

Solution behavior of anionic polymer sodium carboxymethylcellulose (NaCMC) in presence of cationic gemini/conventional surfactants

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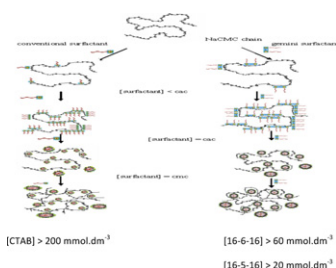
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HIGHLIGHTS

- The cmc values for geminis are 10–14 times smaller than of CTAB.
- The order of interaction of the surfactants $16-5-16 > 16-6-16 > \text{CTAB}$.
- The N_{agg} in presence of NaCMC is the same as those for the corresponding free micelles (conventional as well as geminis).
- Increase in relative viscosity is more for gemini surfactant with shorter spacer.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 9 June 2012

Received in revised form 27 August 2012

Accepted 19 September 2012

Available online 25 September 2012

Keywords:

Polymer
Surfactant
Conductivity
Fluorescence
Viscosity

ABSTRACT

The interaction of sodium carboxymethylcellulose (NaCMC) and cationic gemini surfactants (16-s-16, s = 5, 6)/conventional surfactant (CTAB) in aqueous solutions is investigated by conductivity, fluorescence and viscosity techniques. Electrostatic and hydrophobic interactions play a dominant role in such systems. The conductivity results showed that the geminis interact strongly with NaCMC as compared to CTAB. Fluorescence measurements were used to calculate aggregation number for the three combinations which were found about the same as those for the corresponding free micelles. Addition of surfactants leads to an increase in relative viscosity after certain concentration of the surfactants. Further increase in relative viscosity is significant in case of geminis and this increase is ascribed to the physical cross-linking of surfactant micelles with NaCMC chains.

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1. Introduction

Along with fundamental interest in intermolecular interactions and aggregation phenomena, as polymer/surfactant mixtures possess properties of varied nature (of course, due to availability of molecular structure variations in both the constituents), the interest in investigating such systems is continuing since long. It is pertinent to mention here that polymer/surfactant mixtures have found, as a result of significant research, use in a wide range of domestic, industrial and technological applications. A mixture

in which the polymer and surfactant bear opposite charges is of special interest because association in these systems is strong due to very strong force of electrical attractions. Alkyl chain aggregation of the bound/adsorbed surfactant molecules provides further reinforcement [1]. There is growing interest in associated polymer–surfactant systems based on environmentally friendly biodegradable natural polymers. Among ionic derivatives of cellulose, sodium carboxymethylcellulose (NaCMC), an anionic polymer as shown in Fig. 1 and is most widely used in paints, food, and cosmetic industry, pharmaceuticals, oilfield and paper industries due to its superior properties such as binding, thickening and stabilizing agent in these end uses. The application field of NaCMC can be extended further by chemical modifications such as by its interaction with oppositely charged surfactant micelles [2–9]. The mixture

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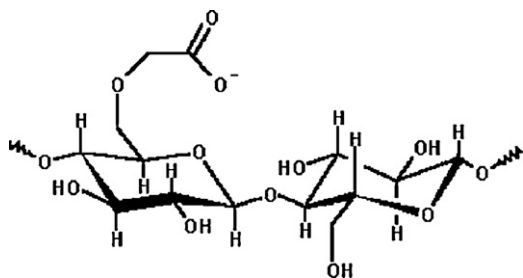


Fig. 1. Structure of NaCMC.

of the polyelectrolyte and oppositely charged surfactant would precipitate in case of the charge ratio of surfactant to polyelectrolyte approaches unity, due to the improvement of hydrophobicity, for more surfactant binding to polyelectrolyte. As excessive surfactant is added, the precipitate would be re-dissolved, because the adsorption of additional surfactants on the precipitates increases the hydrophilicity of the precipitates [8]. The structure, concentration, size and configuration of surfactant clusters, and ratio of the two components can have important effects on the properties.

In the last few years, a novel class of surfactants called gemini (dimeric), consisting of two hydrophobic chains and two polar/ionic head-groups united covalently by a spacer (rigid or flexible), has emerged [10–13]. These gemini surfactants impart better surface properties as compared to their single-head, single-tail counterparts. The geminis have much smaller cmc values, much greater efficiency in reducing surface tension than expected, better wetting properties, and other unusual behaviors. Due to their superior performance in applications and their tunable molecular geometry, geminis have been generating increasing interest among researchers.

Despite many studies on interactions between ionic polymers and surfactants, studies involving gemini surfactants are lacking. Therefore, in the present work, the solution behavior of NaCMC with gemini surfactants α,ω -bis(hexadecyldimethylammonium)alkane dibromides (16-s-16) and the corresponding monomeric counterpart cetyltrimethylammonium bromide (CTAB) has been assessed by using conductivity, fluorescence and viscosity measurements. Thus, the aim of this study is to gain insight into how the cationic gemini surfactant and their monomeric counterpart affects the structure of polymer/surfactant complex of the anionic polymer NaCMC at various surfactant concentrations. The mechanism of interactions between the polymer and the surfactants are proposed, and the changes at different concentrations of surfactants in the systems have been elucidated, which provide important information for the further applications of the above systems.

2. Materials and methods

2.1. Materials

NaCMC (mol. wt $\sim 90,000$, D.S. 0.70, Sigma–Aldrich, USA) and CTAB ($\geq 99.0\%$, Merck, Germany) were used as received. Gemini surfactants α,ω -bis(hexadecyldimethylammonium)alkane dibromides (16-6-16 and 16-5-16) were prepared and purified as described elsewhere [14], which gave ^1H NMR spectra and C,H,N analyses data consistent with their assigned structures [15]. For synthesis, the following materials were used without further purification: 1,5-dibromopentane (Himedia, India), 1,6-dibromohexane ($\geq 98\%$, Fluka, Switzerland), N,N-dimethylhexadecylamine ($\geq 95\%$, Fluka, Switzerland) and dry ethanol (99.9%, Changshu Yangyuan, China).

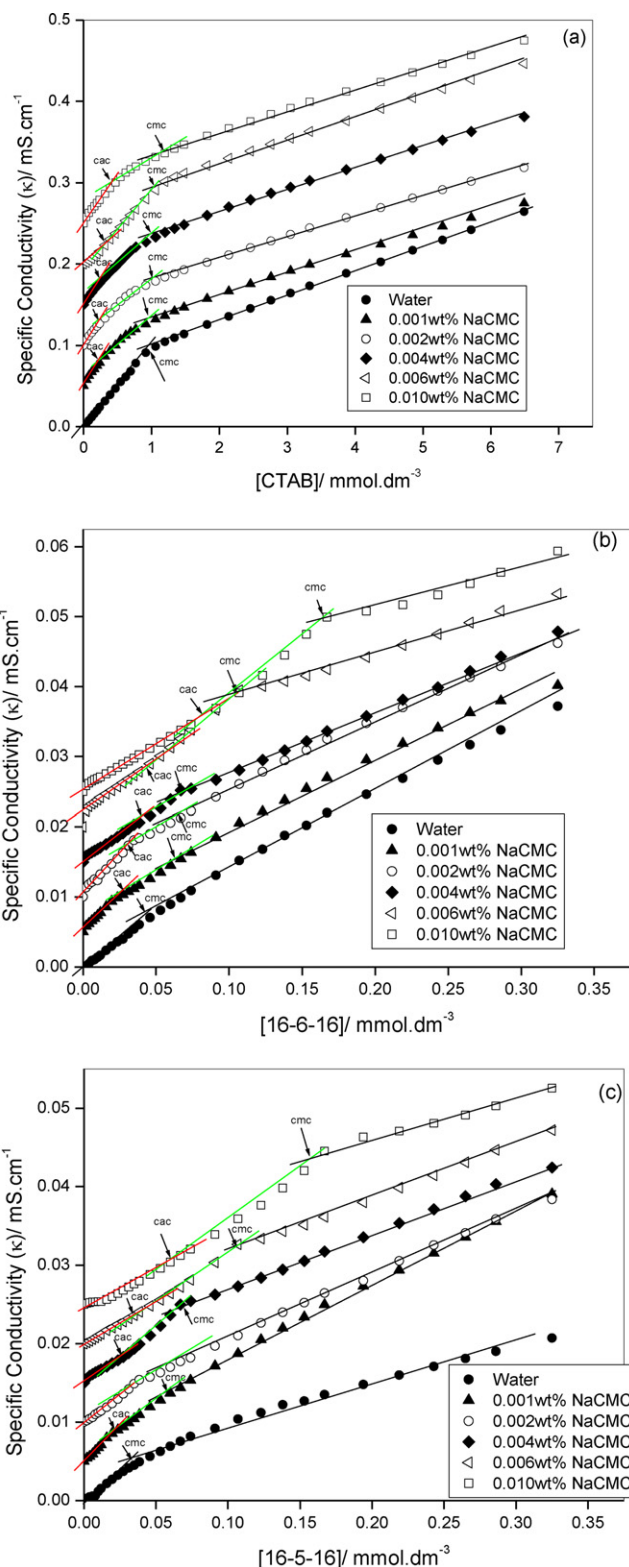


Fig. 2. Representative plots of specific conductance (κ) versus [surfactant] in water at different weight percentages of NaCMC solutions at 298.15 K. (a) CTAB, (b) 16-6-16, and (c) 16-5-16.

2.2. Conductivity measurements

For conductivity measurements, NaCMC reference solution was prepared by dissolving a known amount of NaCMC in demineralised double-distilled water of specific conductivity $1\text{--}2 \times 10^{-6} \text{ S cm}^{-1}$.

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