Effect of Soft Segment Composition on the Physical Properties of Nonionic Aqueous Polyurethane Containing Side Chain PEGME

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ABSTRACT: In this study, a series of nonionic waterborne polyurethanes containing side chain poly(ethylene glycol) monomethyl ether (PEGME) were synthesized using 4,4-methylene bis(isocyanatocyclohexane), the nonionic dispersing center produced by isophorone diisocyanate, N-diethanol amine, and PEGME, and a series of soft segments [polyethylene glycol (PEG), polypropylene glycol (PPG), polytetramethylene glycol (PTMG), and polycaprolactone (PCL)] with a different molecular weight. The solution properties of the polyurethane (PU) dispersions, thermal properties of the PU casting films, and the water vapor permeability (WVP) of the PU coated fabrics were investigated. Our results show that the surface tension of PU dispersion and its contact angle to untreated nylon fabric are higher for the ether-based PU dispersion than for the ester-based PU dispersion, with a sequence of PEG-PU > PTMG-PU > PPG-PU > PCL-PU. Both the surface tension and the contact angle between PU dispersion and nylon fabric increase upon increasing the M_w of the soft segment. As for the thermal property of PU film, the T_gs of PCL-PU is the highest, followed by PEG-PU, PTMG-PU, and PPG-PU, and the T_gs decreases with the increase in the length of the soft segment. The T_ms follows the order: PEG-PU > PCL-PU > PPG-PU > PTMG-PU, and the T_ms increases upon increasing the soft segment length. As regard to PU coated fabric, the trend of WVP is PEG-PU > PTMG-PU > PPG-PU > PCL-PU, which increases with the increasing the soft segment length. With regarding to application for the coating, the Coated-PU with PEG-PU has an excellent WVP. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 105: 1391–1399, 2007

Key words: polyurethanes; water-soluble polymers; thermal properties; coatings

INTRODUCTION

Polyurethane (PU) is a block copolymer comprising hard and soft segments. The solubility parameter between the soft segment and hard segment is significantly different and generates a large phase incompatibility that leads to a two-phase structure with some degree of microphase separation. Therefore, the composition of the soft and hard segments has great influence on the physical properties of PU, such as repellent elasticity, film-forming ability, and resistance to abrasion.[1–3] Recently, due to the awareness of environmental protection and the limited use of volatile organic carbons (VOCs), the development of VOCs-free waterborne PU (WBPU) resin has attracted great attention.[4] The WBPU can be divided into three types: cationic, anionic, and nonionic.[5] Generally speaking, cationic WBPU and anionic WBPU are built by the tertiary-nitrogen group and carboxylic acid group, respectively, and they get emulsified in water by the neutralization of ionic group. The nonionic WBPU

mostly introduces polyether with hydrophilic segment into the PU structure to achieve hydrophility. On the whole, the WBPU is excluded from the category of environmental pollutants because it is VOC-free, and that the potential public hazards can be avoided in the resin storage.[4] However, the water-resistance and mechanical properties of PU casting film are lower than those of solvent-based PU because of the introduction of ionic or nonionic groups with hydrophilic structure.[5–8] There is still much room for improvement in terms of soft and hard segments within the PU structure for its application on coating materials.

In the past, several investigations have been carried out on the soft segment component in anionic WBPU.[9–14] For example, Kim and Lee[9] synthesized the anionic WBPU by using dimethylol propionic acid as the dispersing center, isophorone diisocyanate (IPDI) as the hard segment, and polyethylene glycol (PEG), polypropylene glycol (PPG), or polytetramethylene glycol (PTMG) as the soft segment, and investigated its physical properties. The results showed that the PU dispersion with PEG had the smallest particle size, and the particle size became smaller in direct proportion to the increase in M_w of PEG. Both the surface tension of PEG-PU dispersion and the water swelling of the PU

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casting film were greater than those of other PUs; the PTMG-PU film presented excellent strength. Lee et al.[10] synthesized anionic WBPU by altering the molecular weights of polytetramethylene adipate glycol (PTAd). The results show that the particle size of PU dispersion becomes smaller as the M_w of PTAd becomes greater. The swelling of PU casting film becomes bigger as the M_w of PTAd increases; however, the soft segment of PU is difficult to crystallize because the M_w of PTAd is less than 1500 g/mol. Yang et al.[14] varied the soft segments (PEG, PPG, and PTMG), and used PEG-PPG-PTMG copolymer as the soft segment to synthesize PU anionomers. They pointed out that the viscosity of the PU dispersion with PEG-PPG-PTMG copolymer was higher than that of other PUs. The PTMG-based PU film has the largest strength and elongation, with a sequence of the PEG-PPG-PTMG-based PU, and the PU with PEG-PPG-PTMG can effectively reduce the particle size of dispersion. As far as the research on the soft segment of cationic PU[6,15,16] is concerned, our laboratory synthesized the cationic WBPU with N-methyl diethanolamine (N-MDEA) as the dispersing center, 4,4-Methylene bis(isocyantocyclohexane) (H$_{12}$MDI) as the hard segment, and a series of polydiol as the soft segment.[16] Our results showed that the particle size of PU dispersion followed the order: PCL-PU > PPG-PU > PEG-PU > PTMG-PU and it increased as the M_w of soft segment is increased, and the trend became gradual as the M_w of soft segment reached above 2000 g/mol. This result is different from that shown in the case of the anionic WBPU dispersion.[9,10] From the past researches, it is shown that the composition of soft segment has a great influence on the physical properties of WBPU. However, an investigation concerning the effect of soft segment on the physical properties o nonionic PU with side chain poly(ethylene glycol) monomethyl ether (PEGME) is scarce in the literature.

In the past studies on solvent-based nonionic PU, the PEG component was almost directly introduced into the PU backbone or as the side chain to facilitate the hydrophilicity in PU. Christina and Bengtet,[17] for example, reserved the end functional —NCO group of hexamethylene diisocyanate in the PU side chain and then grafted the PEG into the —NCO group, and discussed the effect of the side chain length on the thermal and mechanical properties of PU film, and the contact angle of water to PU membrane. The results showed that the introduction of PEG into the side chain would effectively promote the hydrophilicity of PU membrane, but the melting point and tensile strength would be significantly lowered. Our laboratory has synthesized cationic-nonionic PU with PEGME side chain and cationic PU without PEGME side chain.[18] We found that the PU film with side chain had higher moisture absorption; however, with lower T_gs and T_ms. Until now, the effect of soft segment composition on the PEGME-containing side chain of nonionic WBPU has not been discussed.

This study focused on the effect of the soft segment composition on the solution properties and thermal properties of nonionic WBPU with PEGME side chain. First, nonionic dispersing center with PEGME was synthesized, and then the nonionic PU was synthesized by using ether-type (PEG, PPG, and PTMG) or ester-type polycaprolactone (PCL) polydiol of different molecular weights as the soft segment. In addition, the synthesized PU was coated on nylon fabrics to find out the effect of the soft segment composition on the WVP.
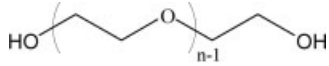
EXPERIMENTAL

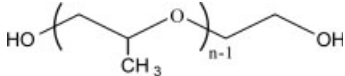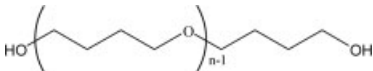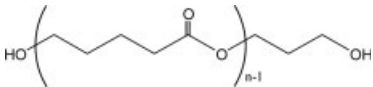### Synthesis of nonionic dispersing center

Before synthesizing nonionic WBPU, we first synthesized a nonionic dispersing center (N-PDEA) following the method described in our previous research.[18] One mol of poly(ethylene glycol methyl ether) (PEGME, M_w = 750, reagent, Acros) was mixed with 1 mol of IPDI
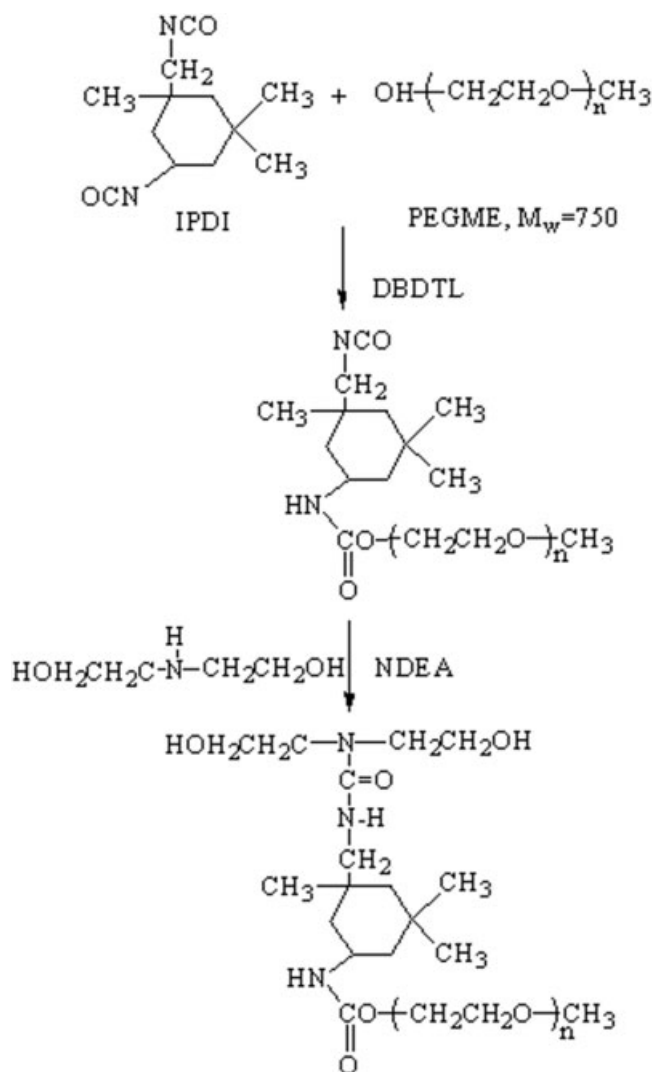
Scheme 1 The preparation process for nonionic dispersing center.

TABLE I
The Symbol, Structure of Polyol and Composition of Nonionic WBPU

Symbol	Polydiol			WBPU composition (wt %)		
	Type/structure	M_w (g/mol)		Hard segment	Soft segment	PEGME content
PEG-1000 PU	Poly (ethylene glycol), PEG	1000		36.00	30.80	33.20
PEG-2000 PU		2000		27.53	47.09	25.38
PEG-3000 PU		3000		22.28	57.18	20.54
PPG-1000 PU	Poly (propylene glycol), PPG	1000		36.00	30.80	33.20
PPG-2000 PU		2000		27.53	47.09	25.38
PPG-3000 PU		3000		22.28	57.18	20.54
PTMG-1000 PU	Poly (tetramethylene glycol), PTMG	1000		36.00	30.80	33.20
PTMG-2000 PU		2000		27.53	47.09	25.38
PTMG-2900 PU		2900		22.71	56.35	20.94
PCL-1250 PU	Poly (caprolactone), PCL	1250		33.43	35.75	30.82
PCL-2000 PU		2000		27.53	47.09	25.38
PCL-3000 PU		3000		22.28	57.18	20.54

H_{12}MDI : Polyol : N-PDEA : EDA = 4.05 : 1 : 1 : 2.
The hard segment wt % was calculated by:

$$\frac{H_{12}MDI + EDA}{H_{12}MDI + polydiol + (N - PDEA) + EDA} \times 100\%$$

The soft segment wt % was calculated by:

$$\frac{Polydiol}{H_{12}MDI + polydiol + (N - PDEA) + EDA} \times 100\%$$

The side chain PEO wt % was calculated by:

$$\frac{N - PDEA}{H_{12}MDI + polydiol + (N - PDEA) + EDA} \times 100\%$$

(Bayer, Germany) in a reactor for dehydration. Then, 0.05% of di-n-butyltin dilaurate (DBDTL, [CH$_3$(CH$_2$)$_3$]$_2$ Sn[OCO(CH$_2$)$_{10}$CH$_3$]$_2$, TCI) was added and reacted at 35°C under pure nitrogen for 4 h. The content of terminal —NCO group during the reaction was determined using standard —NCO titration method.[19] The temperature was raised to 45°C. Finally, 1 mol of N-diethanol amine (NDEA, reagent, Acros) was added to react for 2 h to obtain the nonionic dispersing center. The preparation processes are shown in Scheme 1.

Synthesis of nonionic WBPU

The nonionic WBPU was synthesized according to the prepolymer process method.[18] We placed 1 mol of polydiol PEG, PPG, PTMG or polycaprolactone (PCL)] with different molecular weights (molecular weights = 1000, 2000, and 3000) and 1 mol of N-PDEA in a vacuum reactor. After mixing and removing moisture, the mixture was added with 4 mol of 4,4'-methylenedicyclohexyl diisocyanate (H$_{12}$MDI, Bayer, Germany). The reaction was
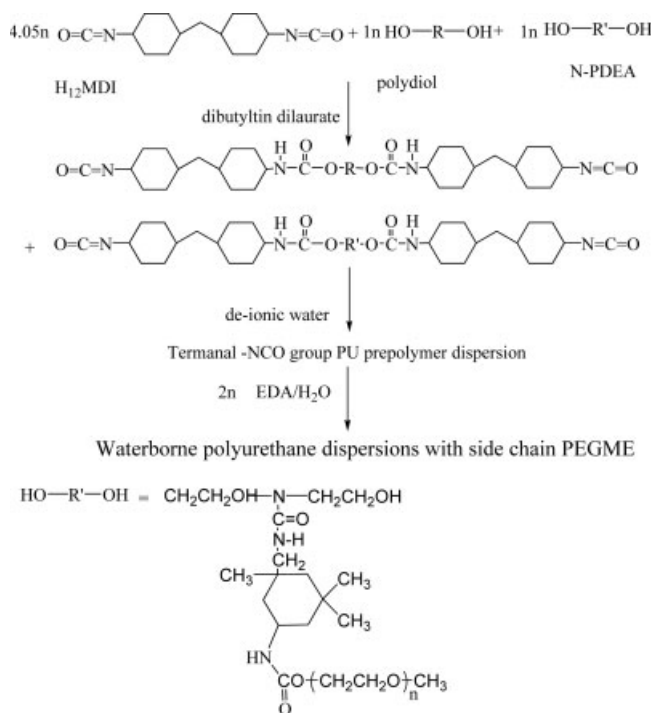
Scheme 2 The preparation process for WBPUs with side chain PEGME.

performed at 50°C (under nitrogen atmosphere) initially, and increased gradually to 80°C. The reaction continued until the —NCO content reached half of the reactant by titration.[19] The mixture was cooled to 10°C and emulsified by deionized water, and then added ethylenediamine (EDA, reagent, Acros) to process the chain extending reaction. The composition and symbol of PU synthetic are displayed in Table I. The preparation processes are shown in Scheme 2.

Identification and analysis of synthetic compound

The Fourier transform infrared (FTIR) spectra were analyzed using BIO-RAD FTS-7 spectrometer at 25°C. The scanning range was 4000–400 cm^{-1} with a resolution of 4 cm^{-1} for 64 times. Figure 1 shows the FT-IR analyses of nonionic dispersing center (*N*-PDEA). The isocyanate (—N=C=O) absorption peak of *N*-PDEA at 2270 cm^{-1} has diappeared indicating that the —N=C=O group of IPDI terminal of the synthetic compound is completely reacted. Moreover, there are respectively, the absorbion peaks of —OH, —NH—, —C=O and —CNH at 3450, 3333, 1712, and 1530 cm^{-1}, respectively, and there is a clear absorbion peak of ether group (—O—) at 1111 cm^{-1}. It shows that the synthetics are ether compounds whose ends possess an OH group. Besides, the FTIR spectra of the WBPUs are also illustrated in Figure 1. The —N=C=O group absorption peaks (2270 cm^{-1}) of these synthetic compounds are disappeared, indicating that the

—N=C=O end group of the synthetic compounds has reacted completely. The absorption peaks of —NH, —COO, and —RCNH— groups occurred at 3330, 1708, and 1531 cm^{-1}, respectively. The formation of —NHCOO (urethane) group in the structure of synthetic compounds was confirmed by these three groups. Furthermore, the absorption peaks at 1112 cm^{-1} and 2850 cm^{-1} or 2940 cm^{-1} belong to —O—, and —CH$_2$— or —CH$_3$ groups, respectively.

Solution properties of WBPU

The particle size of WBPU dispersion was measured by the light scattering method (Malvern, He-Ne laser, 633 nm). The sample was diluted in deionized water to 0.5%, followed by ultrasonic treatment. The surface tension of PU dispersions was measured by CBVP-A3 (Kyowa Kaimenagaku Ltd.). The contact angle of PU dispersion to untreated nylon fabric surface was tested by FACE CA-4 150. The testing condition of the contact angle is the dropping of PU dispersion with low concentration (2 wt %) onto the surface of nylon fabric.
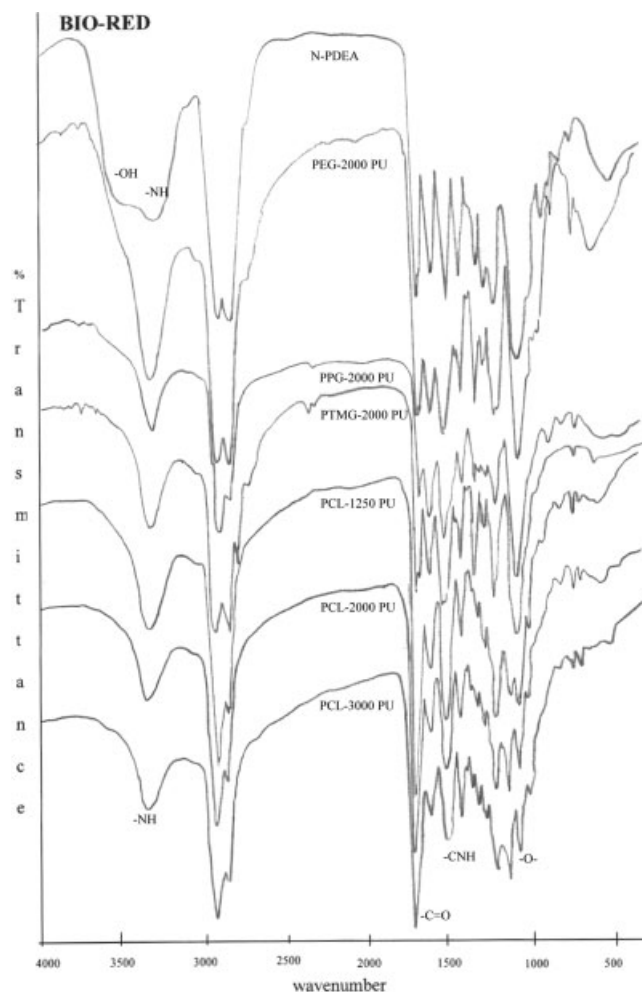

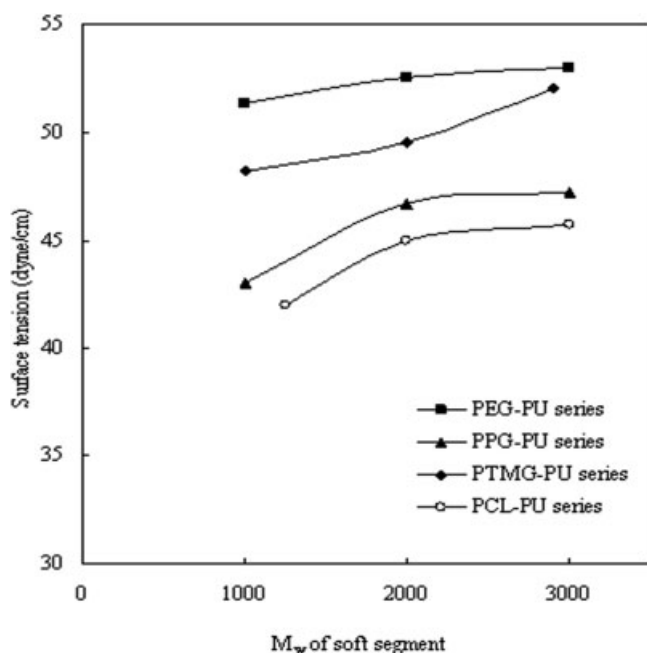**Figure 1** The FTIR spectra of the WBPUs.

Figure 2 The effect of the soft segment composition on the surface tension of PU dispersions.

Preparation and testing of casting films

We took the solid content of 30% aqueous PU coating on a Teflon plate and dried at room temperature for 3 days, and then at 40°C over a week. Differential scanning calorimetry (DSC, Du-Pont Thermal analyst 2000) was used to study the thermal properties of PU casting films with a heating rate of 10°C/min under nitrogen purge of 60 mL/min.
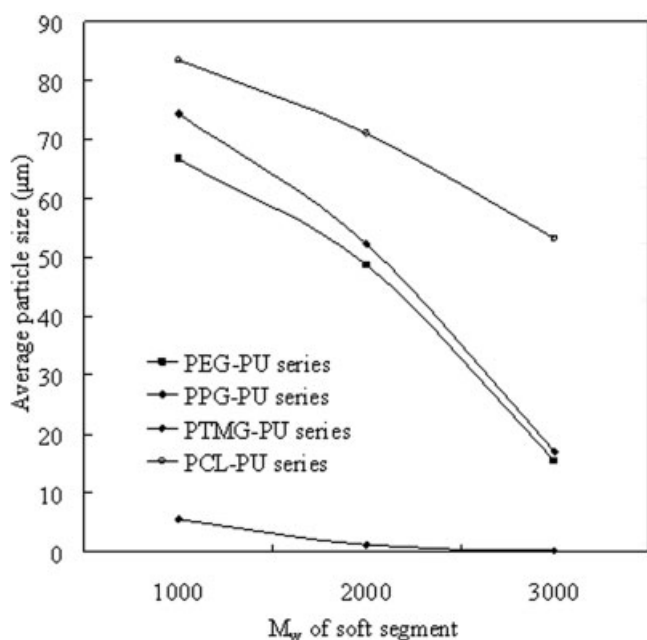
Measurement of coated fabrics

The PU dispersion of 30% solid content blended with 0.4% melamine resin and 10% hydroxyl ethyl cellulose was coated onto the nylon taffeta fabrics (Everest Textile Company, Taiwan), and predried at 100°C for 3 min, and cured at 130°C for 5 min. The contact angle between pure water and PU-coated fabric was tested by FACE CA-4 150, which is dropping pure water on the PU coated fabric. The water vapor permeability (WVP) of the coated fabrics was measured based on the JIS-1099 CaC1 method at 40°C with a 90% relative humidity.

RESULTS AND DISCUSSION

Solution properties

Figure 2 shows the effect of the soft segment composition on the surface tension of PU dispersions. The surface tension follows the order: PEG-PU > PTMG-PU > PPG-PU > PCL-PU for the same M_w of soft segments. The surface tension of ether-based PU (PEG-PU, PTMG-PU, and PPG-PU) is larger than that of ester-based (PCL-PU) because the ether-based PU's soft segment has a hydrophilic ether group (—O—). There is an ether group in every two methylene groups, especially in the PEG-PU, which leads to a more hydrophilic nature and causes the ether-based PU molecular chain difficult to be absorbed at the air–water interface, resulting in a greater surface tension. On the contrary, the PCL-PU with an ester group
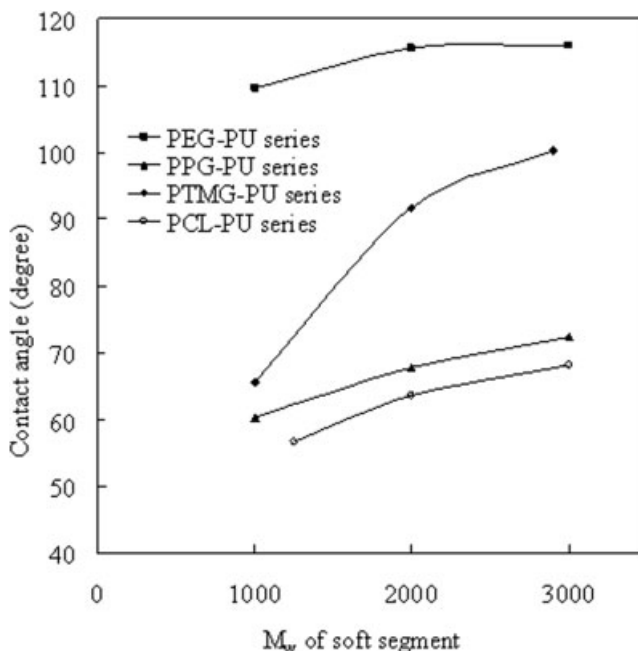

Figure 3 The effect of soft segment composition on the particle size of PU dispersions.

Figure 4 Relationship between soft segment composition and contact angle between PU dispersions and nylon fabric surface.
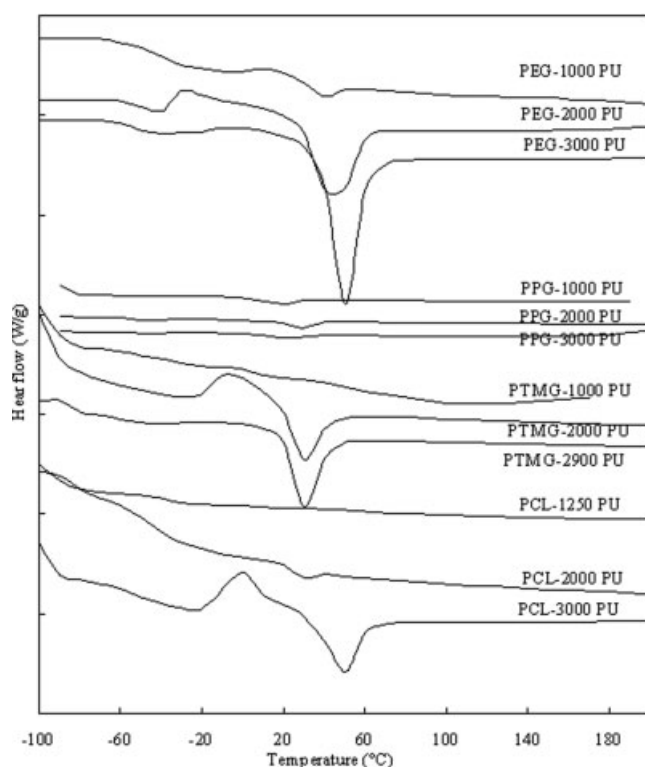
Figure 5 The DSC heating curve of PU casting films.

(—COO) can generate hydrogen bonding with the —NH group of urethane linkage causing intermolecular agglutination making it more hydrophobic. Therefore, the molecular chain of ester-based PU can easily be absorbed at the air–water interface on the surface of PU dispersion, so that the surface tension is the smallest. The surface tension of PPG-PU is lower than those of PEG-PU and PTMG-PU owing to the side chain methylene group (—CH$_3$) with hydrophobic nature. With regard to the M_w of soft segment, the surface tension increases upon increasing the molecular

weight. This is because as the soft segment length increases, the content of hydrophobic hard segment decreases (as shown in Table I), promoting its hydrophilicity, making it hard for the molecular chains to absorb at the air–water interface of the solution. The surface tension of nonionic WBPU is about 40–55 dyn/cm, which is higher than that of cationic WBPU[16] ($\sim$ 35–45 dyn/cm). This is related to the comparatively fewer number of hydrophobic hard segments in the nonionic PU with hydrophilic PEGME.

Figure 3 shows the effect of soft segment composition on the particle size of PU dispersions. The particle size follows the order: PCL-PU > PPG-PU > PEG-PU > PTMG-PU. Except for PTMG-PU, the particle size takes on an opposite trend against that of the surface tension. In general, the particle size of PU dispersion is mainly affected by its molecular weight, molecular chain extending property, and the intermolecular interaction in the solution. The more hydrophobic the PU is, the easier for conglomeration to occur between particles, resulting in a greater particle size. The particle size of PTMG-PU is far less than that of PEG-PU with higher hydrophilic nature, which may be due to the fact that the PTMG-diol possesses four methylene groups to lead a lower cohesive energy of PTMG-PU.[20] With regard to the M_w of the soft segment, all particle sizes of PU dispersions decrease with the increase in M_w of soft segment. This is because the hydrophobic hard segment content declines as the soft segment content increases, causing a decrease in the hydrophilic nature of PU.

Recently, PU has often been used as the coating agent for nylon fabric. To understand the affinity between PU dispersion and nylon fabric, the contact angle between PU dispersion and the untreated nylon fabric was measured. As shown in Figure 4, the contact angle between PU dispersion and nylon fabric (56°–109°) is far less than that of pure water (120°).

TABLE II
Thermal Properties of PU Films and PU-Coated Fabric

| Symbol	DSC analysis			Coated-fabric properties		
	T_gs (°C)	T_ms (°C)	ΔH (J/g)	Thickness (mm)	Contact angle (degree)	W.V.P (g/m$_2$/24h)
Untreated fabric	–	–	–	0	114	7230
PEG-1000 PU	−42.06	36.61	5.165	3.1	41	4580
PEG-2000 PU	−50.35	42.85	51.42	3.0	43	5980
PEG-3000 PU	−53.70	47.69	72.69	3.5	61	6200
PPG-1000 PU	−50.93	28.80	4.56	2.9	63	3220
PPG-2000 PU	−54.5	29.37	3.82	2.8	70	4120
PPG-3000 PU	−57.21	30.18	3.42	2.6	79	4260
PTMG-1000 PU	−42.20	–	–	3.1	53	4510
PTMG-2000 PU	−55.47	29.80	18.99	3.2	55	5660
PTMG-2900 PU	−59.1	30.59	27.74	2.6	70	5880
PCL-1250 PU	−38.84	–	–	3.2	87	2220
PCL-2000 PU	−47.45	32.12	0.35	2.9	98	3370
PCL-3000 PU	−53.39	46.77	35.88	2.7	102	3400
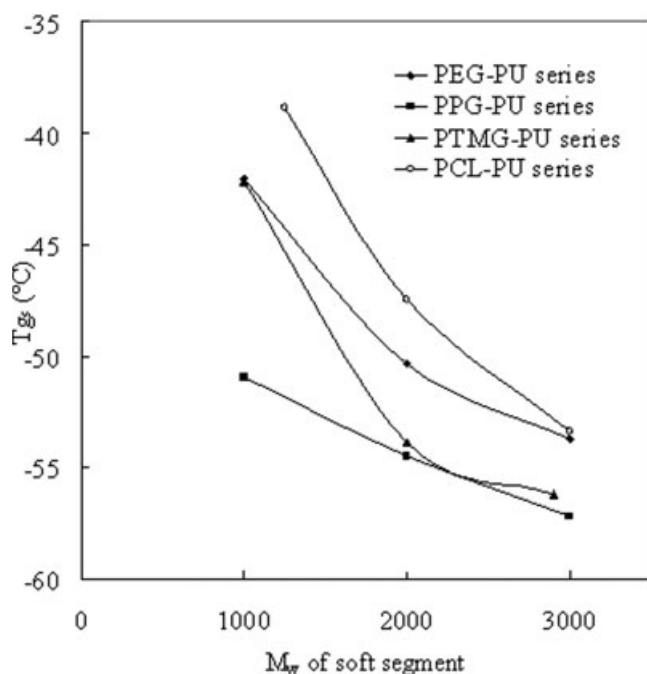
Figure 6 Effect of soft segment composition on T_gs of PU film.

This indicates that all types of PU show a higher affinity with nylon fabric. The contact angle followed the order: PEG-PU > PTMG-PU > PPG-PU > PCL-PU. The ester-based PCL-PU has the largest interaction force with nylon fabric (polyamide structure) and thus has the smallest contact angle, because the urethane linking (—NHCOO—) in the hard segment of PCL-PU as well as the ester group (—COO—) in the soft segment can be formed by the hydrogen bonding with amide group (—NHCO—) in nylon. Contrarily, the ether-based PU is more hydrophilic that leads to the structure not easily to wet the hydrophobic nylon. Especially, in the case of PEG-PU has fewer hydrophobic methylene groups, so that the greater hydrophilicity leads to the largest contact angle with nylon. Furthermore, the contact angle increases upon increasing the M_w of the soft segment. This result is the same as that observed for cationic WBPU.[16] There are two reasons that can be explained. First, the hard segment content in PU becomes lower upon increasing the M_w of soft segment, which decreases the major interaction between PU and polyamide (urethane linking). Second, the soft segment content of PU is increased comparatively, increasing the hydrophilicity of PU. Therefore, the PU is incompatible with the hydrophobic nylon.

Thermal properties

The thermal properties of WBPU containing different polyols as a soft segment were studied by differential scanning calorimetric analysis (DSC) and shown in Figure 5. In this study, the PEGME does not easily form crystals because the M_w of PEGME chain is too short (only 750 g/mol).[18] In this figure, the temperature area of the endothermic peak at −70 to −10°C is the glass transition temperature (T_gs) of the soft segment, and that at 20–40°C is the melting point of the soft segment (T_ms). The data are shown in Table II. The T_gs and T_ms of nonionic PU with side chain are lower than those of other ionic PUs.[16] This is due to the fact that the presence of PEGME in the side chain of PU posses greater steric hindrance to the microstructure of PU causing a loose arrangement of PU molecular chains.

Figure 6 shows the effect of soft segment composition on T_gs of PU film. As seen from Figure 6, the T_gs of PCL-PU is the highest, followed in the right order by ether-type PEG-PU, and PPG-PU, with PTMG-PU showing the lowest T_gs. Since the PCL-PU has a polarity group (—COO) that can generate stronger intermolecular hydrogen bonding between the ester group (—COO—) and urethane group (—NHCOO—) of the hard chain, the polymer molecular chain is difficult to rotate resulting in a higher T_gs. In the case of polyether PU, the T_gs of PEG-PU is the largest that is attributed to its molecular structure that folds easily to crystallize. The amorphous molecular chain is impeded by the molecular chains present in the crystallization area. As for the influence of the M_w of soft segment upon T_gs, the T_gs values decrease with the increase of the soft segment length following a Fox-Flory equation because a greater M_w is not restrained from the hard segment.[21] Because of the presence of four methylenes in the soft segment structure of PTMG-PU, it
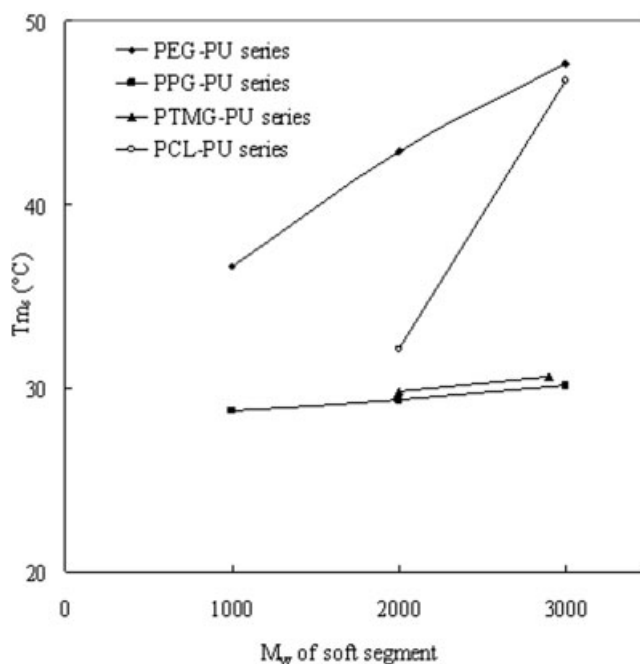

Figure 7 The influence of soft segment composition on T_ms of PU casting film.
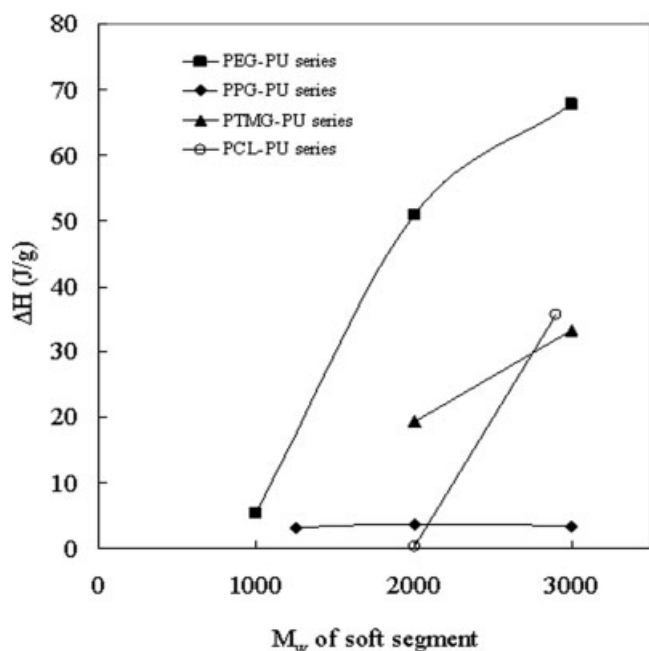
Figure 8 The effect of soft segment composition on the ΔH of PU casting film.

has less cohesive energy[20] than PEG-PU that contains two methylenes. As a result, its T_gs value is the lowest. Figure 7 displays the influence of soft segment composition on T_ms of PU casting film. The T_ms of PEG-PU is also the highest, and follows the order: PCL-PU > PTMG-PU > PPG-PU. The PEG-PU has the highest T_ms, which is due to the molecular structure that folds easily yielding to crystallization. In addition, although the PCL-PU has a polarity group (—COO) that can generate stronger intermolecular hydrogen bonding between the ester group (—COO—) and urethane group (—NHCOO—) of the hard chain, but its T_ms may greatly affected by the hydrophilic side chain PEGME resulting in lower T_ms than PEG-PU and disappear of crystallization of PCL-PU with M_w of 1250. The lowest T_ms of PPG-PU is attributed to its molecular chain with steric hindrance that has a methylene side chain (shown in Table II), which is difficult to orient and crystallize in the microstructure.

Figure 8 shows the effect of soft segment composition on the ΔH of PU casting film, and the trend is similar as that shown in Figure 7. This result is in relation with the degree of crystallization of the soft segment. Both the T_ms and ΔH increase upon increasing the polyol length. This is because the cohesive energy of the soft segment increases as the M_w of the soft segment increases, and the comparative PEGME content in the side chain of PU decreases. As a result, the steric hindrance of the soft segment is reduced. Thus, the arrangement of molecular chain and the degree of crystallization of the soft segment were not affected by PEGME.

Physical properties of coated fabric

The contact angle between pure water and coated fabric and the WVP of coated fabric are also shown in Table II. This contact angle between pure water and PU coated fabric is different from the contact angle between PU dispersion and untreated nylon fabric in solution properties of last section, which is dropping pure water on the PU coated fabric. Table II shows the contact angle with the trend of PCL-PU > PTMG-PU > PPG-PU. The contact angle of coated fabric with PEG-PU is not discussed since the coated fabric with PEG-PU presented a sticky state (against film forming) under a standard atmosphere because of its strong ability to absorb moisture. This result displays that the contact angle increases with the increasing of PU's hydrophilicity. Besides, we found that the contact angles conform with the degree of crystallization of PU casting film. This indicates that the hydrophobicity of PU film increases with the degree of crystallization. The WVP (Fig. 9) follows the order: PEG-PU > PTMG-PU > PPG-PU > PCL-PU. Overall, the ether-based PU has higher WVP than ester-based PU (PCL-PU) because the ester-based PU has a large hydrogen bonding and hydrophilicity, which resulted in lower WVP. Contrarily, the PEG-PU has the largest WVP because the hydrophility of PEG has the advantage of moisture mobility in the film. The WVP of PPG-PU is less than PEG-PU and PTMG-PU because it contains a hydrophobic methylene side chain. Regarding the influence on M_w of soft segment, it has been found that
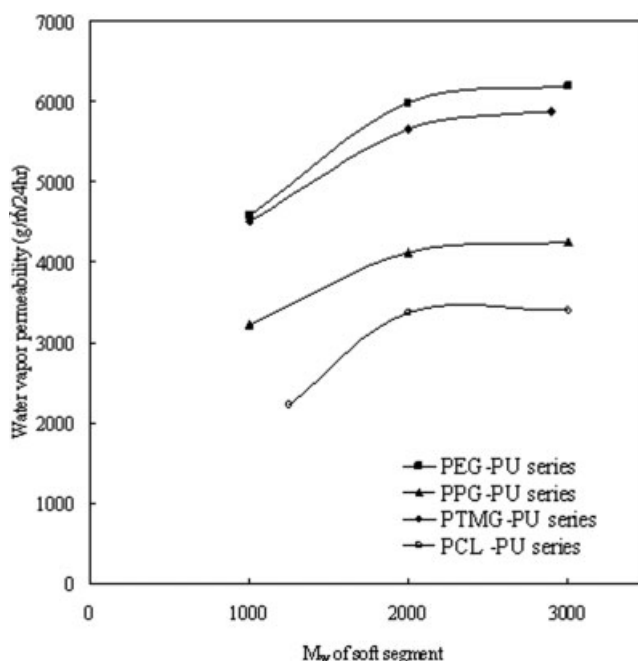

Figure 9 Relationship between soft segment composition and WVP of coated fabric.

the WVP of coated fabric increases upon increasing M_w, the trend is gradual when the M_w of soft segment is over 2000 g/mol. The content of hard hydrophobic segment in PU decreases upon increasing the soft segment, but when the M_w of the soft segment is over 2000 g/mol, the WVP became gradual due to the increase in crystallinity.

CONCLUSIONS

This article focused on exploring the effect of the soft segment composition on the physical properties of PEGME-containing nonionic WBPU. The synthesized PU is also applied on nylon fabric for WVP. The results are summed up in the following.

1. With regard to the solution properties of PU dispersions, the surface tension of PU dispersion follows the order: PEG-PU > PTMG-PU > PPG-PU > PCL-PU, the surface tension increases with the increasing of soft segment length. The particle size of the soft segment dispersion follows the order: PCL-PU > PPG-PU > PEG-PU > PTMG-PU. The contact angle of PU dispersion to nylon fabric increases as the molecular weight of the soft segment increases.

2. As for the thermal property of PU casting film, the T_gs of PCL-PU is the highest, followed by PEL-PU, PTMG-PU, and PPG-PU, and the T_gs decreases upon increasing the length of soft segment. The T_ms of PU casting film is in the order of PEG-PU > PCL-PU > PPG-PU > PTMG-PU, and the T_ms increases upon increasing the length of soft segment.

3. The Coated-PU with PEG-PU has an excellent WVP, and follows the torder: PEG-PU > PTMG-PU > PPG-PU > PCL-PU, and WVP increases upon increasing the length of the soft segment.

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