

ARTICLE

Synthesis, Surface Activity, and Antifogging Property of Triethanolamine Monolaurate Ester

Meng-jun Cai, Qing-qing Li, Xue-mei Chen*

National Engineering Research Centre of Ultrafine Powder, School of Chemical Engineering, East China University of Science and Technology, Shanghai 200237, China

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Triethanolamine monolaurate ester was synthesized by lauric acid and triethanolamine (TEA) with a molar ratio around 1:1 and the esterification process was investigated and optimized. The esterification product of lauric acid with TEA was characterized by infrared spectra and nuclear magnetic resonance. The surface tension (γ_{CMC}) at the critical micelle concentration (CMC) was determined, and the antifogging properties of triethanolamine laurate ester on low-density polyethylene (LDPE) films were also measured. The results indicated that the yield of triethanolamine monolaurate ester was more than 69% under optimized esterification condition, the CMC value and γ_{CMC} of esterification product was 0.91 $\mu\text{g/mL}$ and 22.1 mN/m in aqueous solution at 25 $^{\circ}\text{C}$, respectively. The first-drop time and ten-drop time was 257 and 86 s, respectively, and the antifogging duration of triethanolamine laurate ester on the surface of LDPE film at 60 $^{\circ}\text{C}$ was more than 150 h.

Key words: Triethanolamine, Laurate, Surfactant, Antifogging property

I. INTRODUCTION

Agricultural films are widely used as greenhouse cover to improve the efficiency of crop production. Normal polyethylene films used in greenhouses have no affinity with water. There is a significant difference between the surface tensions of the water and the hydrophobic polymer, which contributes to the forming of droplets when saturated water vapor condenses on the cold surface of plastics, resulting in scattering sunlight, reducing the transmission of light, weakening the efficiency of photosynthesis and affecting the growth of crops [1].

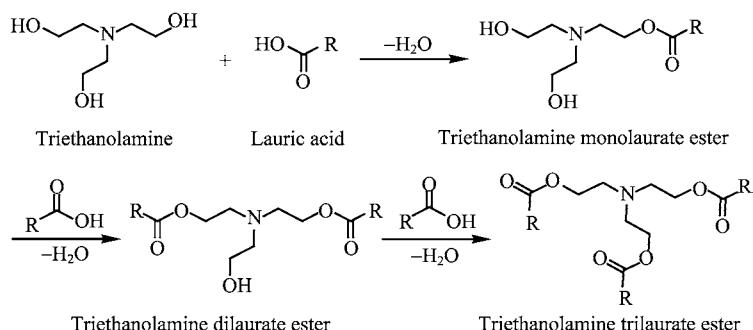
In order to solve these problems, the addition of antifogging agents is a preferred solution to modify the surface tension in industry because of its lower costs. A small quantity of antifogging agents dissolving in the water droplets can decrease the surface tension of the droplets, and the droplets spread into an invisible continuous thin layer on the surface of the films. The structures of antifogging agents possess typical characteristics of surfactants, such as sorbitan, glycerol, polyglycerol, polyoxyethylene esters, and so on [2, 3].

Triethanolamine aliphatic esters and their quaternary ammonium salts [4] are widely used as softening agents [5, 6], foam indicators [7, 8], antistatic agents [9], grinding aids [10] and antimicrobials [11] in recent years and are becoming favorable because of their eco-friendly na-

ture and excellent biodegradability [12]. However, the esterification product of aliphatic acid with TEA is a kind of mixture of mono-, di- and tri-ester [13], and the ultimate performance of the product is corresponding to the molar ratio of the three of them. Higher mono-ester content gives better dispersibility and stability. Higher tri-ester content results in better softening but poor stability [14]. Some researches concerned about the di-estercompounds of aliphatic acid with TEA [15], while few concerned about the mono-ester compounds, especially for the mono-ester compound of lauric acid with TEA. Furthermore, some catalysts, such as hypophosphorous acid, sodium methoxide, *p*-toluene sulfuric acid and so on, are employed in conventional esterification process of aliphatic acid with TEA, which bring about a series of environmental impacts and applied problems [16]. To tackle these problems, whether to use environmentally friendly catalysts or not is becoming a new research hotspot.

In recent years, our research group have studied several metal-organic coordination complexes [17–22], which could be used as antimicrobials in plastics greenhouse films. In this work, we prepare some organic compounds used as antifogging agents in plastic greenhouse films, and synthesize triethanolamine monolaurate ester by esterification reaction of lauric acid with TEA. The esterification products are characterized by infrared spectra (IR) and nuclear magnetic resonance (NMR) and their surface activity are estimated by surface tensions (γ_{CMC}) at the critical micelle concentrations (CMC). The antifogging properties of triethanolamine laurate ester on the surface of low-density

* Author to whom correspondence should be addressed. E-mail: xmchen@ecust.edu.cn, Tel.: +86-21-64252439, FAX: +86-21-64253317



Scheme 1 The synthetic procedure for preparation of triethanolamine laurate ester, where $R=CH_3(CH_2)_{10}$.

polyethylene (LDPE) films, such as the first-drop time, ten-drop time and antifogging duration, are also investigated.

II. EXPERIMENTS

A. Reagents and materials

Lauric acid (CP) and triethanolamine (AR) were purchased from Shanghai Lingfeng Chemical Reagent Co. Ltd., China. Acrylic acid-hydroxyl ethyl methacrylate copolymer (AA-HEMA, solid content 10wt%) was performed using a reported procedure [23]. LDPE films with a thickness of 0.08 mm, some commercial antifogging films and other analytical grade chemical reagents, were purchased commercially and used without purification.

B. Synthesis of triethanolamine monolaurate ester

Lauric acid and TEA was mixed with a molar ratio around 1:1 and the reaction was carried out at a set temperature in an oil bath under nitrogen buffer gas. A moderate vacuum was used to remove the produced water in esterification process until the acid value was less than 5 mg KOH/g. In order to refine the resultant, a small quantity of 10wt% sodium hydroxide solution was dropwise added to the reaction solution to make the lauric acid form soluble sodium laurate. Then sodium chloride saturated solution (the volume ratio of sodium chloride saturated solution to triethanolamine laurate ester is about 2:8) was added to crude product under gentle agitation. The oil layer was separated from the mixture with a separatory funnel and the aqueous layer was removed. The yellowish triethanolamine laurate ester, with certain viscosity, was obtained after the resultant was dehydrated by anhydrous magnesium sulfate.

The esterification product of lauric acid with TEA could be a kind of mixture of mono-, di- and tri- ester (Scheme 1).

The acid value (AV) of free aliphatic acid was determined according to GB/T 5530-2005 [24] and calculated by Eq.(1) [16].

$$AV = \frac{56.1VC}{m} \quad (1)$$

V is the volume of KOH standard solution used in the titration, C is the concentration of KOH standard solution, m is the mass of sample.

C. Preparation of antifogging films

Antifogging coatings were prepared by mixing 1 g triethanolamine laurate ester, 1 g AA-HEMA and 98 g deionized water, and slowly stirred at room temperature for 15 min. 3 mL of them were coated on the surface of cleaned LDPE films (50 mm×50 mm) by means of roller. The antifogging films were obtained after the antifogging coatings dried at room temperature for 24 h.

D. Characterization

Infrared spectra (IR) were recorded from 4000 cm^{-1} to 400 cm^{-1} using a Bruker Vector 33 spectrometer equipped with a diamond reflection accessory. Proton nuclear magnetic resonance ($^1\text{H NMR}$) spectra were obtained on a Bruker DRX spectrometer operating at 400 MHz using CDCl_3 as solvent. Surface tension values were used to calculate CMC using a BZY-1 tensiometer equipped with a platinum ring.

E. Surface tension test

The tension-meter was calibrated using three distilled water. A series of aqueous solution of triethanolamine laurate ester were prepared with a concentration range of $10^{-3}\text{--}10^{-9}\text{ g/mL}$ at 25°C . The platinum ring was washed with distilled water and cleaned by burning method. Record the tension-meter reading as the surface tension value (γ) when the bottom of the platinum

ring was immersed into the aqueous solution of prepared ester. Each surface tension value was averaged by at least three consistent measured values.

F. Optical transmittance

The optical transmittance of LDPE film was determined using a UV-2700 ultra-vis spectrophotometer ranging from 400 nm to 800 nm by a piece of rectangular LDPE film, 43 mm in length and 14 mm in width, perpendicularly inserted in the spectrophotometer along the diagonal of colorimetric ware. Each optical transmittance was parallelly measured three times.

G. Antifogging properties

Beakers containing water, covered with LDPE films forming an angle of 15° with the horizontal, was kept in the water bath at 50 °C. Then the timer started immediately. The first-drop time (the time of the first droplet dripping down from the surface of LDPE film) and ten-drop time (the time of continuous ten droplets dripping down from the surface of LDPE films, after ten minutes from the first-droplet dripping down) at 50 °C was recorded according to the timer, respectively. The antifogging duration (the service life of the antifogging film) can be determined under the similar condition as testing the first-drop time except that the water bath temperature was 60 °C.

III. RESULTS AND DISCUSSION

A. Synthesis of triethanolamine monolaurate ester

In order to confirm the optimal esterifying condition of the triethanolamine monolaurate ester, some factors were investigated, such as reaction temperature and time, molar ratio of lauric acid to TEA, catalyst and so on.

1. Temperature and time effect

The conversion of triethanolamine laurate ester, expressed by acid value, was significantly affected by the reaction temperature and time (Fig.1). The conversion of triethanolamine laurate ester was obviously improved with the increase of reaction temperature from 130 °C to 170 °C. The higher the temperature, the shorter is the reaction time at the same conversion. However, the thermal degradation and/or oxidation reaction could make the color of the reaction system become dark at high temperature. After being esterified in a 1:1 molar ratio of lauric acid to TEA at 150 °C for 6 h, the conversion was more than 97% and tended to be a certain constant, while the acid value (3.15 mg KOH/g) is in good

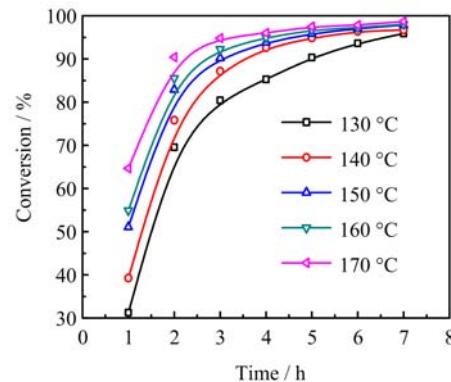


FIG. 1 The influence of reaction temperature and time on the conversion of triethanolamine laurate ester in a 1:1 molar ratio of lauric acid to triethanolamine without catalyst.

agreement with the required (less than 5 mg KOH/g) (Fig.1). Accordingly, considering the color of esterification product and the complexity of reaction process, it seemed that the appropriate reaction temperature was 150 °C and the corresponding reaction time was 6 h when the molar ratio of lauric acid to TEA was about 1:1.

2. Catalyst

Generally, esterification reactions of fatty acid with TEA have been performed by using homogeneous catalysts such as phosphorous acid, sodium methoxide, *p*-toluene sulfuric acid, sulfuric acid, and so on [25]. The effects of catalyst on the conversion of triethanolamine laurate ester were studied in a 1:1 molar ratio of lauric acid to TEA at 150 °C (Fig.2).

Sodium methoxide and *p*-toluene sulfuric acid were used as catalysts to investigate the relationship between the reaction time and conversion of triethanolamine laurate ester at 150 °C in the esterification process of lauric acid with TEA. The results showed that the conversion of triethanolamine laurate ester was high up to 98% when *p*-toluene sulfuric acid, a common homogeneous catalyst in the esterification process of aliphatic acid ester with TEA, was used as the esterification catalyst of lauric acid with TEA at 150 °C for 6 h. Accordingly, the conversion of triethanolamine laurate ester also reached 97.6% (closed to 98%) without catalyst (Fig.2). However, considering the toxicity and the corrosion of catalysts, the esterification reaction of lauric acid with TEA could be performed without catalyst.

3. Molar ratio of lauric acid to TEA

In general, the application performance of esterification product is determined by the molar ratio of mono-, di- and tri-ester of triethanolamine laurate, which is

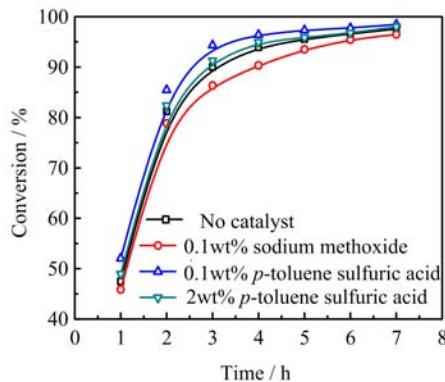


FIG. 2 The effect of catalyst on the conversion of triethanolamine laurate ester in a 1:1 molar ratio of lauric acid to triethanolamine at 150 °C.

mainly accommodated by the molar ratio of lauric acid to TEA in esterification process. In order to obtain the esterification product with a higher content of triethanolamine monolaurate ester, the esterification reaction was studied in a different molar ratio of lauric acid to TEA at 150 °C for 6 h (Fig.3).

The conversion of lauric acid was increased with the increase of molar ratio of lauric acid to TEA in an appropriate range. When the molar ratio of lauric acid to TEA was about 0.95:1, the conversion of lauric acid reached 97% (Fig.3). Increasing the molar ratio of lauric acid to TEA could promote further esterification. However, the reaction system became more and more viscous, and the content of di- and/or tri-ester of triethanolamine laurate ester was increased. In order to obtain the higher purity and yield of triethanolamine monolaurate ester, the molar ratio of lauric acid to TEA sounded to be 0.95:1.

From the above investigation, the results showed that the optimized esterification reaction of lauric acid with TEA could be carried out in a 0.95:1 molar ratio of lauric acid to TEA at 150 °C for 6 h under nitrogen buffer gas without catalyst.

B. IR spectra

In order to determine the structure of esterification product, the IR spectra of triethanolamine laurate ester (synthesized under the optimized condition), lauric acid and TEA are recorded in Fig.4. In the spectrum of prepared triethanolamine laurate ester (Fig.4(c)), the characteristic absorption bands of -COOH groups of free lauric acid at 1703 cm⁻¹ disappear [26] (Fig.4(a)), and the bands of stretching vibration of COO- groups appear at approximately 1740 cm⁻¹. The stretching vibrations of OH- and C-N absorption bands of TEA around 3393 and 1028 cm⁻¹ show upward shifts to 3400 and 1100 cm⁻¹ after being esterified [27] (Fig.4(b)). The distinct feature in the IR spectrum of prepared

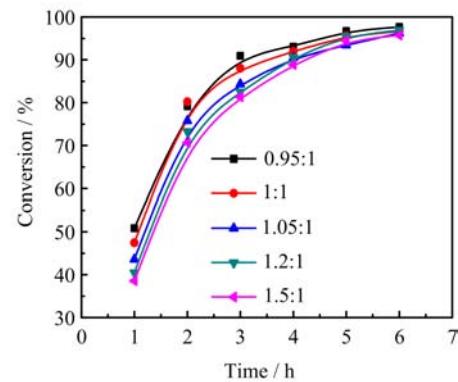


FIG. 3 The relationship of molar ratio and reaction time with the conversion of triethanolamine laurate ester at 150 °C without catalyst.

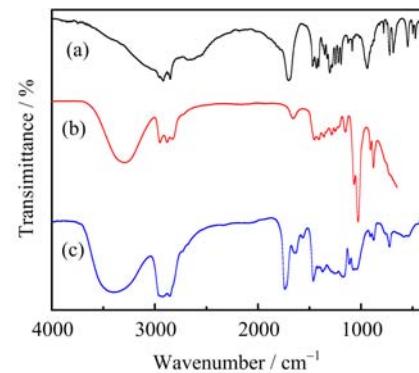


FIG. 4 IR spectra of (a) lauric acid, (b) triethanolamine, and (c) triethanolamine laurate ester.

triethanolamine laurate ester is the presence of vibration absorption of -C=O and -C—O—C around 1740 and 1172 cm⁻¹ respectively, which is in agreement with the long chain aliphatic ester [5].

C. ¹H NMR

The ¹H NMR spectrum for esterification product of lauric acid with TEA under optimized condition and the proton signals assigned as numbered in the formula are shown in Fig.5. The values of chemical shifts of proton and integrated areas of peaks depicted from Fig.5 are grouped in Table I [28].

¹H NMR spectroscopy was used to quantitatively estimate the content of mono-, di- and tri-ester in the esterification product. The chemical shifts, at 0.87 and 1.25 ppm, assigned to the methyl groups and methylene groups of laurate, indicate that laurate group is present in the structure of esterification product of lauric acid with TEA. Similarly, the resonance, at 2.73 ppm attributed to the N(CH₂)₃ groups of TEA, indicates that parts of TEA groups are present in the structure of

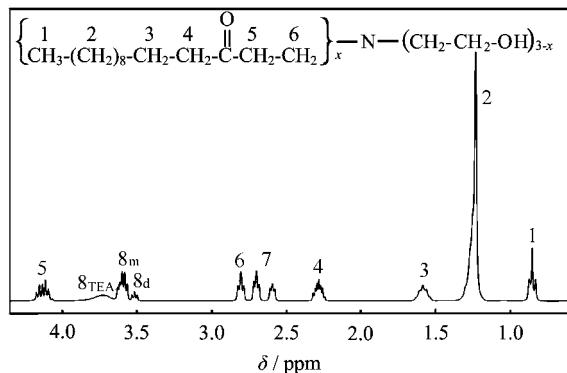


FIG. 5 ^1H NMR spectrum of esterification product in a 0.95:1 molar ratio of lauric acid to triethanolamine at 150 $^{\circ}\text{C}$ for 6 h.

esterification product of lauric acid with TEA. The signals at 3.56 and 3.62 ppm, corresponding to the mono-ester and di-ester, are in good agreement with the ^1H NMR of aliphatic ester reported in Ref.[27]. It could be concluded that the triethanolamine laurate ester was synthesized successfully.

According to the ^1H NMR of esterification product, the content of triethanolamine mono-laurate ester is calculated as follows. Values of I_{H} (integral area of each hydrogen) and X (average esterification degree) are calculated using the Eq.(2) and Eq.(3).

$$I_{\text{H}} = \frac{1}{12} (I_5 + I_6 + I_7 + I_{8_{\text{TEA}}} + I_{8_m} + I_{8_d}) \quad (2)$$

$$X = \frac{I_1 + I_3 + I_4}{7I_{\text{H}}} \quad (3)$$

The average molecular weight (M_p) of esterification product can be calculated according to Eq.(4).

$$M_p = (M_{\text{acid}} - 18)X + M_{\text{TEA}} \quad (4)$$

The moles of mono-ester n_m and di-ester n_d of triethanolamine laurate ester can be calculated using Eq.(5) and Eq.(6).

$$n_m = \frac{A_{8_m}}{4I_{\text{H}}} \quad (5)$$

$$n_d = \frac{A_{8_d}}{2I_{\text{H}}} \quad (6)$$

The content of mono-ester and di-ester in esterification product is calculated according to Eq.(7) and Eq.(8), respectively.

$$\omega_m = \frac{n_m M_m}{M_p} \times 100\% \quad (7)$$

$$\omega_d = \frac{n_d M_d}{M_p} \times 100\% \quad (8)$$

M_{acid} is the molecular weight of lauric acid, M_{TEA} is the molecular weight of TEA, M_m is the molecular weight

TABLE I Chemical shifts and integrated areas of proton of the esterification product in a molar ratio of lauric acid to triethanolamine of 0.95:1 at 150 $^{\circ}\text{C}$ for 6 h.

^1H identification	δ/ppm	Integration area/Arb. unit
1	0.87	1.52
2	1.25	7.67
3	1.61	1.12
4	2.30	1.12
5	4.15	1.00
6	2.82	1.03
7	2.73	1.01
8_m	3.62	1.42
8_d	3.56	0.19
8_{TEA}	3.71	0.81

of triethanolamine monolaurate ester, M_d the molecular weight of triethanolamine dilaurate ester, A_{8_m} is the hydrogen integral area of 8_m , A_{8_d} is the hydrogen integral area of 8_d .

According to the data in Table I and the equations above, the content of mono-ester and di-ester of triethanolamine laurate ester is about 69% and 28%, respectively. The high content of triethanolamine monolaurate ester makes the esterification products more soluble in water, which also contributes to its application effect as antifogging agent.

D. Surface tension

The surface tension values of triethanolamine laurate ester aqueous solution with concentration of 10^{-6} g/mL, prepared different reaction molar ratios of lauric acid to TEA at 150 $^{\circ}\text{C}$ for 6 h without catalyst, are shown as follows. The surface tension values are 22.1, 23.2, 24.2, 25.8, 37.9 mN/m for ester prepared with different molar ratio of lauric acid to triethanolamine of 0.95:1, 1:1, 1.05:1, 1.2:1, 1.5:1.

The surface tension of esterification product ranges from 22 mN/m to 38 mN/m. During the esterification process, the yield of di- and tri- ester increases with the increase of molar ratio of lauric acid to TEA, which is not likely to reduce the surface tension effectively. The minimum value of surface tension under the optimized esterification condition is 22.1 mN/m attributed to the presence of one ester group between the long hydrocarbon chain and two TEA hydroxyl groups in the product, which is in good agreement with the content of triethanolamine monolaurate ester in the esterification process of lauric acid with TEA.

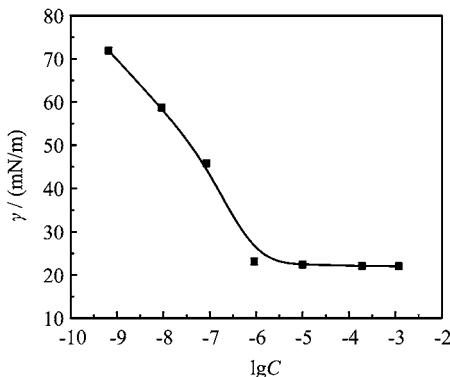


FIG. 6 Surface tension γ versus logarithmic concentration of esterification product prepared with a molar ratio of lauric acid to triethanolamine of 0.95:1 at 150 $^{\circ}\text{C}$ for 6 h.

E. Critical micelle concentration

The critical micelle concentration (CMC) is defined as the concentration of surfactant in which the micelles are spontaneously formed. The surface tension of the prepared triethanolamine laurate ester aqueous solution at 25 $^{\circ}\text{C}$ against the logarithm of the solute concentration ($\lg C$) is plotted in Fig.6.

In the range of low concentration, the surface tension declines with the increase of the concentration. It is suggested that all the prepared triethanolamine laurate esters are adsorbed in the interface layer of air and water [27]. Further increasing the concentration, the surface tension remains a constant, which indicates that the micelles of prepared triethanolamine laurate ester are formed in bulk aqueous solution above a given concentration [29].

The CMC value of esterification product of lauric acid with TEA in optimized condition is about 0.91 $\mu\text{g}/\text{mL}$ ($10^{-6.04}$ g/mL in Fig.6). Compared with other surfactants with high CMC, it is appreciated for triethanolamine laurate ester to have a low CMC because it can be used in less amount to achieve the same effect [30, 31]. The lower the CMC of a given surfactant, the more stable is the micelle. It is very important for antifogging agents to obtain excellent antifogging performance. Because the antifogging agents can be diluted and washed away from the surface of greenhouse covers when blended with droplets, and shorten the antifogging duration of antifogging film. Consequently, the prepared triethanolamine laurate ester, with the low critical micelle concentration, is a promising candidate antifogging agent in greenhouse cover.

F. Antifogging property

To assess the antifogging efficiency of triethanolamine laurate ester, the relationship between antifogging performance and the content of triethanolamine laurate es-

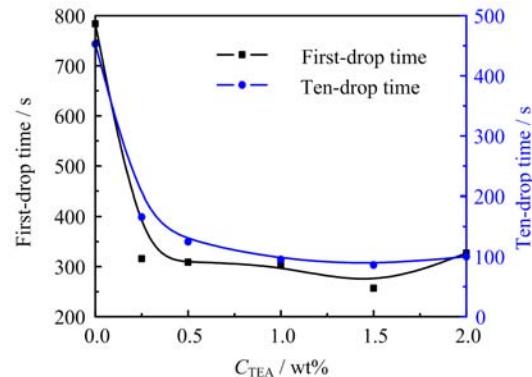


FIG. 7 The effect of the content of triethanolamine laurate ester in antifogging coatings on the first drop time and ten-drop time.

ter in antifogging coatings was investigated and the results are shown in Fig.7. For comparison, the blank experiment is performed without AA-HEMA in the antifogging coatings, and the first-drop time and ten-drop time of triethanolamine laurate ester is 160 and 85 s respectively, when the concentration of triethanolamine laurate ester is about 1.5wt%.

Increasing the content of triethanolamine laurate ester in antifogging coatings from 0wt% (no triethanolamine laurate ester) to 2wt%, the first-drop time and ten-drop time is gradually decreased until reaching a minimum of 257 and 86 s, respectively. Further increasing the content of triethanolamine laurate ester resulted in an increase of the first-drop time and ten-drop time (Fig.7). In the range of low content, the higher the content of triethanolamine laurate ester, the easier the droplets spread out on the surface of the films. Furthermore, the first-drop time and ten-drop time will be shortened. Both the first-drop time and ten-drop time approximate a minimum values when the content of triethanolamine laurate ester is about 1.5wt%. Further increasing the amount of triethanolamine laurate ester in the antifogging coatings, the antifogging coatings is a slight opaque owing to the crosslinking reaction of triethanolamine laurate ester with AA-HEMA. The crosslinking results lead to the decrease of surface activity of antifogging coatings and the increase of the first-drop time and ten-drop time. Therefore, the optimum concentration of triethanolamine laurate ester in the antifogging coatings is 1.5wt%.

A series of first-drop time, ten-drop time and light transmittance property of the prepared antifogging films were performed and the results are summarized in Table II.

The LDPE film covered with antifogging coatings has a shorter first-drop time and ten-drop time than the commercial antifogging film, which is also superior to the standard value in GB/T 19603-2004 [32]. At the same time, the antifogging duration of the LDPE film

TABLE II The first-drop time t_1 , ten-drop time t_{10} , antifogging duration t_{anti} and optical transmittance η_t of the prepared coatings.

	t_1/s	t_{10}/s	t_{anti}/h	$\eta_t/\%$
GB/T 19603-2004	600	120		
Commercial antifogging film	410	105	72	76.9
Prepared antifogging film	257	86	>150	75.4

covered with antifogging coatings is longer than that of the commercial antifogging film. The light transmittance property of LDPE film covered with antifogging coatings displays a negligible loss attributed to the darker of the antifogging coatings color and the thicker of the antifogging film. In addition, the increase of surface roughness of LDPE film causes a decrease of the light transmittance owing to the surface scattering, which is in agreement with the results reported in the previous literatures [33, 34]. The LDPE film covered with antifogging coatings containing triethanolamine laurate ester has a better antifogging performance and smaller surface tension than the commercial antifogging film. Therefore, as a high-effective antifogging agent, triethanolamine laurate ester has a tempting application prospect in greenhouse cover.

IV. CONCLUSION

Triethanolamine monolaurate ester is successfully synthesized in a 0.95:1 molar ratio of lauric acid to TEA at 150 °C for 6 h without catalyst, and the yield of triethanolamine monolaurate ester is more than 69wt%. The novel synthesized triethanolamine monolaurate ester has a good solubility in water and a good surface activity. The CMC and γ_{CMC} of esterification product of lauric acid with TEA is 0.91 $\mu\text{g}/\text{mL}$ and 22.1 mN/m at 25 °C in aqueous solution respectively. It shows that triethanolamine monolaurate ester is a good kind of non-ionic surfactant and can be used as antifogging agent in greenhouse cover.

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