cool. Disposable diaper construction PSA formulations evaluated are given in Table III.

Viscoelastic properties of the PSAs were evaluated using DMA. DMA of the blends was performed using a TA Instruments Ares RDA3[®] Rheometer in a parallel plate geometry. The diameter of the plates was 8 mm and the gap was set at 2.33 mm. Temperature sweep experiment was performed between –80°C and 300°C with a heating rate of 6°C/min, by keeping the frequency at 10 Hz and the maximum strain at 5%.

Viscosity profiles and rheological master curves were generated to determine the processability characteristics of the disposable

Table III. Disposable Diaper Construction PSA Formulations with OBC/PE–PP Blends in wt %

Sample name	Formulation description	OBC	PE–PP	Resin	Calsol [®] 5550	Irganox [®] 1010
Disposable diaper construction PSAs with unsaturated hydrocarbon resins						
F1	With resin R1	12.5	12.5	54 (R1)	20	1
F2	With resin R2	12.5	12.5	54 (R2)	20	1
F3	With resin R3	12.5	12.5	54 (R3)	20	1
Disposable diaper construction PSAs with saturated hydrocarbon resins						
F4	With resin R4	12.5	12.5	54 (R4)	20	1
F5	With resin R5	12.5	12.5	54 (R5)	20	1

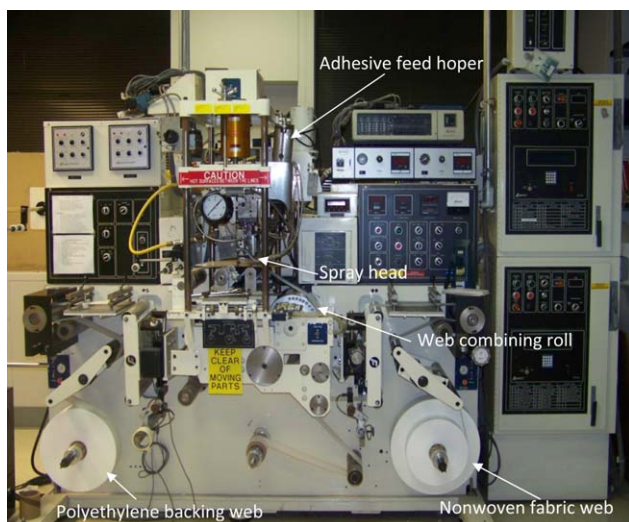


Figure 3. Acumeter Spray Coater hot melt PSA-spraying equipment. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

diaper PSA. Viscosity measurements were carried out using a Brookfield viscometer (DV II, spindle #27) over a range of temperatures (130°C, 150°C, 170°C, 190°C, and 210°C). Rheology master curves were generated using time-temperature superposition (TTS) analysis using Rheumatics RDA3 instrument. The dynamic mechanical test was conducted on a Rheumatics RDA3 using a 25-mm diameter parallel plates with a 1 mm gap setting. The dynamic frequency sweep was run at a frequency range from 1 to 400 rad/s with 10% strain amplitude starting at 180°C. The frequency sweep was repeated while the temperature was decreased at an interval of 10°C down to 25°C. A master curve was generated by using the built-in software Orchestra (Rheometrics, Inc.) on the basis of the TTS principle. The individual curves were shifted by a two-dimensional optimization scheme with guessing mode of calculation and linear interpolation algorithm.

Disposable diaper construction PSA formulations shown in Table III were evaluated for adhesive peel strength after the adhesive has been applied between a nonwoven fabric and PE backing (using Acumeter Spray Coater). Spiral spray adhesive patterns were created using an Acumeter Spray Coater, air-assisted spraying equipment, which is shown in Figure 3.

The hot melt adhesive is melted in the 500 mL feed hopper at a controlled temperature. The adhesive is then pumped with a gear pump through a nozzle, which is designed to use direct spray air to create a high-frequency pattern of hot melt adhesive. The spray air draws the adhesive into a fine fiber. This directed air causes the monofilament to spiral and cool as it is dispensed, delivering a highly consistent helical pattern as shown in Figure 4. The run speed (X) for the Acumeter Spray Coater was set at 350 ft/min (1.78 m/s) to obtain a targeted coat weight (C) of 6 gsm (grams per square meter) and also to keep the open time of the adhesives constant. The nozzle temperature was controlled between 149°C and 163°C and the spray head temperature was controlled between 163°C and

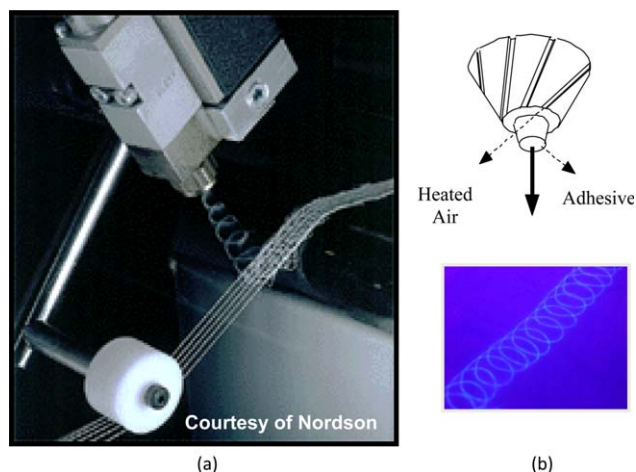


Figure 4. (a) Disposable diaper adhesive spraying for an elastic attachment adhesive (for illustration), (b) a typical air-assisted spraying head configuration in Acumeter Spray Coater spraying equipment and the resulting spiral spray patterns of PSA (under UV light). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

177°C. The width of the spiral patterns (W) was controlled to 0.5 in (12.7 mm), and the number of spiral loops within an inch was controlled to 5–8 loops/in (5–8 loops in 12.7 mm) as shown in Figure 5. After the adhesive was applied between the nonwoven fabric and PE back sheet, the peel adhesion testing on the samples was performed using Instron.

RESULTS AND DISCUSSION

The viscoelastic properties of disposable diaper construction PSAs based on OBCs and a comparative SBS-based control is given in Figure 6. As can be seen (Figure 6), the viscoelastic response for OBC/PE-PP-based PSA is different than that of the typical SBS-based control formulation. The PSA formulation (F1) containing unsaturated aliphatic resin (R1) and PSA formulation (F2) containing unsaturated slightly aromatic (5%)

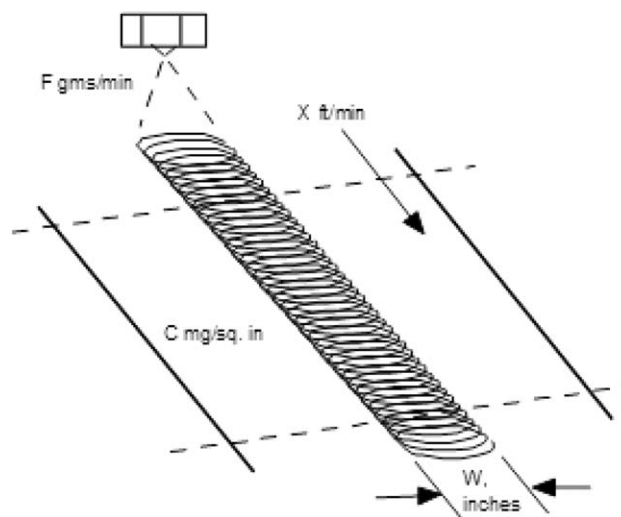


Figure 5. Disposable diaper construction adhesive spiral pattern measurements.

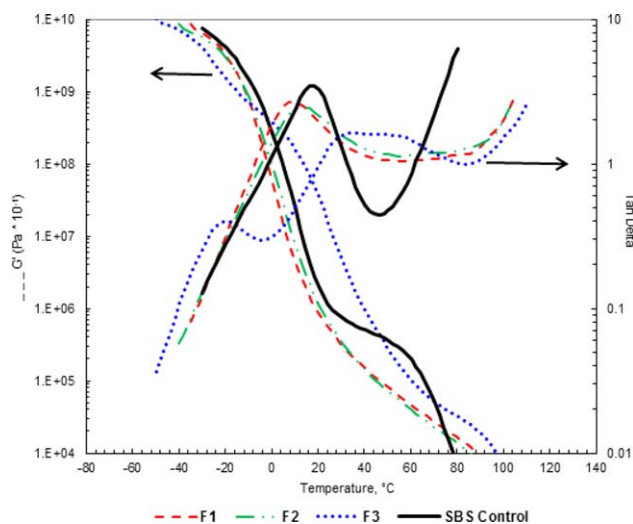


Figure 6. Disposable diaper construction PSAs formulations based on OBC and a comparative SBS-based control. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

resin (R2) PSAs show a single broad T_g . However, the PSA formulation (F3) containing highly aromatic (14%) unsaturated resin (R3) PSA shows two T_g 's, which is a clear indication of immiscibility. The first transition is around -20°C and a second broad transition around 42°C , which corresponds to the highly aromatic unsaturated resin (R3) T_g . Both olefin-based PSA formulations (F1 and F2) showed predominantly flow properties and less elastic characteristics which can be observed from storage modulus (G'). The storage modulus plateau (from room temperature up to 55°C) representing the elastic characteristics of SBS-based control over a wide application temperature range is very clear (Figure 6). The shape of the storage modulus plateau typically represents the strength of the adhesive over a measured temperature range. After the plateau, the adhesive will start to flow. Figure 7 shows the viscoelastic properties of disposable diaper construction PSAs with saturated hydrocarbon

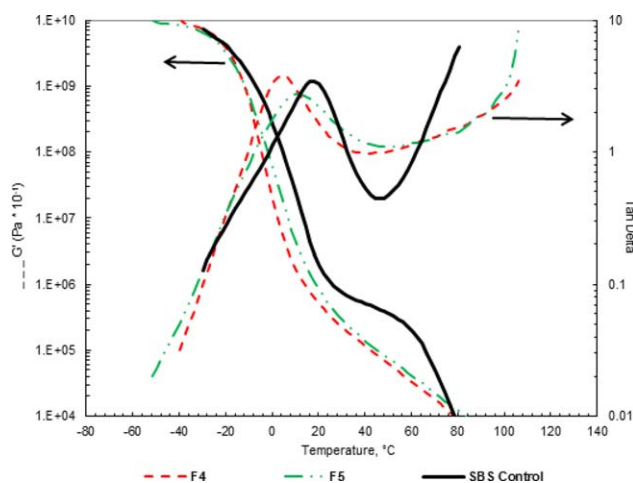


Figure 7. Disposable diaper construction PSAs with saturated hydrocarbon resins and comparative SBS-based control. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

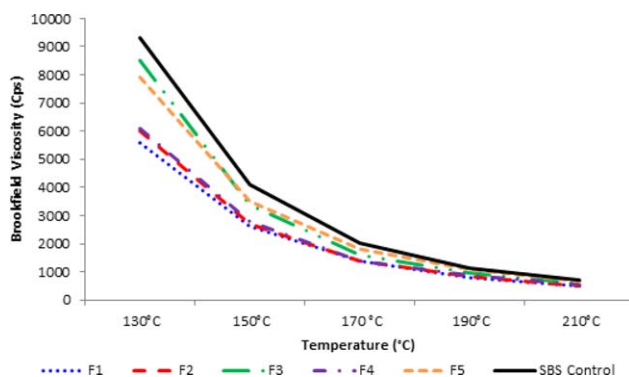


Figure 8. Viscosity profiles of disposable PSAs with hydrocarbon resins at five different temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

resins and comparative SBS-based control. The PSA formulation (F4) containing cycloaliphatic resin (R4) shows a lower T_g , narrower $\tan \delta$, and slightly lower modulus. The slightly lower T_g can be correlated to the 10°C lower softening point of the cycloaliphatic resin compared to both linear aliphatic–cycloaliphatic resin used in formulation F5, and the comparative SBS control. However, the elastic modulus plateau representing the strength of the adhesive bond was still not clear for the OBC/PE–PP with saturated aliphatic resin-based PSA formulations, compared to the SBS-based control. Figure 8 shows the viscosity profiles at five different temperatures for the disposable diaper PSA formulations.

As can be seen (Figure 8), all OBC/PE–PP blend-based disposable diaper formulation containing hydrocarbon resins show lower viscosity profiles at all five temperatures evaluated compared to the SBS-based control adhesive. This is a clear indication that these adhesive formulations can be applied at lower application temperatures. The PSA formulation containing unsaturated aliphatic resin (R1), slightly aromatic unsaturated resin (R2), and cycloaliphatic saturated resin (R4) shows the lowest viscosity profiles, which are almost half the viscosity compared to the SBS-based control, and they are virtually the same from 150°C to 210°C . Better miscibility of the different adhesive formulations component can also be correlated well to the lower viscosity of the same blends. The immiscibility behavior of PSA formulation containing highly aromatic unsaturated resin (R3) may be correlated to the higher viscosity profiles, which are closer to the linear aliphatic–cycloaliphatic saturated resin (R5) and also to the SBS-based control.

Figures 9 and 10 shows the rheological profiles generated using TTS experiments for the adhesive formulations. As can be seen (Figure 9), the complex viscosity (η^*) at different frequencies shows shear thinning behavior. To understand flow characteristics during coating operation on the Acumeter coater, shear rate at the coater nozzle and Reynolds numbers were calculated using eqs. (1) and (2), respectively.^{25,26} The rheology profiles for each adhesive and the Acumeter coater parameters are as follows: Diameter of nozzle (D) = 0.0457 cm [radius (R) = 0.0229 cm], width of spirals = 0.127 m (0.5 in), line speed = 350 ft/min (1.778 m/s), coating weight = 6 gsm, melt density

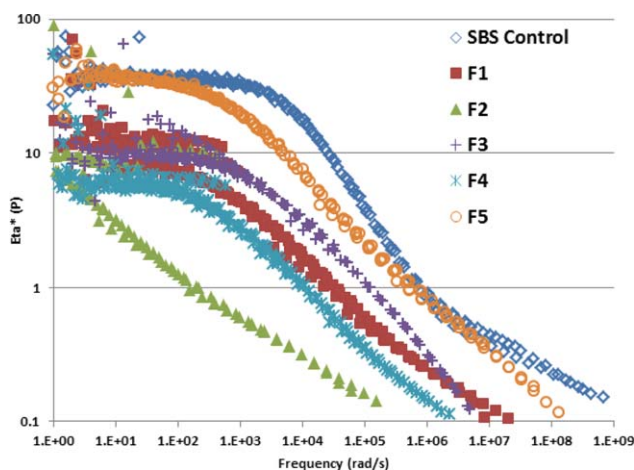


Figure 9. Complex viscosity (η^*) profiles of disposable PSAs with hydrocarbon resins from TTS experiments at different frequencies. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

$= 0.78 \text{ g/cc}$,^{25,26} approximate Newtonian melt viscosity = 3–35 poise, power law index $n = 0.3$, and mass flow rate = 0.1355 g/s (volume flow rate $Q = 0.1129 \text{ cc/s}$), where n = power law index estimated from the slope of the shear thinning region of viscosity profiles in Figure 9.

$$\text{Shear rate (Power law)} = \left(\frac{4Q}{\pi r^3} \right) \frac{3n+1}{4n} \quad (1)$$

$$\text{Reynolds number (non-newtonian)} = \rho D \frac{u^{n-2}}{K \left(\frac{3n+1}{4n} \right)^n (8n-1)} \quad (2)$$

The calculated shear rate for all adhesives at nozzle is between $20,000$ and $30,000 \text{ s}^{-1}$. Reynolds number is calculated to be less than 500 . As Macosko²⁷ points out, the critical Reynolds number for the onset of turbulence in the tube flow is 2100 , and in our case, the calculated Reynolds number is less than 500 for all the adhesive formulations. This means that there is no major

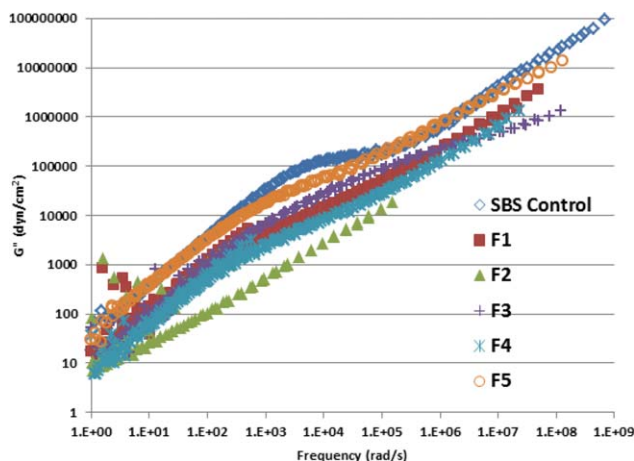


Figure 10. Loss modulus (G'') profiles of disposable PSAs with hydrocarbon resins using TTS experiments at different frequencies. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

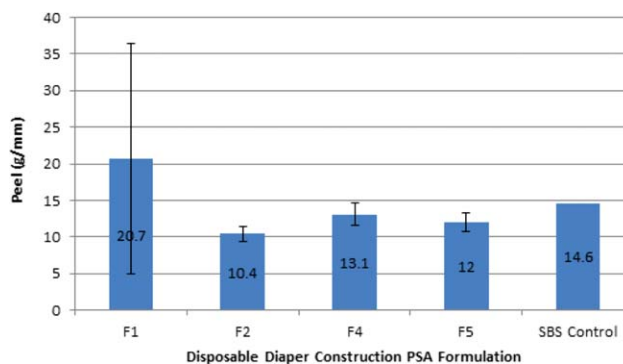


Figure 11. Adhesive peel of disposable diaper construction PSAs with hydrocarbon resins. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

turbulence occurring at the nozzle and should provide very good spiral spray patterns.

Because we have seen immiscibility behavior with PSA formulation (F3) containing highly aromatic (14%) unsaturated resin, we did not proceed with this formulation any further. We eliminated this particular formulation (F3) from spraying (using Acumeter Spray Coater). PSA formulations including, F1, F2, F4, F5, and the SBS control formulations were applied between a nonwoven fabric and PE backing using Acumeter Spray Coater to make a diaper construction-like article, employing the same spraying conditions. It was able to get very good spiral spray patterns with all four OBC/PE–PP-based adhesive formulations. After the adhesives were applied between the nonwoven fabric and PE back sheet, the peel adhesion testing on the samples was performed.

The peel adhesion results are shown in Figure 11. It can be seen that PSA formulation containing saturated cycloaliphatic resins (F4) gave the best adhesive peel strength followed by a formulation containing a saturated, linear aliphatic–cycloaliphatic resin (F5). Both of these formulations show similar adhesive performance compared to the SBS-based control formulation. Even though the PSA formulation containing unsaturated aliphatic resins (F1) showed higher peel strength, the standard deviation was very high, making this an inconsistent adhesive. The main reason for a very high standard deviation can be because of the insufficient penetration of this particular adhesive when sprayed on to polyethylene backing, resulting in variation of bond strength between the PE and nonwoven fabric at different areas of the diaper construction article. PSA formulation containing slightly aromatic unsaturated resin (F2) showed the lowest peel strength of all the formulations. Peel adhesion results (Figure 11) can be correlated to loss modulus (G'') characteristics (Figure 10) as explained by Chu,²⁸ Shih and Hamed.²⁹ Formulations with high G'' at higher frequencies ($>100 \text{ Hz}$) corresponds to higher peel strength^{28,29} (Figure 10) and correlates with the peel results (Figure 11).

The main advantages of the OBC/PE–PP-based disposable diaper construction adhesive, compared to standard SBS-based comparative formulation is that, the OBC/PE–PP-based PSA formulations can be applied at lower applications temperature

(from viscosity profiles), and these formulations also show a big formulation latitude with very good tolerance for different hydrocarbon resin chemistries (especially saturated aliphatic and slightly aromatic–aliphatic unsaturated resins), with most of them giving good adhesive performance properties.

CONCLUSIONS

- Viscoelastic performance of the OBC/PE–PP-based adhesive formulations revealed that these formulations show predominantly flow characteristics than elastic performance as measured via Storage Modulus. The adhesive formulation (F3) containing highly aromatic (14%) unsaturated hydrocarbon resin showed some immiscibility.
- Disposable diaper construction adhesives based on OBC/PE–PP blends can be potentially applied at lower temperatures than that of the comparative SBS control formulation because all the OBC/PE–PP-based adhesives show lower viscosity profiles at a wide temperature range, irrespective of the resin chemistry.
- OBC/PE–PP blend-based disposable diaper construction adhesives showed good sprayability characteristics, when applied using an air-assisted spiral spray equipment, Acumeter Spray Coater.
- Peel adhesion evaluations of the disposable diaper construction article showed good adhesive peel properties, especially for the adhesive formulations containing saturated aliphatic hydrocarbon resin, which were comparable to the SBS-based control.
- It can be concluded that OBC/PE–PP-based PSA formulations can be applied at lower applications temperature (from viscosity profiles), and these formulations show a big formulation latitude with very good tolerance for different hydrocarbon resin chemistries (especially saturated aliphatic and slightly aromatic–aliphatic unsaturated resins), with most of them giving good adhesive performance properties, when compared to the evaluated comparative SBS-based control.

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REFERENCES

1. Fornes, M. *Nonwovens Industry*, May **2009**, 12.
2. Jagisch, F. C.; Tancrede, J. M. In *Hand Book of Pressure-sensitive Adhesive Technology*; Satas & Associates: Warwick-RI, **1999**; 3rd ed., p 346.
3. Harlan Jr., J. T. US Patent No. 3239478, **1966**.
4. Godfrey, D. A. US Patent No. 4299745, **1981**.
5. Puletti, P. P. US Patent No. 4419494, **1983**.
6. Schmidt Jr., R. C.; Decowski Jr., S. K.; Puletti, P. P. US Patent No. 4460728, **1984**.
7. Schmidt Jr., R. C.; Puletti, P. P. US Patent No. 4526577, **1985**.
8. Raterman, J. *TAPPI Hotmelt symposium*, **1987**, 63.
9. Dunckley, P. M. *Adhesives Age*, Nov. **1993**, 17.
10. Sheriff, M.; Knibbs, R. W.; Langley, P. G. *J. Appl. Polym. Sci.* **1973**, *17*, 3423.
11. Kraus, G.; Rollmann, K. W. *J. Appl. Polym. Sci.* **1977**, *21*, 3311.
12. Kraus, G.; Rollmann, K. W.; Gray, R. A. *J. Adhesion.* **1979**, *10*, 221.
13. Class, J. B.; Chu, S. G. *J. Appl. Polym. Sci.* **1985**, *30*, 805.
14. Chu, S. G.; In *Hand Book of Pressure Sensitive Adhesive Technology*; Van Nostrand Reinhold: New York, **1989**; 2nd ed., p 159.
15. Satas, D. In *Handbook of pressure Sensitive Adhesive Technology*; Satas & Associates: Warwick-RI, **1999**; 3rd ed.
16. Yuan, B.; McGlinchey, C.; Pearce, E. M. *J. Appl. Polym. Sci.* **2006**, *99*, 2408.
17. Park, V. K. US Patent No. 3239478, **1974**.
18. Lindquist, J. S.; Cameron, J. C.; Simmons, E. R. US Patent No. 5869562, **1999**.
19. Trotter, J. R.; Petke, F. D. US Patent No. 4022728, **1977**.
20. Ives, C. L.; Middleton, I. P.; Lucas, D. M.; Riley, D. US Patent Application No. US2003/0232905 A1, **2003**.
21. Arriola, D. J.; Carnahan, E. M.; Hustad, P. D.; Kuhlman, R. L.; Wenzel, T. T. *Science* **2006**, *312*, 714.
22. Wang, H. P.; Khariwala, D. U.; Cheung, W.; Chum, S. P.; Hiltner, A.; Baer, E. *Macromolecules* **2007**, *40*, 2852.
23. Khariwala, D. U.; Taha, A.; Chum, S. P.; Hiltner, A.; Baer, E. *Polymer* **2008**, *49*, 1365.
24. Li Pi Shan, C.; Yalvac, S.; Diehl, C.; Marchand, G.; Rickey, C.; Karjala, T.; Carvagno, T.; Dunckley, P. M.; Germinario, L. *30th International PSTC Technical Seminar Orlando-FL*, **2007**.
25. Meissner, j.; Hostettler, J. *Rheol. Acta* **1994**, *33*, 1.
26. Metzner, A. B.; Reed, J. C. *AICHEJ* **1955**, *1*, 434.
27. Macosko, C. W.; In *Rheology-Principles Measurements and Applications*; Wiley-VCH: New York, **1994**; p 237.
28. Chu, S. G. In *Adhesive Bonding*; Plenum Publishing: New York, **1991**; p 97.
29. Shih, H. H.; Hamed, G. R. *J. Appl. Polym. Sci.* **1997**, *63*, 323.