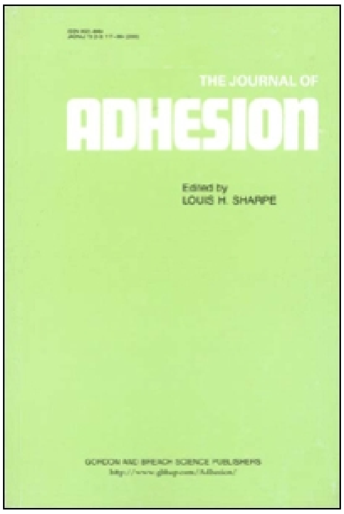

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Melt Synthesis and Characterization of Aliphatic Low-T_g Polyesters as Pressure Sensitive Adhesives

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Polyesters, which are readily synthesized in the absence of solvent, are excellent candidates for a new generation of pressure sensitive adhesives (PSAs) due to their low cost and potential biodegradability. In this study, linear, all-aliphatic polyesters with low glass transition temperatures (T_g) were synthesized using a solvent-free, environmentally friendly melt polycondensation methodology. Polyesters of various compositions were synthesized from different diol and diester monomers to adjust the glass transition temperature and achieve optimum adhesive properties. Melt polycondensation of an isomeric mixture of dimethyl-1,4-cyclohexane dicarboxylate (DMCD), dimethyl adipate (DMAP), diethylene glycol (DEG), and triethylene glycol (TEG) generated a series of linear low-T_g polyesters. The synthesized polyesters were characterized using size exclusion chromatography (SEC), differential scanning calorimetry (DSC), and ^{1}H NMR spectroscopy. The frequency- and temperature-dependent properties of the low-T_g polyesters were characterized using dynamic mechanical analysis (DMA). The adhesive performance of the polymers was evaluated using tack, peel, and shear strength measurements at ambient humidity and temperature. The low-T_g polyesters exhibited peel and tack properties comparable with commercial acrylic adhesives.

Keywords: Adhesion; Aliphatic polyesters; Polyesters; Pressure sensitive adhesives

INTRODUCTION

Pressure sensitive adhesives (PSA) represent a class of adhesives that adhere to a surface upon application of light pressure [1]. PSAs are

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utilized as self-adhesive labels, double-sided tapes, packaging tapes, and in biomedical applications such as drug-loaded adhesive patches [1–3]. Acrylic copolymers [4–6], styrene-isoprene-styrene (SIS) block copolymers [7], styrene-butadiene-styrene (SBS) block copolymers [8], styrene-butadiene rubber (SBR) [9], and polysiloxane [10]-based polymers are important families of polymers that are commonly used for PSA applications. In addition, a recent study describes the incorporation of carbon nanotubes and fullerenes [11] to improve the adhesive properties of PSAs. The performance of adhesives strongly depends on the viscoelastic properties of polymers [12]. PSAs must possess viscous properties in order to flow and dissipate energy during adhesive bonding; however, elastic properties are also essential for storing bond rupture energy. A balanced combination of viscous and elastic properties, therefore, is required for good adhesive performance. Chang [13,14] demonstrated the four-quadrant viscoelastic window concept to classify different types of PSAs using dynamic storage and loss modulus measured at frequencies of 10^{-2} and 10^{2} rad/sec, which corresponds to the time scale for bonding and debonding in an adhesive. A low glass transition temperature is another important property associated with polymers for PSA applications. It was shown, for example, that the T_g of polymers that are intended for use in PSA applications at room temperature generally falls between −20 and −60°C [1].

Tack, peel, and shear strength typically define the basic performance of a PSA. Tack corresponds to an adhesive's ability to adhere quickly to a surface and its resistance to separation after short contact time [15,16]. Tack is highly dependent on the modulus of the polymer; therefore, adjusting the composition and molecular weight of a polymer enables one to regulate tack for specific applications [17]. Moreover, tack not only depends on the bulk property of the polymer, but also on the surface energy of the adhesive and adherend surface [17,18]. Peel strength corresponds to the ability of an adhesive to resist removal from a surface upon peeling at a certain angle from the substrate. Peel strength is not a direct measure of the bond strength, but rather the sum of the energy needed to break the bond and deform the backing material and the adhesive. Shear strength represents an adhesive's ability to resist flow as shearing forces are applied and measured in a static manner. The adhesion behavior of polymers was characterized using a combinatorial approach [19]. Literature studies indicate that the molecular weight distribution of the polymer influences both resistance to peel and resistance to shear [1]. Additives such as tackifiers and plasticizers are commonly incorporated into PSA formulations to adjust T_g and viscoelastic properties.

In the literature, new types of polymers were investigated for use as novel PSAs, as well as to improve the adhesive properties of existing polymers [20]. More recently, Chan *et al.* [21,22] studied patterned adhesives to investigate the influence of surface patterns on adhesion of elastomeric interfaces. In addition, a recent study describes the incorporation of carbon nanotubes and fullerenes [11] to improve the adhesive properties of PSAs. In addition, incorporation of varying degrees of crosslinking using UV curing [23] resulted in balanced tack, peel, and shear strength; hence, improved the performance [24,25]. Incorporating hydrogen bonding represents an alterative method for improving the adhesive properties of polymers. Improved peel strength, tack, and cohesive strength were achieved upon incorporation of acrylic acid units, since they are able to hydrogen bond to other acrylic acid groups [26]. Recently, new families of PSAs have been derived from the existing synthetic processes and addressed the reduced availability of conventional raw materials [27–29]. For example, polyesters are viable candidates for a new generation of PSAs due to their low cost and potential biodegradability [30]. Polymerization involving a solvent-free melt phase also offers environmental benefits and volatile organic solvents are eliminated from the reaction system [31,32]. Moreover, polyester-based adhesives degrade in a more environmentally friendly manner due to the presence of hydrolyzable ester bonds on the polymer backbone [30]. Incorporating co-monomers and tailoring the topology of the polyesters facilitate efficient modification of both their thermomechanical properties and hydrolytic stability [31–34]. The diester monomer, DMCD, used in this study, is commonly utilized to synthesize polyesters and polyamides. Moreover, the influence of the *cis* and *trans* ratios of DMCDs on polymer properties was also investigated partially in response to an earlier study [35,36] that confirmed with the ratio of *cis* and *trans* isomers in a polymer composition affected the physical properties of the resulting polymer [37]. Specifically, the *trans* isomer produces a more crystalline structure compared with the *cis* isomer. Conversely, the *cis* isomer is less symmetrical and introduces kinks into the polymer backbone; therefore, it reduces the crystallinity of the polymer [38]. The UV-stability of this monomer also enables the utilization of DMCD-based polyesters for coatings and outdoor applications where UV radiation has a detrimental effect on the mechanical properties [39].

The current research investigates the synthesis of linear all-aliphatic polyesters for PSA performance. A series of low T_g polyesters for use as pressure sensitive adhesives was synthesized to determine the dependence of viscoelastic properties on temperature and frequency. In this study, careful selection of the comonomer composition enabled us to tailor the glass transition temperature over a broad range. The dependence of adhesive properties on molecular weight and composition was analyzed in detail using differential scanning calorimetry (DSC), ^{1}H NMR spectroscopy, and adhesive testing methods.

EXPERIMENTAL

Materials

Dimethyl adipate (DMAP), diethylene glycol (DEG), triethylene glycol (TEG), titanium tetraisopropoxide (99%), and toluene were purchased from Sigma-Aldrich, St. Louis, MO, USA Dimethyl-1,4-cyclohexane dicarboxylate (DMCD, *cis/trans* mixture) was donated by Eastman Chemical Company, Kingsport, TN, USA. All reagents and solvents were used as received without further purification.

Polymer Characterization

Molecular weight was determined at 40°C in tetrahydrofuran (THF) (ACS grade) using a Waters size exclusion chromatograph (SEC) equipped with 3 in-line PLgel 5 μm MIXED-C columns with an autosampler, a 410 RI detector, a Viscotek 270 dual detector (Houston, TX, USA), and an in-line Wyatt Technologies miniDawn multiple angle laser light scattering (MALLS) detector (Santa Barbara, CA, USA). ^{1}H NMR analyses were performed on a Varian (Palo Alto, CA, USA) Unity 400 spectrometer at 400 MHz in $CDCl_3$. Thermal analysis using a TA Instruments DSC (New Castle, DE, USA) determined thermal transition temperatures at a heating rate of 10°C/min under nitrogen; all reported values were obtained from the second heating cycle.

Synthesis of Linear Low-T_g Polyesters

Diester monomer (DMCD or DMAP) and 50 mol% excess diol (DEG or TEG) were charged to a 250-mL round-bottomed flask equipped with a mechanical stirrer, nitrogen inlet, and condenser. Titanium tetraisopropoxide (60 ppm $Ti(OR)_4$) catalyst was added to facilitate transesterification. The flask was degassed three times using vacuum and nitrogen flush and subsequently heated to 200°C. The reaction proceeded under nitrogen atmosphere at 200°C for 2 h and the temperature was raised to 220°C over 3 h. Vacuum (0.1 to 0.2 mm Hg) was gradually applied for 2 h at 275°C. The reaction products were characterized without further purification.

Adhesive Sample Preparation

Samples were prepared from solution casting of the polymer solutions (30 wt% in toluene) on poly(ethylene terephthalate) (Mylar®) backing material using a doctor's blade. Coated films were dried at room temperature for 10 min and subsequently dried in a convection oven at 60°C for an additional 10 min. The coating weight of each film specimen was measured prior to adhesive testing, with a coating weight of 20 g/m^2 targeted for each sample. The surfaces of the stainless steel substrates were carefully cleaned with acetone prior to peel, tack, and holding power tests.

180° Peel Testing

The peel strength of all samples was measured as follows. Strips of 2.54 cm width were cut from the polymer coated Mylar® and the coating weight of the each strip was measured. Upon application of the specimen onto a stainless steel substrate, a 2-kg roller was passed over the sample two times under its own weight before each peel test. After storage under controlled temperature (25°C) and humidity (50% R.H.) conditions, the specimen strip was peeled from a stainless steel substrate at an angle of 180° and at a speed of 300 mm/min using an Instron tensile tester (Instron 4201, Instrou, Norwood, MA, USA) [40]. The reported results represent an average of five specimens.

Loop Tack and Rolling Ball Tack (RBT)

Rolling ball tack experiments were performed using a 7/16(1.1 cm) inch diameter steel ball and a 2.54 × 2.54 cm strip of tape that was conditioned at 72° F and 50% R.H [41]. Loop tack was measured using a ChemInstruments loop tack tester (Fairfield, OH, USA) at a cross-head displacement rate of 300 mm/min [42]. Low-T_g polyester coated Mylar substrate was brought into contact with a predetermined stainless steel surface area (2.54 × 2.54 cm). The maximum force per width of the specimen was recorded at a dwell time of 1 sec.

Holding Power

Resistance-to-shear was measured as the time required for a specific area of polymer to fail under continuous applied force [43]. Polymer films were cut into strips and a 2.54 × 2.54 cm square of the specimen was placed onto a stainless steel panel. A 2-kg roller was passed over the sample two times under its own weight before each test. A clamp was placed on the free end of the specimen ensuring that the clamp extended evenly across the width and was properly aligned to uniformly distribute the load. The specimen was then placed in the test stand and a 1-kg weight was applied to the clamp. The elapsed time during which the specimen completely separated from the test panel was recorded. Holding power was tested at controlled room temperature and humidity. The reported values represent an average of five tests.

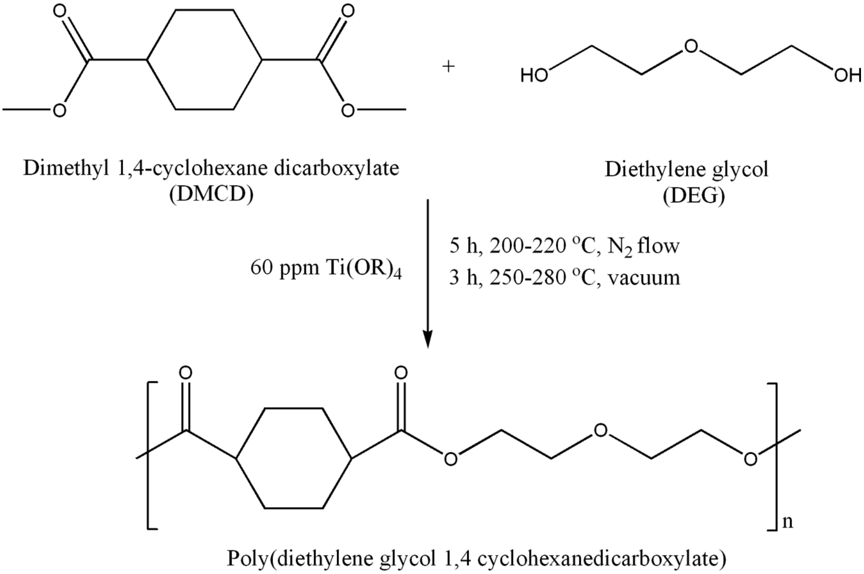
Dynamic Mechanical Analysis

The rheological properties of the polymers were studied using a TA Instruments AR 1000 stress-controlled rheometer with 8 mm parallel plate geometry. Strain amplitudes were limited to the linear viscoelastic regime over a frequency range of 0.1 to 100 Hz. The time-temperature superposition method was utilized to investigate the frequency dependence over a wide frequency range.

RESULTS AND DISCUSSION

The synthesis of solvent-free, water-based acrylic PSAs has become increasingly important because of their environmental advantages [44]. In this study, all linear low-T_g polyesters were also synthesized using an environmentally friendly, solvent-free melt-polycondensation reaction using the following procedure (Scheme 1). DMCD was reacted with DEG to form an oligomeric precursor, which underwent subsequent polycondensation to produce high molecular weight polyesters when excess diethylene glycol and methanol were removed under vacuum. Titanium isopropoxide was used as the transesterification catalyst. The transesterification of DMCD and DEG was conducted at 200–220°C with removal of the methanol. Polycondensation of the oligomers occurred at a higher temperature under vacuum to remove the DEG by-product. The diester monomer, 1,4-dimethyl cyclohexanedicarboxylate, was composed of a mixture of 90% *trans* and 10% *cis* isomers. The *cis* to *trans* ratio in the starting monomer and the polymer was calculated from the integration of the corresponding peaks in ^{1}H NMR spectrum.

The thermal properties and molecular weight of DMCD-, DMAP-, DEG-, and TEG-based low-T_g polyesters are summarized in Table 1. The glass transition temperatures of the synthesized low-T_g polyesters ranged from -49 to -18°C depending on the monomer used in the synthesis. Low-T_g polyesters based on DMCD were characterized



SCHEME 1 Synthesis of linear low-T_g polyester *via* melt polycondensation.

using ^{1}H NMR spectroscopy to determine the ratio of *cis* to *trans* isomers in the synthesized polymers. This was calculated using the integration of the peaks corresponding to the *cis* and *trans* protons in the cyclohexane ring (Fig. 1). ^{1}H NMR resonances for the cycloaliphatic protons in the cyclohexane dicarboxylate residue were identified at the 2.8–1.5 ppm region. The calculation of the areas under the peaks, which correspond to the *cis* and *trans* cyclohexanedicarboxylate isomer, indicated that the *cis* to *trans* ratio did not change after polycondensation. In the literature, however, studies have indicated that the *cis* to *trans* ratio changes under certain reaction conditions [36].

TABLE 1 Summary of Thermal Analysis Results (DMAP: dimethyl adipate, DMCD: 1,4-dimethyl cyclohexanedicarboxylate, DEG: diethylene glycol, and TEG: triethylene glycol)

Composition	M_w (g/mol)	M_n (g/mol)	M_w/M_n	T_g (°C)
DMAP + DEG	67,000	44,000	1.53	−47
DMAP + TEG	59,000	27,000	2.22	−49
DMCD + DEG	55,000	35,000	1.56	−12
DMCD + TEG	41,000	19,000	2.13	−25

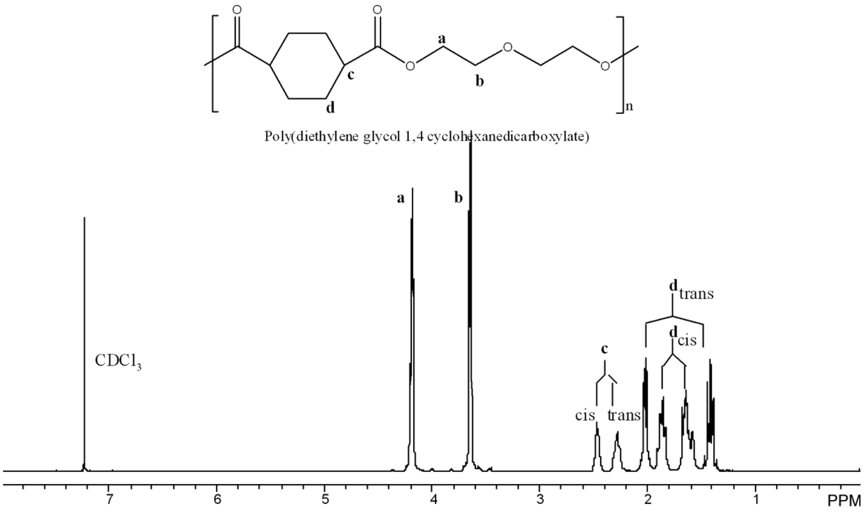


FIGURE 1 ^{1}H NMR of linear DMCD-DEG-based polyester.

The peel test results confirmed that low-T_g polyesters based on DMCD and DEG exhibited higher peel strengths compared with DMAP, DEG, and TEG compositions under applied test conditions. All compositions showed cohesive failure, as suggested by the presence of a residue on the stainless steel substrate after the peel test. The summary of peel test results is shown in Table 2. In order to evaluate the dependence of the adhesives properties of the DMCD-DEG-based low-T_g polyesters, polymers of varying molecular weights were selected based on the molecular weight information obtained from SEC analysis. A summary of the thermal analysis and SEC results of polyesters of varying molecular weights (28,000 to 57,000 g/mol) is shown in Table 3. Characterization of the thermal properties of the synthesized polyesters indicated that all polymers were amorphous and the

TABLE 2 Summary of Molecular Weight and Peel Properties of Linear Low-T_g Polyesters

Composition	M_w (g/mol)	M_n (g/mol)	M_w/M_n	Thickness (μm)	Peel adhesion (N/mm)
DMCD + DEG	55,000	35,000	1.56	24 ± 3	10.53 ± 0.40
DMCD + TEG	41,000	19,000	2.13	27 ± 6	0.31 ± 0.04
DMAP + DEG	67,000	44,000	1.53	22 ± 2	1.01 ± 0.05
DMAP + TEG	59,000	27,000	2.22	30 ± 5	1.23 ± 0.05

TABLE 3 Thermal Analysis and SEC Results of DMCD- and DEG-Based Low-T_g Polyesters Having Various Molecular Weights

cis/trans DMCD[a]	M_w (g/mol)[b]	M_n(g/mol)[b]	M_w/M_n	T_g (°C)[c]
95/5	28,000	13,000	2.14	−15
95/5	34,000	14,000	2.43	−15
95/5	44,000	17,000	2.66	−13
95/5	57,000	21,000	2.71	−11

[a] Calculated from ^1H NMR of the polymer.
[b] SEC conducted in THF.
[c] DSC, 2nd heating cycle.

T_g varied between −15 to −11°C. This temperature range is slightly higher compared with other adhesive compositions studied in earlier literature; however, it should be noted that these T_g values correspond to neat polymers in the absence of any tackifiers or plasticizers.

Selected low-T_g polymers utilized in this study were characterized using DMA and adhesive tests to evaluate the influence of molecular weight on adhesive properties. DMA revealed the glass transition temperature, storage modulus at room temperature, and the plateau region, which reflects the holding power of the polymer (Fig. 2). DMA also confirmed that the peak tan delta value increased as the molecular weight of the polymer increased. DMA analysis indicated a high storage modulus at low temperatures, where the polymer behaved like a glass. As expected, the storage modulus decreased as

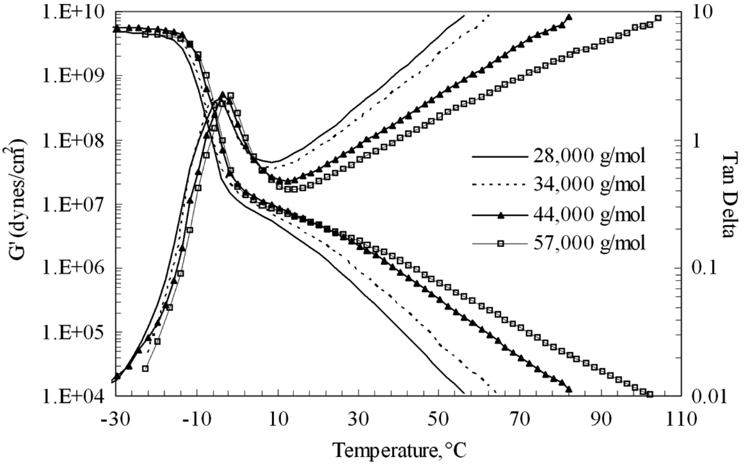


FIGURE 2 Dynamic mechanical analysis of molecular weight series.

the temperature was increased and a short plateau region was observed. It should be noted that these polymers did not yield a long plateau region, which is also typical for acrylic adhesives. This was attributed to the lower molecular weights of these polyesters compared with acrylic adhesives, whose molecular weights were as high as 500,000 g/mol [45]. In comparing the behavior of varying molecular weight polymers, we determined that as molecular weight increased, the storage modulus plateau extended and the storage modulus measured at room temperature increased. The molecular weight demonstrates a strong correlation with the adhesive properties due to its influence on several polymer properties such as modulus and viscosity. An increase in molecular weight is expected to improve adhesive properties and an extended plateau region due to higher degree of entanglements. It is important to note that very high molecular weights results in poor wetting due to reduced flow. Another important measurement, following the Dahlquist's criterion, correlates tack with the compliance of an adhesive. It states that the 1 sec creep compliance of a polymer, which is associated with bonding, must be $3 \times 10^{-7}\,\mathrm{cm}^2/\mathrm{dyne}$ or larger to exhibit good tack characteristics. DMA analyses of all of the samples indicated that storage modulus at room temperature was clearly below 3×10^6 dynes/cm^2, which is the Dahlquist criterion for tack.

The molecular weight effect of the low-T_g polyesters was measured with rolling ball and loop tack methods (Fig. 3). Tack measurements indicated that as molecular weight increased, tack properties also increased, but then leveled off. The dependence of tack on molecular weight is confirmed in the literature [46], which shows that as

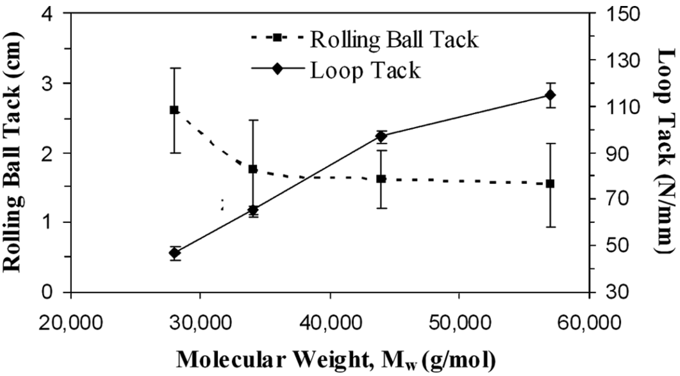


FIGURE 3 Tack properties of molecular weight series measured with rolling ball and loop tack techniques.

molecular weight increases, the viscosity of the polymer also increases and reaches a high viscosity, thereby efficiently reducing the wetting by the polymer. In some applications, therefore, additives such as tackifiers or plasticizers are required to improve the tack of high molecular weight polymers for adhesive applications [47]. However, in the current study all the samples were tested in the absence of plasticizers or tackifiers.

As we reported herein, both the resistance-to-shear and peel strength improved as the molecular weight of the low-T_g polymers increased (Fig. 4). The polymers exhibited peel strengths comparable with commercially available polymers. However, the holding power of the polymers indicated inadequate cohesive strength. The typical holding power for commercially available polymers is reported to be 10,000 min. However, the holding power for our low-T_g polyesters was limited to 500 min. It should be noted, however, that the reported adhesive results were determined from characterization of neat samples. Peel performance may be improved through the incorporation of additives such as tackifiers, which are compatible with this polymer composition. Moreover, the molecular weights of the samples under used in this study were lower when compared with conventional polymers typically used as pressure sensitive adhesives. Since the molecular weight of a polymer is important for achieving superior mechanical properties due to entanglements, the cohesive strength of these samples may be improved as the molecular weight is increased. Moreover, all these polyesters are linear; therefore, the absence of crosslinking is another factor resulting in low cohesive strength. As reported in the literature [23], incorporation of low levels of crosslinking results in enhanced cohesive strength. It is important to note that low crosslink

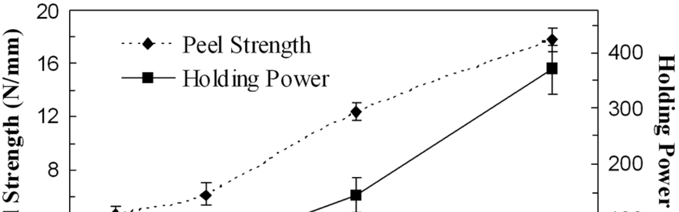


FIGURE 4 Peel and shear properties of molecular weight series.

density and high molecular weight between crosslink points are required to attain balanced tack and shear properties [48].

CONCLUSIONS

Linear all-aliphatic polyesters having low glass transition temperatures (T_g) were synthesized *via* solvent-free, environmentally friendly melt polycondensation. The polymerization of isomeric mixtures of dimethyl-1,4-cyclohexane dicarboxylate (DMCD) and diethylene glycol (DEG) in the melt phase generated a series of linear low-T_g polyesters with a T_g varying from -15 to -11°C. Rheological studies and the characterization of adhesive properties indicated an influence of molecular weight on adhesive performance for the synthesized low-T_g polyesters. Peel strength and resistance to shear was enhanced as the molecular weight of low-T_g polyesters increased from 28,000 to 57,000 g/mol. Low-T_g polyesters exhibited good tack and peel properties; however, the cohesive strength of the low-T_g polyesters was low compared with polymers used for conventional pressure sensitive applications.

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