

CHAPTER IV
REMOVAL OF SOLVENT-BASED INK FROM PRINTED SURFACE OF
HIGH-DENSITY POLYETHYLENE BOTTLES BY
ALKYLTRIMETHYLAMMONIUM BROMIDES: EFFECTS OF pH,
TEMPERATURE, AND SALINITY

5.1 Abstract

The effects of pH, temperature, and salinity on the removal of solvent-based ink from high-density polyethylene (HDPE) surfaces were investigated using three alkyltrimethylammonium bromides (i.e., dodecyl-, tetradecyl-, and hexadecyl-trimethylammonium bromide or DTAB, TTAB, and CTAB, respectively). Ink removal increases with increasing concentration, increasing pH, decreasing temperature, and increasing salinity of the CnTAB solutions. The zeta potential of ink becomes more positively charged with increasing pH as well as concentration and alkyl chain length of CnTAB, indicating that adsorption of CnTAB on ink pigment occurred more readily with an increase in any of those parameters. The solubilization of epoxy ink binder was found to increase with increasing surfactant concentration, decreasing temperature, and increasing salinity of the CnTAB solutions. Adsorption of cationic surfactant onto the ink pigment particles and solubilization of ink binder molecules into surfactant micelles are important mechanisms causing ink removal from the plastic surface.

Key-words: deinking; alkyltrimethylammonium bromide; high-density polyethylene; zeta potential; solubilization

5.2 Introduction

Steady growth of demand in plastics leads to increasing demand for natural gas and petroleum as the raw materials for plastic production. An increase in the utilization of plastics translates into a steady growth of plastic wastes as a

consequence. A means for dealing with such a problem is through recycling, but it has been reported that recycled plastics often bear poor physical and mechanical properties in comparison with those of virgin polymers [1,2]. For printed plastics, the presence of residual inks can be a primary cause of the deterioration of polymer properties. Previous work, for example, reported the deterioration effect of residual ink on physical properties of re-extruded polyethylene [3]. Therefore, removal of ink from the plastic surface prior to recycling is a prerequisite. Generally, ink can be removed by either physical [4] or chemical [3,5-7] means. Chemically, ink can be removed by organic solvents, but, due to their toxicity, alternative deinking media are desirable. Due to their biodegradability, non-toxicity, and non-volatility, aqueous solutions of surfactants are considered good alternatives for such a task.

Previous studies on surfactant-based deinking process have been carried out on surfaces of either plastic films [3,5,7] or rigid plastics [6] in order to remove either water-based [3,5] or solvent-based [6,7] ink using an anionic surfactant [3], nonionic surfactants [3,5,7], an amphoteric surfactant [3,7], or cationic surfactants [3,6,7]. For either water- or solvent-based ink, cationic surfactants were the most effective in ink removal at concentrations well above their critical micelle concentration (CMC) and at high pH levels (generally greater than or equal to 11) [3,6,7]. Furthermore, increased temperature during deinking, increased pre-soaking time in the surfactant solutions prior to mechanical agitation, and increased shaking time helped increase deinking efficiency [5-7]. The cationic surfactants which have been previously investigated were hexadecyltrimethylammonium bromide (CTAB; $C_{16}H_{33}^+N(CH_3)_3Br^-$) [3,6,7] and hexadecylpyridinium chloride (CPC; $C_{16}H_{33}^+N(C_5H_5)Cl^-$) [3], but CPC was found to degrade at high pH levels.

In the first paper of this series [8], we reported the use of three alkyltrimethylammonium bromides (i.e., dodecyl-, tetradecyl-, and hexadecyl-trimethylammonium bromide or DTAB, TTAB, and CTAB, respectively) to remove blue solvent-based ink from a printed surface of high-density polyethylene (HDPE) bottles. All studies were carried out at a fixed temperature and pH of 30°C and 12, respectively. Both the increase in the alkyl chain length and the surfactant concentration increased the deinking efficiency. Complete deinking was achieved at

concentrations of about 3, 8, and 24 times of the CMC of CTAB, TTAB, and DTAB, respectively. For CTAB, ink removal began at a concentration close to its CMC and increased significantly at concentrations greater than its CMC, while, for TTAB and DTAB, significant deinking was only observed at concentrations much greater than their CMC's. Both the wettability of the surfactant on the ink surface and the solubilization of ink binder in the surfactant micelles increased with an increase in both the alkyl chain length and the surfactant concentration. Finally, we proposed a mechanism for the removal of a solvent-based ink from the HDPE surfaces as consisting of four main steps: 1) surfactant adsorption on both printed and unprinted HDPE surfaces; 2) solubilization of ink binder in micelles; 3) detachment of ink pigment from HDPE surfaces; and 4) stabilization and dispersion of the detached ink particles.

In the present contribution, the effect of pH, temperature and addition of a simple salt, sodium chloride, on deinking for this same system using DTAB, TTAB and CTAB was studied. The effect of their concentration on zeta potential of ink particles and solubilization of ink binder pigment particles was investigated under different deinking conditions [i.e., pH = 11 – 12; temperature = 30, 35, 40, and 45°C; and addition of 0.1 M sodium chloride salt (NaCl)] from that performed in our previous work [8] (i.e., pH = 12 and temperature = 30°C).

5.3 Experimental

5.3.1 Materials

High-density polyethylene (HDPE) sheet samples were obtained from commercial drinking water blow-molded bottles produced by SVB Drinking Water Company (Thailand). Printing of these bottles was done at CK-Plastic Company (Thailand). Prior to printing, the HDPE bottles were flame-treated. A blue solvent-based ink formulation (UPE-B4009/2) from Uni Ink Company (Thailand) was screen-printed on the treated HDPE surfaces. The printed part of the bottles were then cut into 8 mm × 40 mm sheets.

N-dodecyltrimethylammonium bromide (DTAB) (99% purity; powder) and n-tetradecyltrimethylammonium bromide (TTAB) (98% purity; powder) were purchased from Aldrich (USA), while n-hexadecyltrimethylammonium bromide (CTAB) (98% purity; powder) was purchased from Fluka (Switzerland). Sodium chloride (NaCl) was purchased from Aldrich (USA). Sodium hydroxide (NaOH) (98% purity), purchased from EKA Noble (Sweden), and hydrochloric acid (HCl) (AR grade), purchased from BHD (Germany), were used for pH adjustment. All of these chemicals were used as-received.

Ink powder was prepared by mixing the as-received ink with an appropriate solvent and the mixture was evenly pasted onto a 15" × 10" glass plate. The layer of ink was made as thin as possible to hasten the drying. The painted glass was later dried in an oven at 60°C over night, after which time the ink was scraped off from the glass surface, ground in a mortar, and finally sieved into powder of about 100 mesh or less (i.e., <150 μm). The as-prepared ink powder was kept in a desiccator prior to further use. The chemical composition of the ink powder was investigated by a Fourier-transformed infrared spectrometer (FT-IR; Bruker Vector 3.0), operating at a resolution of 4 cm⁻¹ and a frequency range of 4000 to 400 cm⁻¹.

5.3.2 Zeta potential Measurement

A very small amount of the as-prepared ink powder was added into a CnTAB solution. The pH level of the mixture was adjusted to either 11 or 12 and the mixture was stirred for 24 hours. The temperature of the mixture was equilibrated at 30 ± 1°C. The as-prepared mixture was then transferred to an electrophoretic cell of a zeta meter (Zeta Meter, 3+), equipped with a microscope module. After applying a suitable voltage according to the solution conductivity, the time for any visible ink particle to move for a certain distance was measured. For a given data set, at least 20 ink particles were monitored, from which the average time was calculated. The average time was then used to calculate the average zeta potential value for that particular data set.

5.3.3 Deinking Experiments

To investigate the effects of alkyl chain length, surfactant concentration, pH, temperature, and salinity on deinking efficiency, DTAB, TTAB, and CTAB solutions were prepared at different concentrations, pH levels (i.e., between 11 and 12), temperatures (i.e., between 30 and 45°C), and salinity (i.e., with or without 0.1 M NaCl). The as-prepared printed HDPE specimens were first pre-soaked (without shaking) in a 15 mL surfactant solution for 2 hours and further soaked while being shaken at 200 cycles/min for another 2 hours in a shaking water bath. Then, the specimens were washed with deionized water and later dried in open air at room temperature overnight. The amount of ink on the plastic before and after deinking was measured using the optical scanning method [6]. In this method, each plastic specimen was carefully positioned on and scanned by a HP ScanJet 4C optical scanner using the factory settings. The scanned files were analyzed by a copy of Adobe Photoshop 5.5 by counting the number of pixels (proportional to the amount of ink present) on the plastic surface. The amount of ink removed (%) was then calculated based on the following relationship [6]:

$$\text{Ink removed (\%)} = \left[\frac{\text{pixels}_{\text{before deinking}} - \text{pixels}_{\text{after deinking}}}{\text{pixels}_{\text{before deinking}}} \right] \times 100. \quad (1)$$

5.3.4 Critical micelle Concentration Measurement

At $30 \pm 1^\circ\text{C}$, the critical micelle concentration (CMC) of pure DTAB, TTAB, and CTAB solutions was determined based on conductivity, using a conductivity meter (Orion, 125). Surface tension values, used to determine the CMC of the CnTAB solutions in the presence of 0.1 M NaCl, were measured by a drop shape analyzer (KRÜSS, DSA10-Mk2). The temperature was maintained at $30 \pm 1^\circ\text{C}$. For both surface tension and conductivity, the CMC is the surfactant concentration at which there is an abrupt change in the slope of the property as a function of the surfactant concentration.

5.3.5 Solubilization Measurement

The concentration of solubilized ink in CnTAB solutions with or without the presence of 0.1 M NaCl was indicated by first mixing 10 mg of the as-prepared ink powder in a 15 mL CnTAB solution at pH 12 in a 50 mL Erlenmeyer flask. The flask was then placed in a shaking water bath with the temperature of the water being varied between 30 and 45°C and the shaking frequency of the sample

stage being set at 200 cycles/min for 4 hours. The solution was later filtered to remove undissolved ink particles using filter paper (Whatman no. 7402-001; average pore size = 0.2 μm). The UV absorbance of the filtrate was measured by a UV-visible spectrophotometer (Shimadzu, UV-2550) at a wavelength of 273.8 nm. The absorbance for accurate measurement is in the range of 0.1 to 0.8. If the sample solution showed an absorbance greater than this range, it was further diluted. The absorbance versus the concentration of CnTAB solutions is a qualitative measure for ink binder solubilization in the CnTAB solutions as increased absorbance corresponds to increased solubility of the binder in the surfactant solutions due to increased solubilization in micelles.

5.4 Results and Discussion

5.4.1 Zeta potential of Ink

The effects of initial concentration and pH on zeta potential of ink in CnTAB solutions are shown in Figure 1. For a given type of CnTAB solution, the zeta potential of ink pigment particle at pH 12 is consistently greater than that at pH 11 over the entire concentration range investigated. For a given pH, an increase in the initial concentration of the CnTAB solutions increases the zeta potential of ink pigment particles due mainly to the adsorption of the positively-charged head groups of the CnTAB monomers onto the initially negatively-charged ink pigment surfaces. This was possible because the point of zero charge (PZC) of the ink particles in water was observed at a pH of about 3.3 [8], which means that the ink particles exhibit a positive charge at $\text{pH} < 3.3$, while they are negatively charged when $\text{pH} > 3.3$. The particles became more negatively charged with increasing pH. As a result, the adsorption of CnTAB monomers on the ink particles could occur much better at pH 12 than at pH 11, causing the particles to become more positively charged at pH 12. This “charge reversal” effect is commonly observed when ionic surfactants adsorb onto oppositely-charged solid surfaces. In the presence of cationic surfactants, the zeta potential reaches a plateau at approximately the CMC because surfactant adsorption onto solids levels off above the CMC due to the surfactant monomer

concentration (and therefore thermodynamic activity) becoming constant when micelles form at or above the CMC.

Figure 2 shows the effects of alkyl chain length and pH on the zeta potential of ink pigment particles in CnTAB solutions. For a given initial concentration and pH of CnTAB solutions, the zeta potential of pigment increased with increasing alkyl chain length both below and above the CMC. The longer the alkyl chain length, the stronger the van der Waal interaction of the tail groups with the pigment surface [8], which, in turn, helps render more CnTAB monomers to adsorb onto the particles.

Figure 3 shows the FT-IR spectrum of the solvent-based ink used in this study. A typical composition of screen-printing ink for HDPE bottles usually comprises 5% organic pigment, 11% titanium dioxide pigment, 82% epoxy resin, and 2% naphenate catalyst [9]. Since the majority of the ink formulation contains epoxy resin as the binder, it is expected that chemical fingerprints obtained in the FT-IR spectrum should belong to those of the epoxy resin. Normally, the epoxide group exhibits a weak C-H stretching peak around 3050 to 2990 cm^{-1} , while aromatic rings show absorption peaks over the wavenumber range of about 870 to 670 cm^{-1} [10]. According to Figure 3, the absorption peak at about 3026 cm^{-1} and the absorption peaks at about 870 to 670 cm^{-1} were evident. Under the strongly basic condition of the deinking process, the cationic head groups of the CnTAB monomers can associate with the negative moieties of the epoxy molecules (due to the opening of the epoxide rings) via electrostatic interactions, thus enhancing the adsorption of CnTAB monomers on the ink surfaces [8], although the epoxy molecules are simultaneously being solubilized into micelles as will later be discussed, making the deinking process quite complex.

The adsorption of CnTAB monomers on the ink pigment surfaces is driven by both electrostatic and van der Waal forces. Since the possibility for the formation of negatively-charged moieties of the epoxy binder and on the pigment (e.g., TiO_2) surfaces themselves is greater at a higher pH, increasing the pH value also renders greater adsorption of CnTAB monomers onto the ink pigment surfaces. On the other hand, pH does not affect the van der Waal interactions between the tail groups of the adsorbed CnTAB monomers and the hydrophobic part of the pigment

surfaces greatly. In addition, the van der Waal interactions should increase with the length of the hydrophobic part of the CnTAB molecules. We cannot, however, differentiate between the adsorption of CTAB onto the pigment particles themselves or onto epoxy molecules attached to the pigment surface.

5.4.2 Critical Micelle Concentration

5.4.2.1 Effect of Temperature

Plots of the CMC values of CnTAB solutions over a temperature range of 25 to 50°C are illustrated in Figure 4. The CMC of these CnTAB solutions increased very slightly with increasing temperature. Specifically, the CMC values increased from about 0.93 mM at 25°C to about 1.28 mM at 50°C for CTAB, from about 3.67 mM at 25°C to about 4.06 mM at 50°C for TTAB, and from about 16.1 mM at 25°C to about 17.8 mM at 50°C for DTAB. This mild effect of temperature on CMC of ionic surfactants is commonly observed [11] and necessary to know in this work to interpret the temperature dependence of deinking.

5.4.2.2 Effect of Salinity

The CMC of CnTAB with and without 0.1 M NaCl are shown in Table 1. There is good agreement between the two methods of measurement except in one case where we will use the conductivity values in the absence of added NaCl since the change in the slope of the curve used to obtain the CMC is sharper in that case. The CMCs of CnTAB without NaCl addition were 0.96, 3.75 and 16.29 mM, for CTAB, TTAB and DTAB, respectively, whereas, in CnTAB solution with 0.1 M NaCl, the CMCs were reduced to 0.04, 0.74 and 12.77 mM for CTAB, TTAB and DTAB, respectively. The reduction in the CMC with increasing alkyl chain length and the added electrolyte is well known for cationic surfactant and the values shown here are quite in line with a similar system [11].

5.4.3 Solubilization

5.4.3.1 Effect of Temperature

The effects of temperature and concentration on solubilization of epoxy binder from the ink in CnTAB solutions are shown in Figure 5. The concentrations of the CTAB and TTAB solutions for solubilization studies

were in the same ranges as those in the deinking experiments; i.e., 0.96 to 4.8 mM and 15.0 to 30.0 mM, respectively. For DTAB solutions however, the concentration range for the solubilization studies was very low (i.e., about 24.4 to 57.0 mM or about 1.5 to 3.5×CMC) as compared to about 293 to 382 mM or about 18.0 to 23.5×CMC used in the deinking experiments because high concentration of surfactant can solubilize essentially all epoxy molecules, so there is no effect of surfactant concentration at high concentrations.

In our previous work [8], solubilization studies were carried out at pH 12 and 30°C. We found that the solubilization of ink binder in CnTAB micelles increased with an increase in both the alkyl chain length and the concentration of CnTAB. In this work, the temperature was varied between 30 and 45°C. Apparently, for any given CnTAB concentration, the solubilization of ink binder within the micelles decreases with increasing temperature. Interestingly, the effect of temperature on the decrease in the solubilization of ink binder in CTAB micelles was more pronounced at higher concentrations. The results could be explained based on the effect of temperature on the CMC values and the aggregation numbers of CnTAB. As mentioned earlier, a slight increase in the CMC values with increasing temperature is evident (see Figure 5) and this leads to a smaller fraction of surfactant being in micelle form, however, this factor will have a small effect on solubilization capacity at the surfactant concentrations used here. An increase in the temperature causes the aggregation number to decrease [12,13]. The decreased aggregation number translates into less micellar volume that can accommodate solubilized ink binder [14].

Among the three surfactants investigated, the effect of temperature on solubilization of ink binder in TTAB solutions is more pronounced than for DTAB or CTAB. It is possible that the effect of the particular alkyl chain length of TTAB monomers affects the size of micelles more than CTAB and DTAB. It was reported that the size of TTAB micelles decreases with increasing temperature [13]. Specifically, the aggregation number of TTAB micelles decreases from about 157 at 5°C to about 72 at 81°C [14], while the CMC increases from about 3.7 mM at 20°C to about 7.1 mM at 80°C [15]. The decrease in the size of TTAB micelles with

increasing temperature is relatively more important than those of CTAB and DTAB, as evidenced by greater temperature dependence of solubilization in TTAB.

5.4.3.2 Effect of Salinity

The effect of NaCl addition on the solubilization of ink in CnTAB solutions is illustrated in Figure 6. The experiments were carried out at pH 12 and 30°C. For all surfactants, the addition of NaCl significantly improved the solubilization of ink binder. The difference in the solubilization of ink binder in CnTAB in comparison with that with NaCl salt addition was found to be more pronounced at higher CnTAB concentration, except for DTAB. The addition of NaCl contributes two main effects regarding to the solubilization capacity of ink binder. First, it lowers the CMC, which results in the improvement in the solubilization capacity of ink binder at low surfactant concentrations. Second, the aggregation number is increased when NaCl is added.

5.4.4 Deinking

5.4.4.1 Effect of pH

Figure 7 shows the ink removal as a function of pH for different values of normalized CnTAB concentration (i.e., CnTAB concentration divided by corresponding CMC value). These measurements were carried out at a fixed temperature of 30°C. Evidently, deinking is not possible in surfactant-free basic solutions. For a given type and concentration of CnTAB solution, the amount of ink removal increased monotonically with increasing pH of the solutions. For any given CnTAB concentration, the lowest pH value at which deinking began is about <11.0, <11.5, and 11.5 for CTAB, TTAB, and DTAB solutions, respectively. Significant deinking (i.e., at least 40% of ink removal) was observed at about 2×CMC and pH of about 11.5 for CTAB, 6×CMC and pH of about 11.8 for TTAB, and 16.5×CMC and pH of about 11.7 for DTAB, respectively. Evidently, for a given pH, the amount of ink removal increased monotonically with increasing alkyl chain length of CnTAB. Figure 7 also shows that complete deinking was observed at concentrations of about 4×CMC, 8×CMC, and 24×CMC for CTAB, TTAB, and

DTAB solutions, respectively [8]. Clearly, the effect of pH on deinking is less pronounced with CnTAB having a longer alkyl chain length. As previously mentioned, longer alkyl chain length helps promote the adsorption of CnTAB monomers onto the ink pigment surfaces (as evidenced from the observed increase in the zeta potential of ink particles with increasing alkyl chain length of CnTAB; see Figure 2) and adsorption of surfactant on both printed and unprinted HDPE surfaces is the first step in the deinking process of solvent-based ink from HDPE surfaces [8].

5.4.4.2 Effect of Temperature

Figure 8 shows the ink removal as a function of temperature for different values of normalized CnTAB concentration. These measurements were carried at a fixed pH of 12. Deinking is not significant in surfactant-free basic solutions at any temperature. The amount of ink removal decreases monotonically with increasing temperature for all surfactants and concentrations. For CTAB, significant deinking (i.e., at least 40% of ink removal) was observed at concentrations greater than about $1\times\text{CMC}$ when the temperature is 30°C . At 35°C , significant deinking is only observed at $5\times\text{CMC}$. For TTAB, significant deinking is only observed at 30°C , provided that the concentrations are greater than about $5\times\text{CMC}$. For DTAB, at 30°C , significant deinking is observed at all concentration ranges investigated. However, at 35°C , significant deinking is only observed at a concentration of $22\times\text{CMC}$ or greater.

The observed decrease in the deinking performance with increasing temperature is in accord with the observed decrease in the solubilization capacity of ink in CnTAB solutions with increasing temperature for all surfactants and concentrations. According to the four-step mechanism for the removal of solvent-based ink from the HDPE surfaces proposed in our previous work [8], solubilization of the ink binder in CnTAB micelles was postulated to be the most important step for deinking, a hypothesis which is supported by this subsequent work.

5.4.4.3 Effect of Salinity

Figures 9 to 11 show the removal of ink from HDPE surfaces in CTAB, TTAB, and DTAB solutions as function of concentration and normalized

concentration with or without the addition of 0.1 M NaCl. These experiments were carried out at a fixed pH of 12 and a fixed temperature of 30°C. Without the presence of the salt, complete deinking of printed HDPE surfaces was observed at about 2.9, 30, and 380 mM for CTAB, TTAB, and DTAB, respectively. With the salt added, it was observed at about 1.5, 8.2, and 305 mM, respectively. The result indicated again that deinking efficiency increased with increasing alkyl chain length and salinity. Marked improvement in the deinking efficiency was obtained in the TTAB solutions when the salt was added into the solutions, while, in the cases of CTAB and DTAB, only marginal improvement was observed.

The significant improvement in the deinking performance of TTAB with salt addition over those of CTAB and DTAB can be explained based on the results from the solubilization studies (see Figure 6), where the presence of NaCl increased the solubilization capacity of ink binder in the TTAB solutions in a much greater extent than it did with the CTAB and DTAB counterparts. For examples, at the concentration where CTAB solutions with added salt showed complete deinking (i.e., 1.5 mM), the solubilization capacity for that particular concentration was not very different from that when no salt was present (see Figure 6a). On the other hand, for TTAB, at the concentration where complete deinking was observed in the presence of salt (i.e., 8.2 mM), the solubilization capacity for that particular concentration was far greater than that when no salt was added (see Figure 6b). Based on these observations, the observed significant improvement in the deinking efficiency of TTAB solutions in comparison with the other two surfactants is reasonable.

If we are to consider the deinking performance as a function of normalized concentration with or without the presence of 0.1 M NaCl, it is obvious that, without the presence of salt, complete deinking was observed at a concentration equivalent to about 3xCMC, 8xCMC, and 23.5xCMC for CTAB, TTAB, and DTAB, respectively. On the other hand, with the presence of salt, complete deinking was observed at about 40xCMC, 11xCMC, and 23.5xCMC for CTAB, TTAB and DTAB, respectively. Apparently, with or without the presence of salt, complete deinking was only observed at concentrations greater than the CMC for all surfactant solutions studied.

5.6 Conclusions

The present contribution investigates the effects of pH, temperature, and salinity on the removal of solvent-based ink from high-density polyethylene (HDPE) surfaces based on the use of three alkyltrimethylammonium bromides (i.e., dodecyl-, tetradecyl-, and hexadecyl-trimethylammonium bromide or DTAB, TTAB, and CTAB, respectively). The critical micelle concentration (CMC) values of these surfactant solutions was found to increase very slightly with increasing temperature and decreased significantly with the addition of 0.1 M NaCl. Ink removal was found to increase with increasing concentration, increasing pH, decreasing temperature, and increasing salinity of the CnTAB solutions. The zeta potential of ink became more positively charged with increasing pH as well as concentration and alkyl chain length of CnTAB, suggesting that the adsorption of CnTAB molecules on ink pigment occurred more readily with an increase in any of those parameters. The solubilization of epoxy ink binder was found to increase with increasing surfactant concentration, decreasing temperature, and increasing salinity of the CnTAB solutions. Without the NaCl salt addition, complete deinking was observed at a concentration equivalent to about 3xCMC, 8xCMC, and 23.5xCMC for CTAB, TTAB, and DTAB, respectively, while, with the salt addition, it was observed at about 40xCMC, 11xCMC, and 23.5xCMC for CTAB, TTAB and DTAB, respectively.

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5.7 References

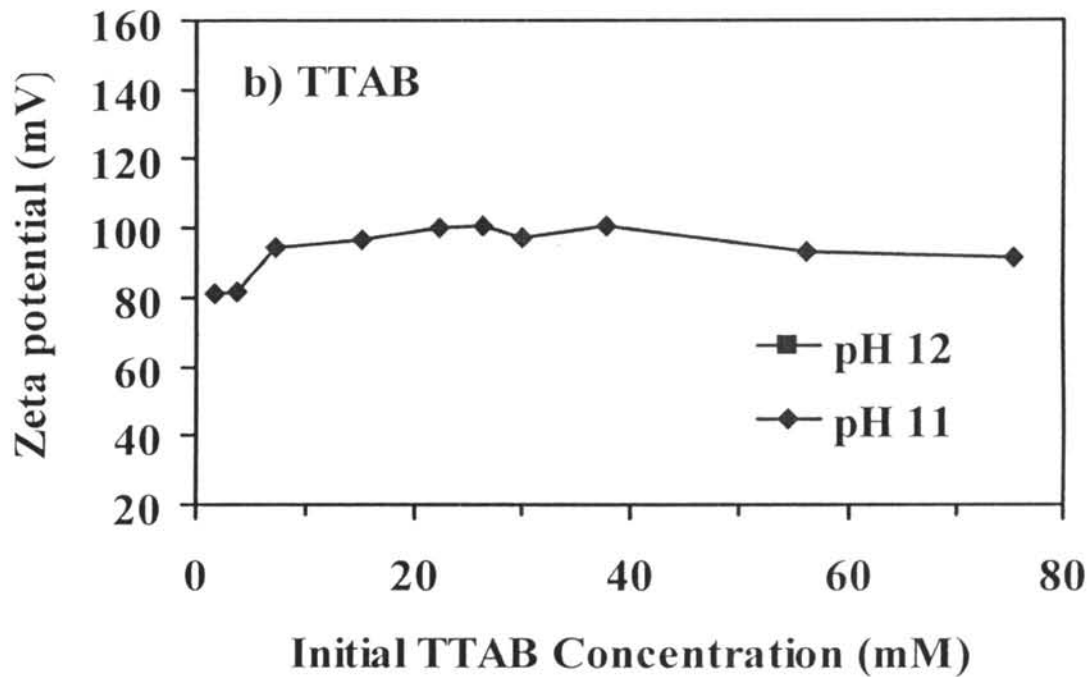
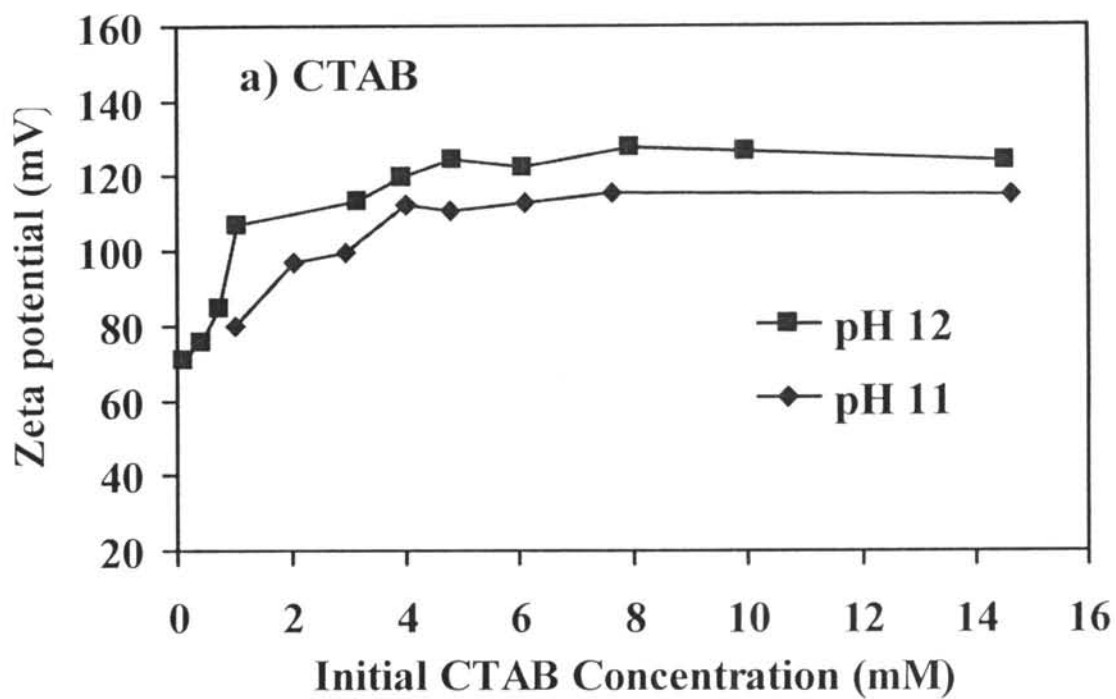
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Table 5.1 CMC of CnTAB solutions with or without the presence of 0.1 M NaCl at 30°C

CnTAB	CMC of CnTAB (mM)		
	without NaCl (conductivity)	without NaCl (surface tension)	with NaCl (surface tension)
CTAB	0.96	0.93	0.04
TTAB	3.75	3.70	0.74
DTAB	16.29	13.77	12.77



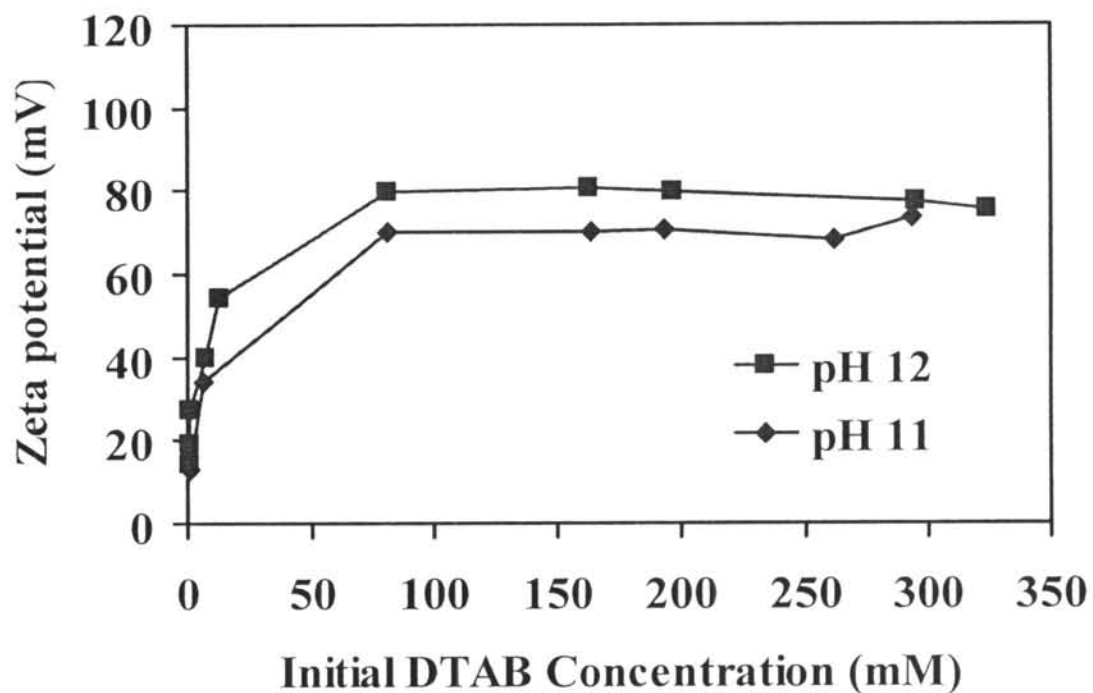


Figure 5.1 Zeta potential of ink in (a) CTAB, (b) TTAB, and (c) DTAB solutions as a function of concentration (in a linear scale) at pH levels of 11 and 12.

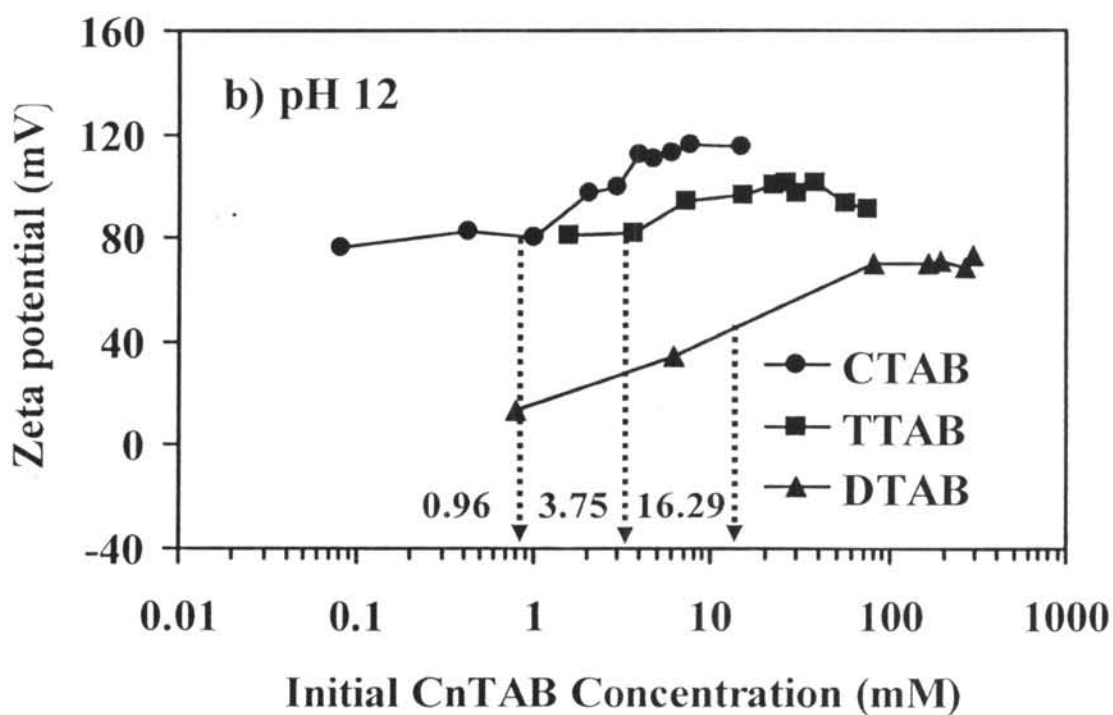
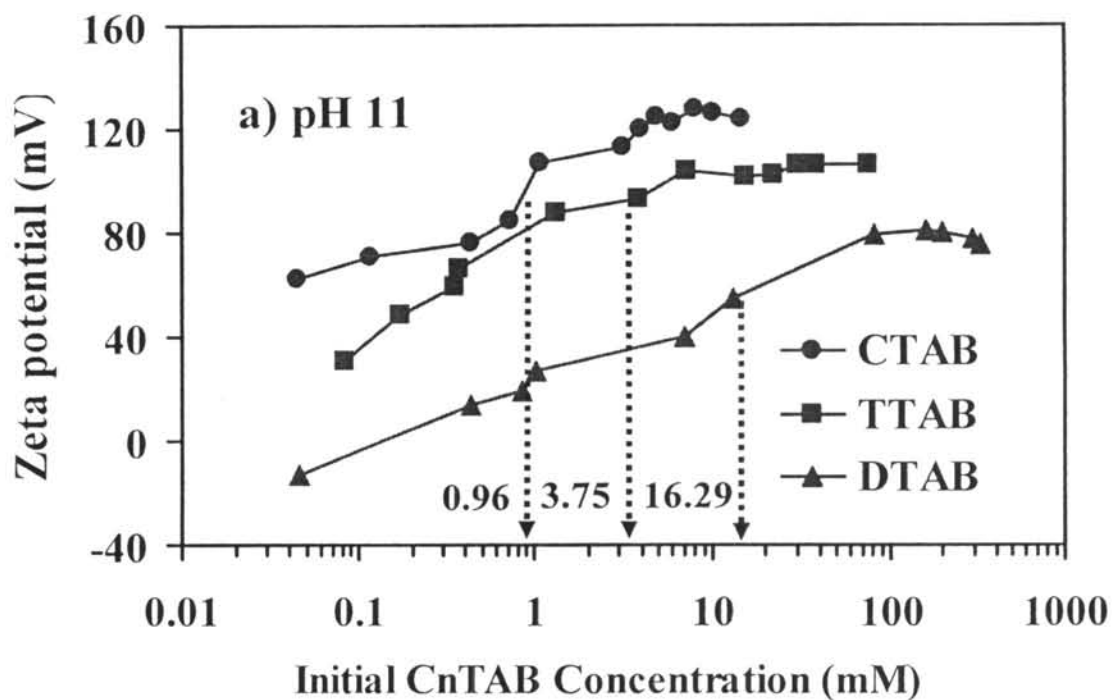


Figure 5.2 Zeta potential of ink in CnTAB solutions as a function of concentration (in a semi-logarithmic scale) at pH levels of (a) 11 and (b) 12. Dashed lines refer to CMC values.

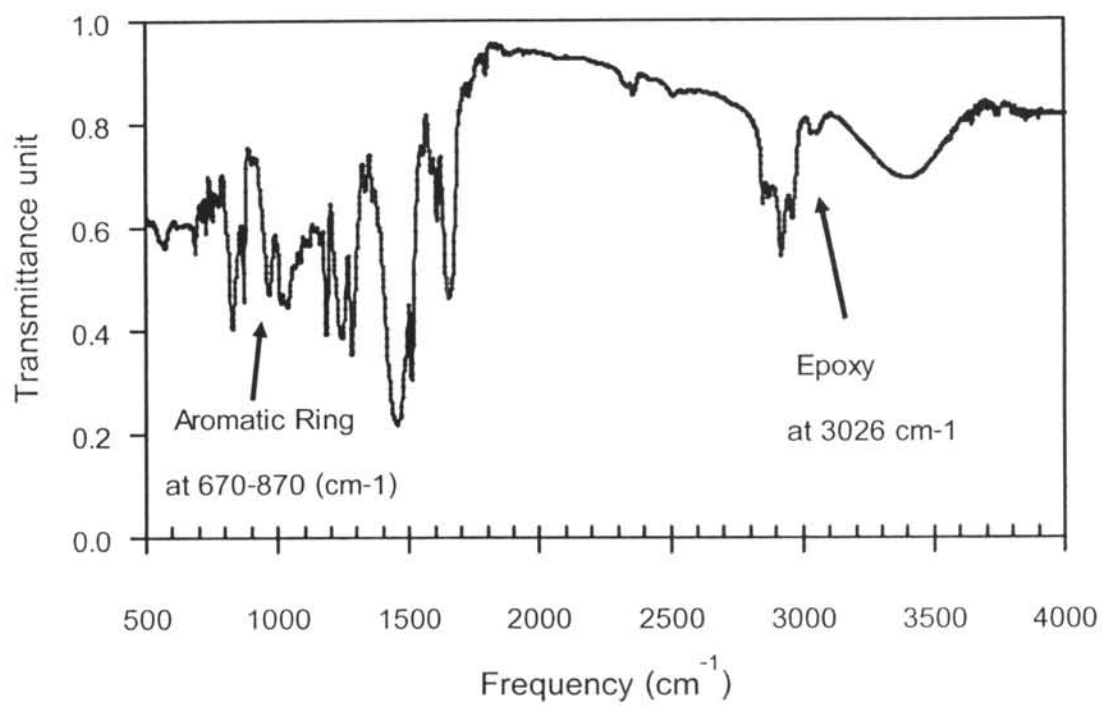


Figure 5.3 FT-IR spectrum of ink powder.

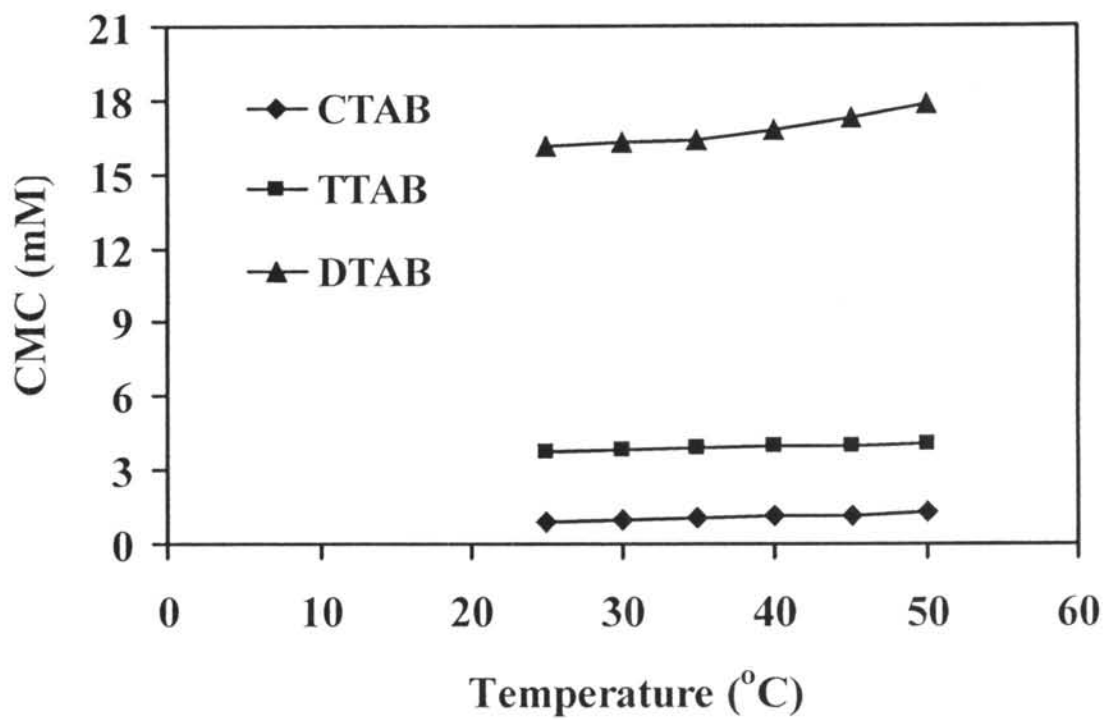
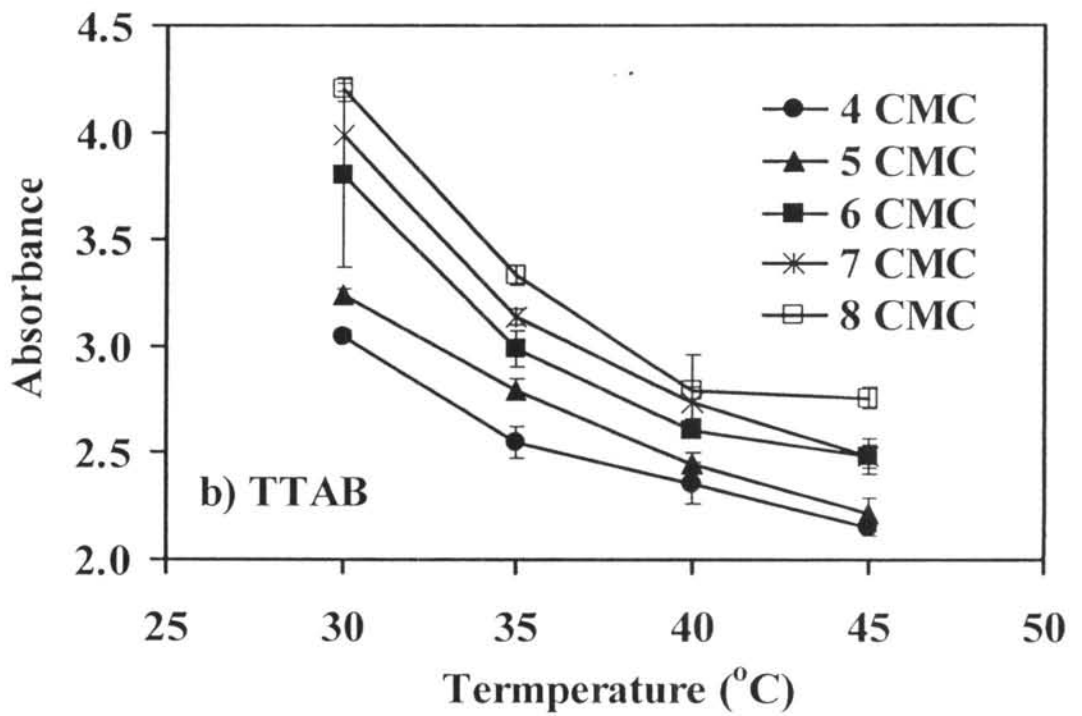
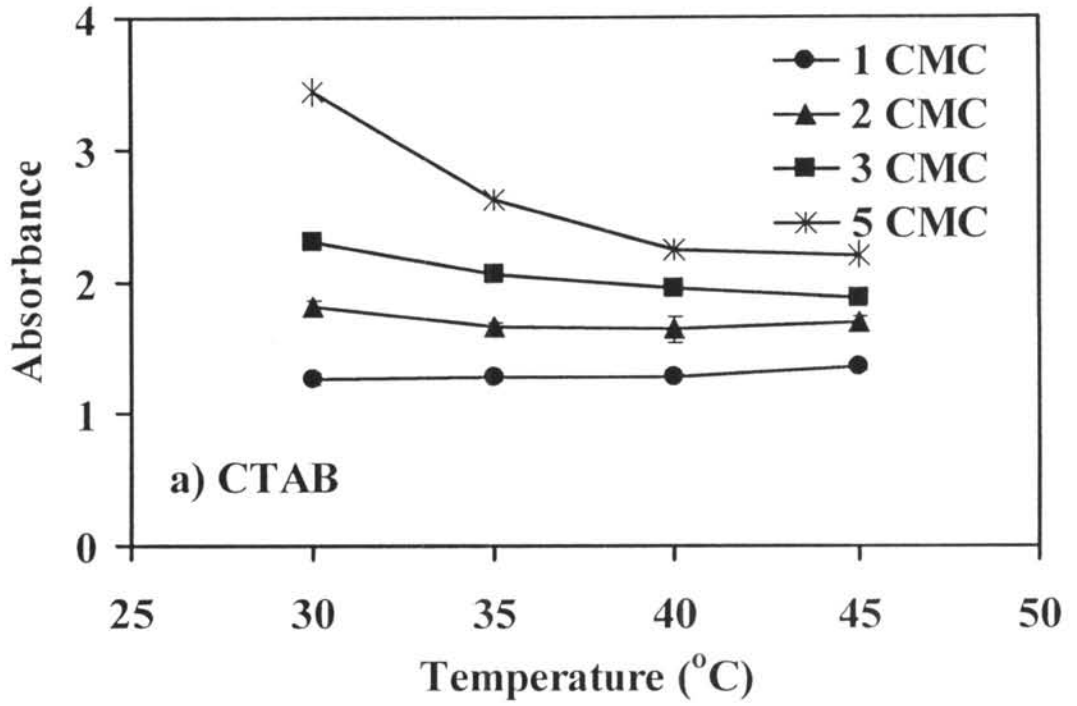


Figure 5.4 CMC of CnTAB solutions as a function of temperature.



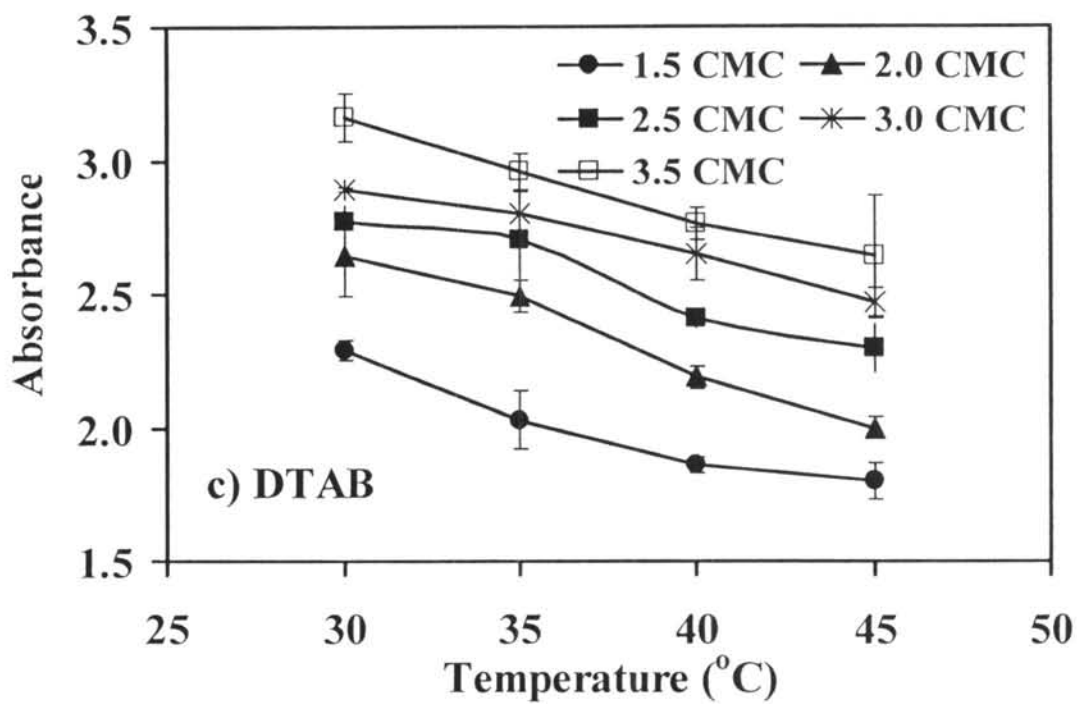
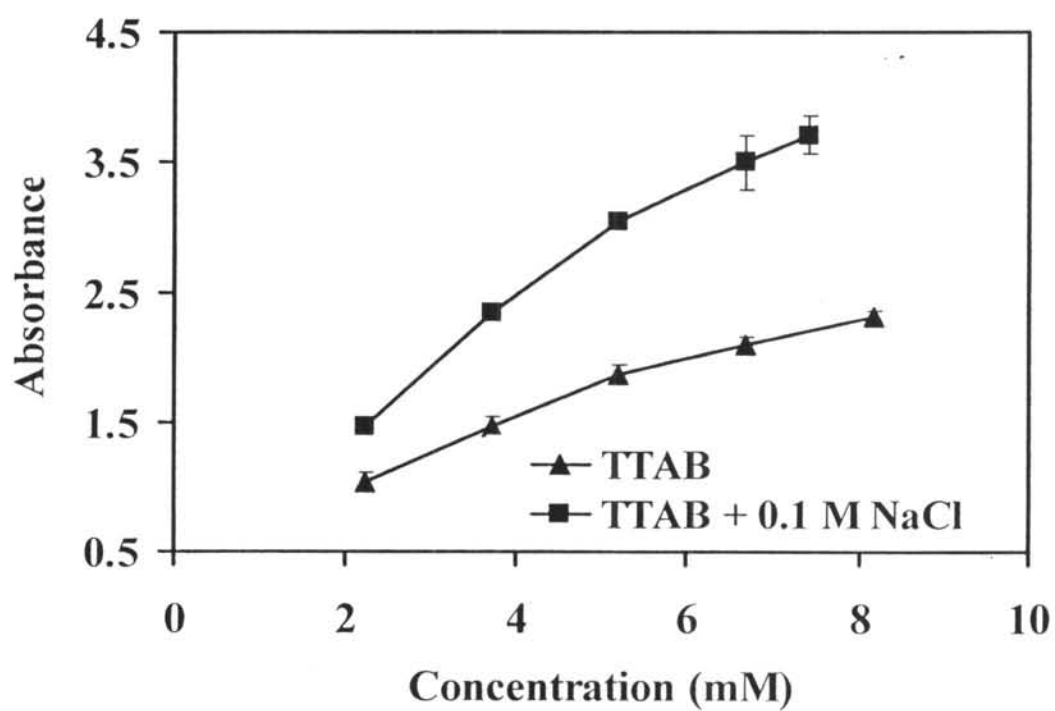
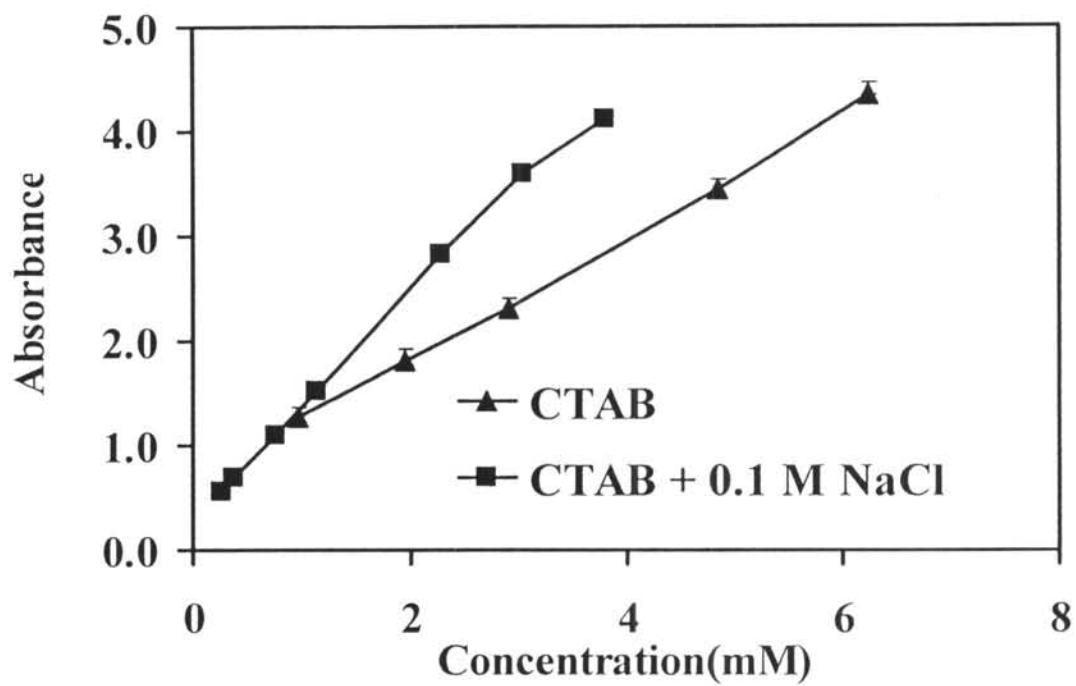


Figure 5.5 Solubilization capacity of ink components in (a) CTAB, (b) TTAB, and (c) DTAB solutions at various normalized concentrations (i.e., concentration divided by CMC of the corresponding CnTAB solution) at pH 12 as a function of temperature.



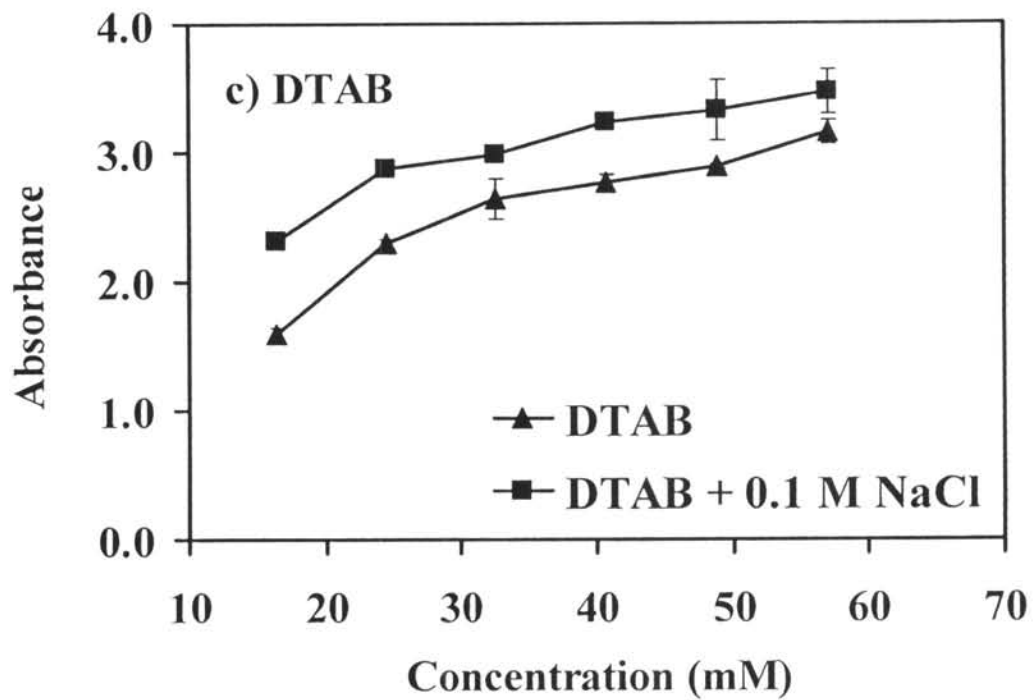
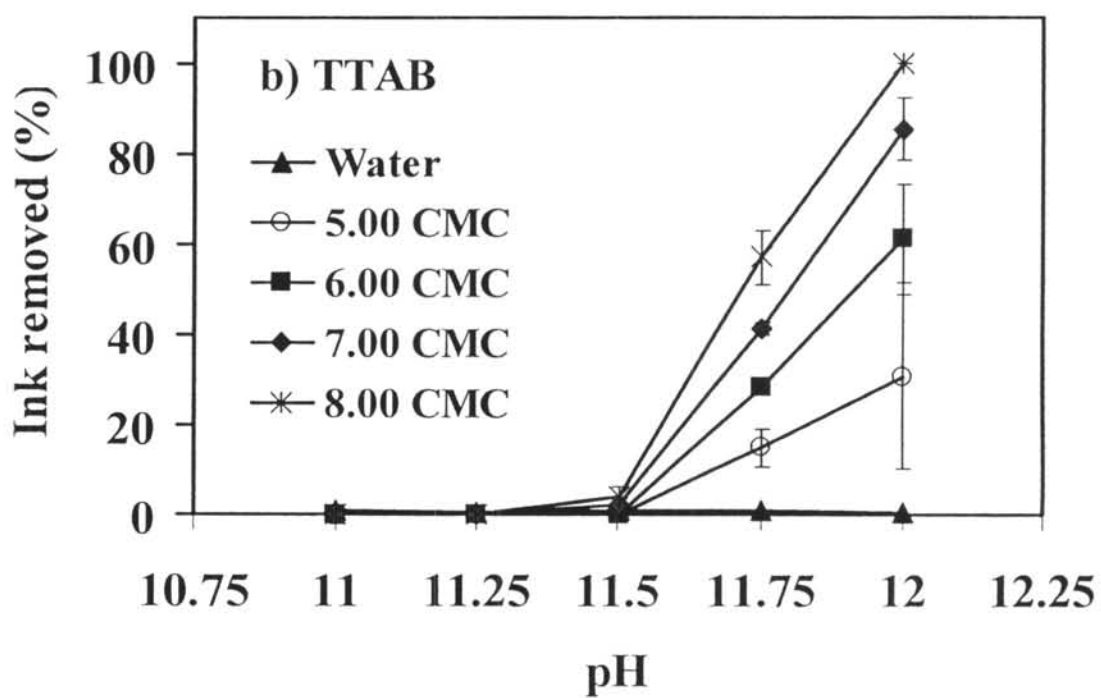
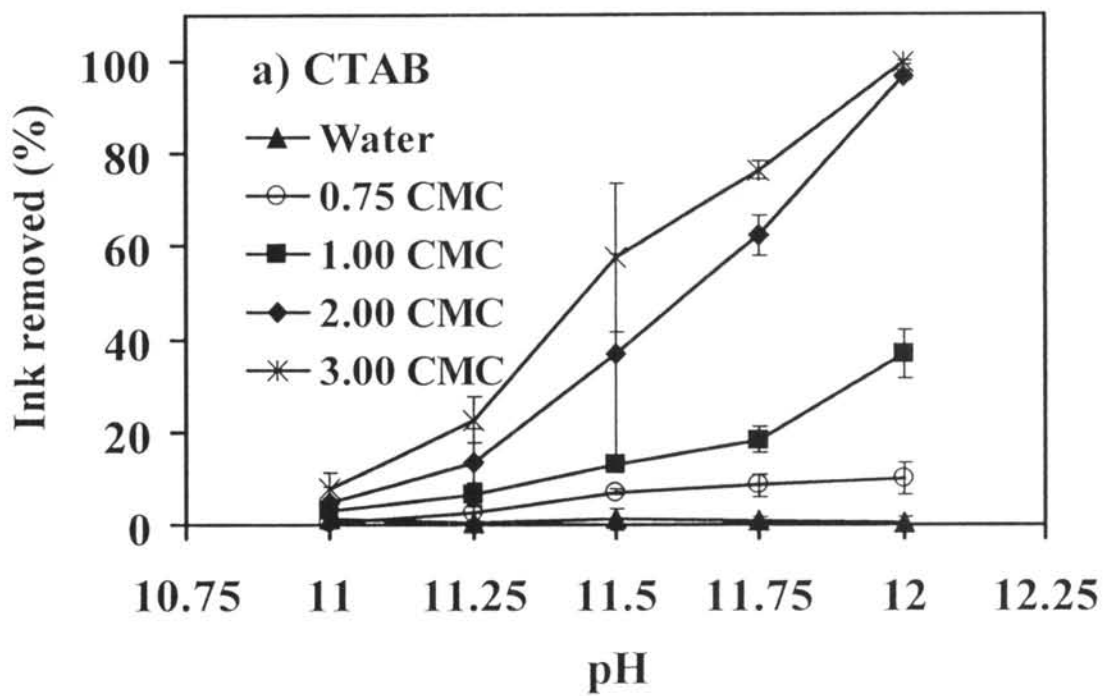


Figure 5.6 Solubilization capacity of ink components in (a) CTAB, (b) TTAB, and (c) DTAB solutions with or without the presence of 0.1 M NaCl at pH 12 and 30°C as a function of concentration.



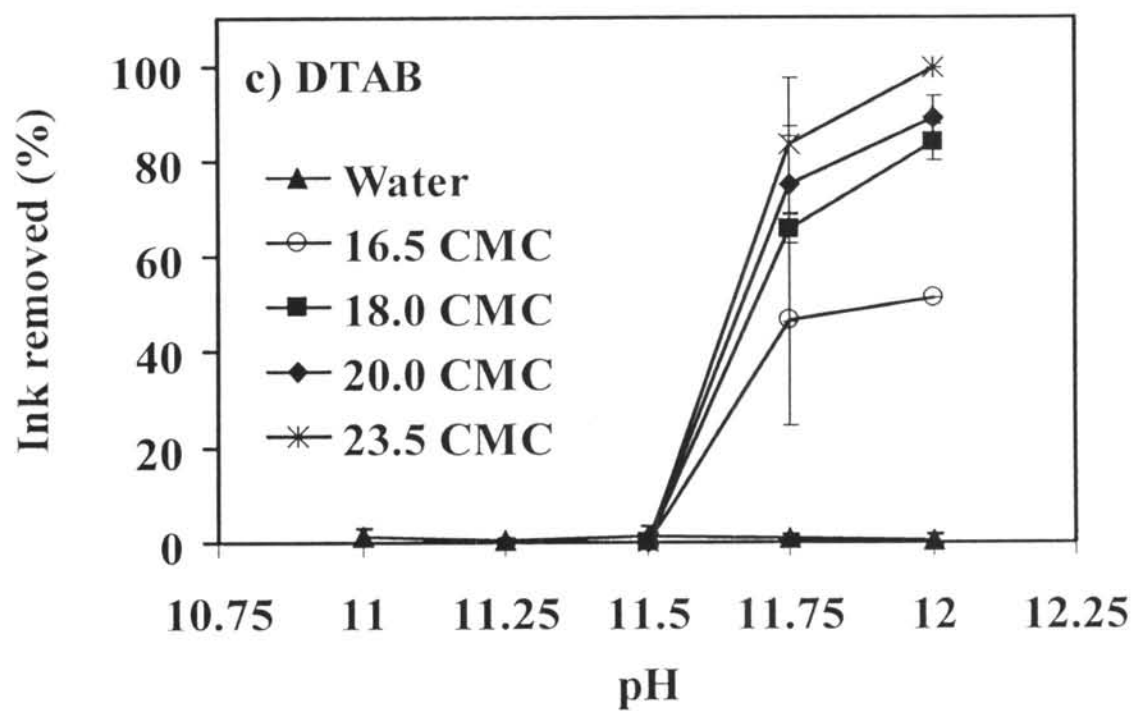
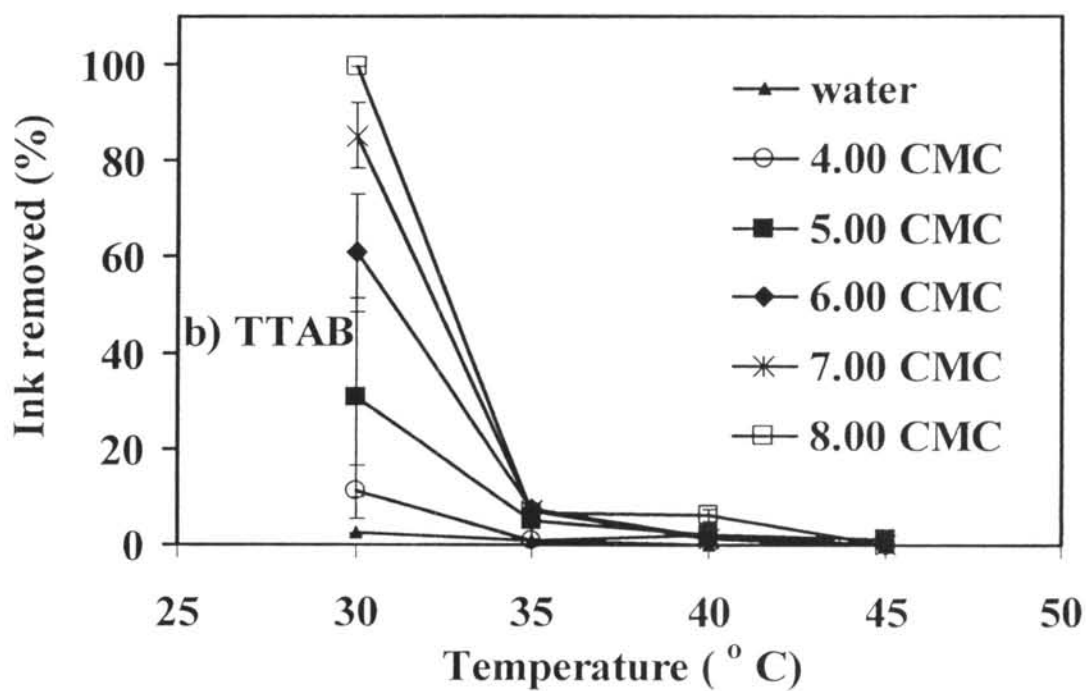
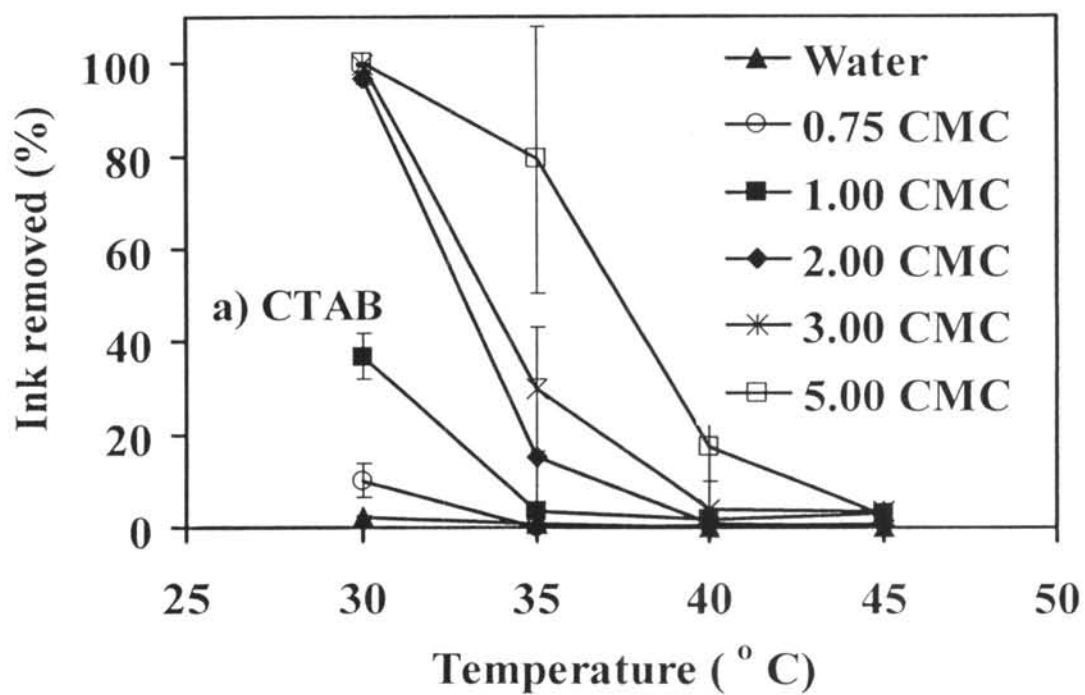


Figure 5.7 Percentage of ink removal from printed HDPE surfaces in (a) CTAB, (b) TTAB, and (c) DTAB solutions at various normalized concentrations at 30°C as a function of pH.



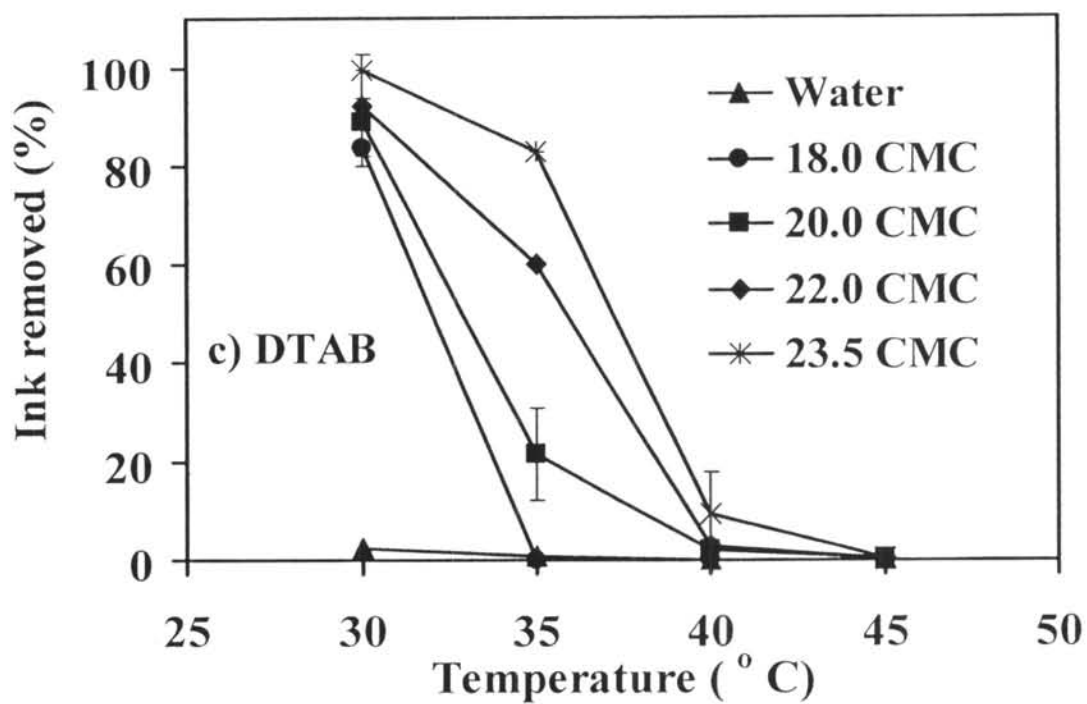


Figure 5.8 Percentage of ink removal from printed HDPE surfaces in (a) CTAB, (b) TTAB, and (c) DTAB solutions at various normalized concentrations at pH 12 as a function of temperature.

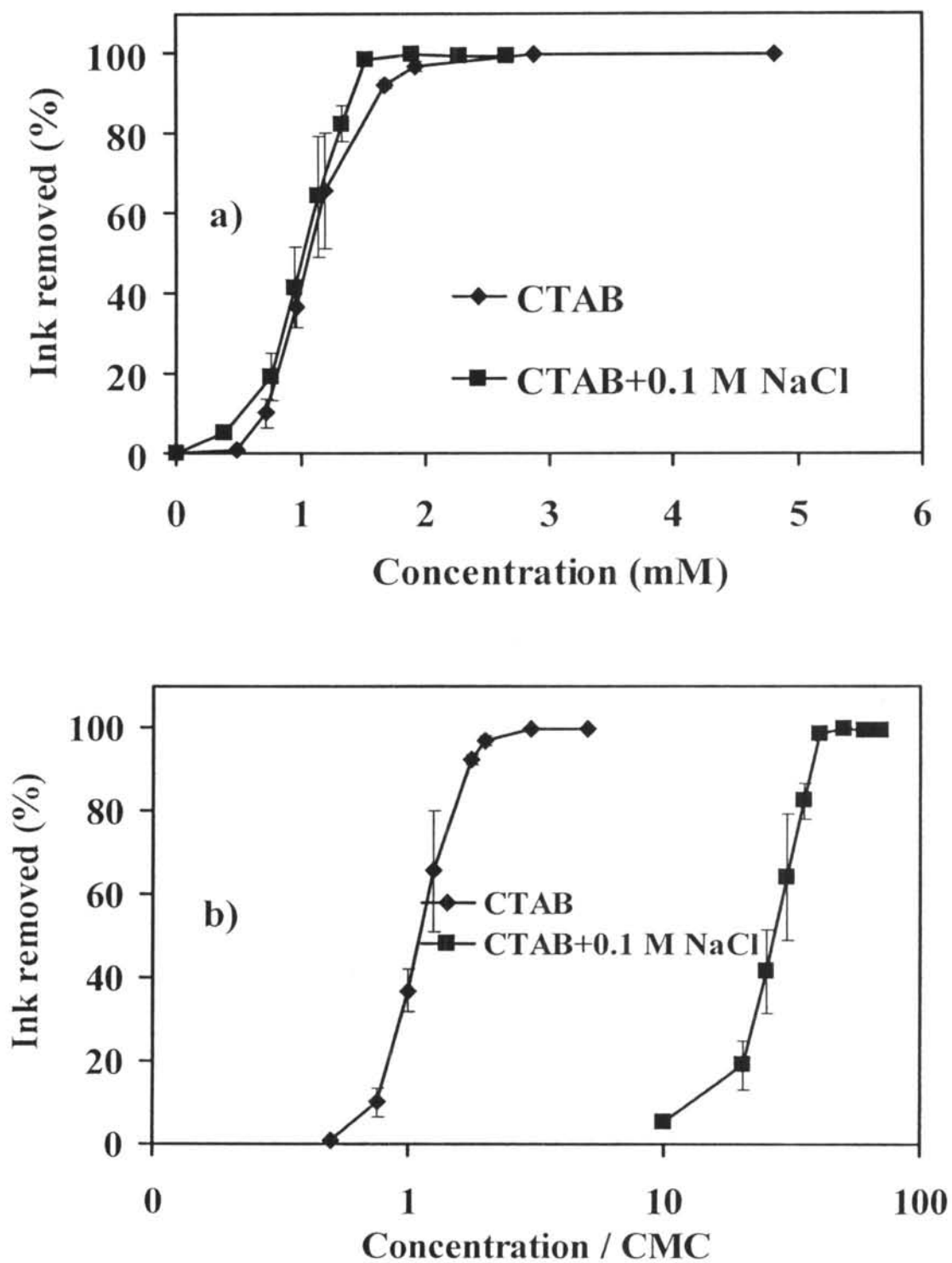


Figure 5.9 Percentage of ink removal from printed HDPE surfaces in CTAB solution with or without the presence of 0.1 M NaCl at pH 12 and 30°C as a function of concentration: (a) on a normal scale and (b) on a normalized scale (i.e., concentration/CMC).

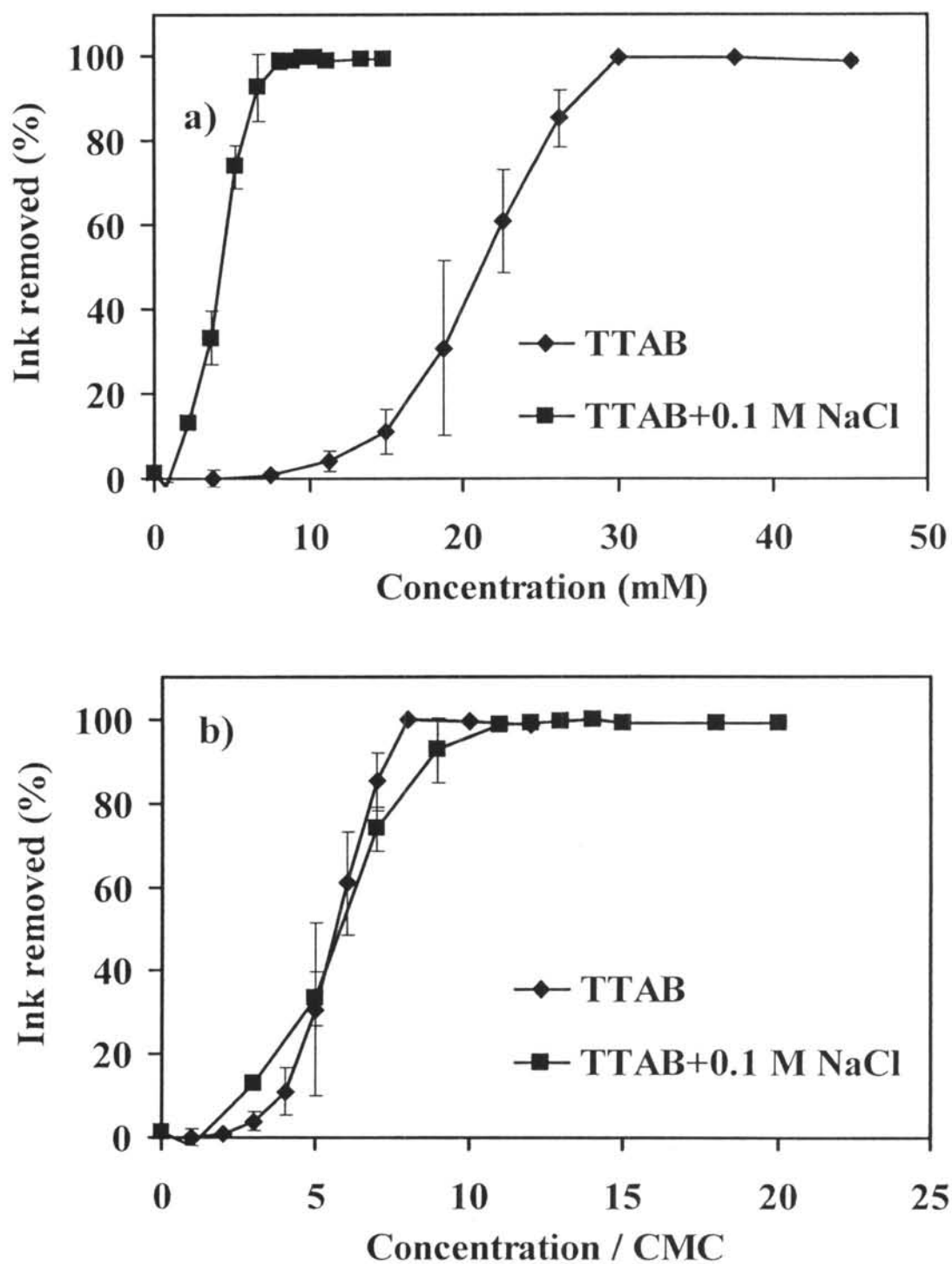


Figure 5.10 Percentage of ink removal from printed HDPE surfaces in TTAB solution with or without the presence of 0.1 M NaCl at pH 12 and 30°C as a function of concentration: (a) on a normal scale and (b) on a normalized scale (i.e., concentration/CMC).

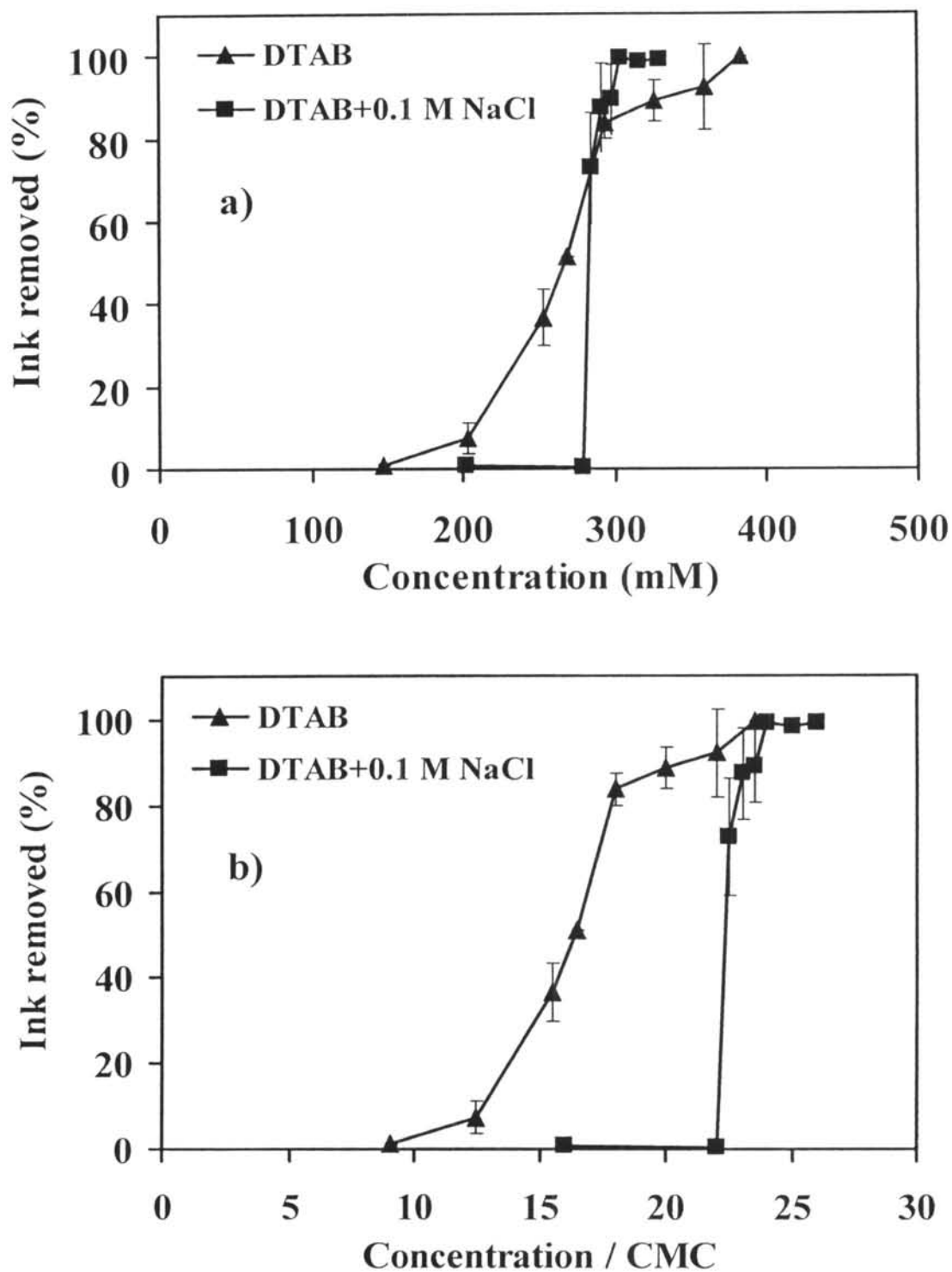


Figure 5.11 Percentage of ink removal from printed HDPE surfaces in DTAB solution with or without the presence of 0.1 M NaCl at pH 12 and 30°C as a function of concentration: (a) on a normal scale and (b) on a normalized scale (i.e., concentration/CMC).