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Manipulation of lyotropic liquid crystal behavior of ionic liquid-type imidazolium surfactant by amino acids

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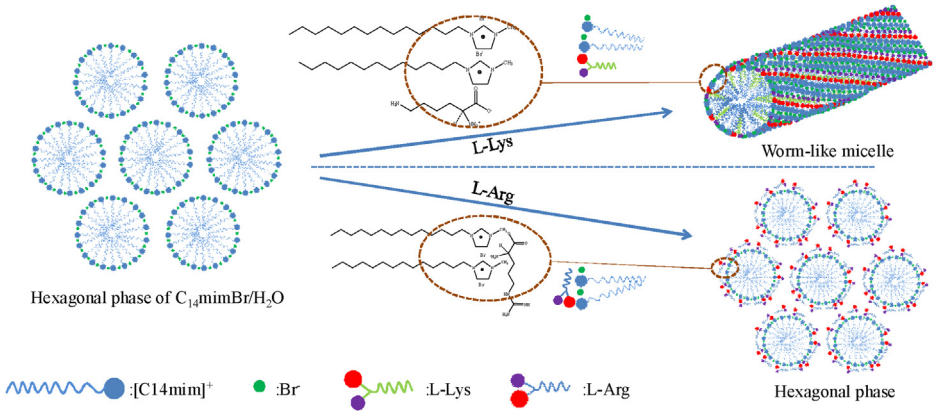
HIGHLIGHTS

- The effects of amino acids on the phase behavior of ionic liquid-type imidazolium surfactant were investigated.
- With the addition of Lys, C_{14}mimBr LLC transformed to worm-like micelles.
- For C_{14}mimBr/Arg system, it remained the hexagonal phases.
- Our work contributes to a better understanding of the effect of amino acids on the influence of surfactant aggregates.

GRAPHICAL ABSTRACT

Schematic illustrations of the phase transition between LLCs and the micelle solution by the introducing of amino acids.



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ABSTRACT

The effects of alkaline amino acids L-Lysine (L-Lys) and L-Arginine (L-Arg) on the lyotropic liquid crystal (LLC) behavior of ionic liquid-type imidazolium surfactant (1-tetradecyl-3-methylimidazolium bromide, C_{14}mimBr) were investigated systematically. The corresponding properties were investigated by polarized optical microscopy (POM), small angle X-ray scattering (SAXS), field emission-scanning electron microscopy (FE-SEM), Fourier transform infrared (FT-IR) spectroscopy and rheological measurements. The results indicated that with the introduction of L-Lys, LLC phase of C_{14}mimBr gradually undergoes the transition to an isotropic homogeneous phase, which demonstrated to be worm-like micelles (WLMs). While for C_{14}mimBr/L-Arg system, it remained the hexagonal phases and merely induced the variation

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of the mechanical strength of C_{14}mimBr LLCs. It can be speculated that the balance between electrostatic interaction, H-bond interaction, and hydrophobic interaction plays an important role in the phase transition of C_{14}mimBr/amino acids. Our work can contribute to a better understanding of the effect of the additions especially amino acids on the influence of the surfactant aggregates and their macroscopic properties, which maybe open the door for wide applications in the biological system.

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1. Introduction

Ionic liquids (ILs) have received a lot of attention due to their unique chemical and physical properties, such as nonvolatility, nonflammability, high stability, high ionic conductivity, and easy recyclability [1–3]. Among various kinds of ILs, surface-active ILs (SAILs), referred to the ILs containing long alkyl chains which exhibit amphiphilic character, have emerged as a novel kind of amphiphiles [4–6]. SAILs are capable to form supramolecular aggregates including micelles, vesicles, microemulsions, lyotropic liquid crystals (LLCs), and gels, which can be controlled and regulated by variation in the ratio of cationic and anionic surfactants, polar additives, temperature, salts, solvent, etc. [7–13]. The aggregation behavior of SAILs based on imidazolium, pyrrolidinium, pyridiniumcations, and morpholinium based cations has been extensively investigated in the field of colloid and interface chemistry [8,14,15]. Among them, the self-aggregation of imidazolium-type SAILs, particularly 1-alkyl-3-methylimidazolium salts, plays an important role in the processes such as the extraction of compounds from IL-containing systems, modified functional materials, modifying the various types of chemical reactions, controlling the phase behavior of new aqueous biphasic or multiphasic systems [16,17]. The results demonstrated that the surface activities of the long-chain imidazolium ILs are somewhat superior to that of conventional ionic surfactants when compared at the same hydrocarbon chain length [7,9,10].

The interactions between surfactant and protein have also been investigated which is not only fundamental in theoretics, but also practical in industrial applications [18–21]. It is well known that amino acids are the basic unit of proteins, which gives the specific molecular structure features of the proteins. Therefore, they are always considered to be the model compounds for specific aspects of the more complex proteins. Much attention has also been paid to study the interactions between surfactant and amino acids. For example, He et al. employed surface tension and oscillating bubble measurements to investigate the aggregation behaviors of sodium deoxycholate (NaDC) at the air/water surface in the absence and presence of three alkaline amino acids [22]. It is worth mentioning that both electrostatic and hydrophobic interactions are responsible for the interaction between NaDC and amino acids. Our group also investigated the effect of the amino acids on the properties of hydrogels formed by NaDC [23,24]. The results indicated that the addition of amino acids modifies the balance of the weak interactions for the formation of hydrogels and determines the phase structure transition. Geng et al. studied the interactions between a surface active imidazolium ionic liquid and BSA [25]. Compared with traditional cationic surfactants, C_{14}mimBr stabilizes the secondary structure of BSA at low concentration (below the CMC), but destroys the secondary structure of BSA at high concentration (above the CMC). Ruiz et al. investigated the effect of glycine on the surface activity and micellar properties of N-decanoyl-N-methylglucamide [26]. It was found that both the hydrodynamic radius and the micellar aggregation number increased with the glycine concentration.

Among various supramolecular aggregates mentioned above, LLCs, an important type of self-assembled aggregates, have attracted increasing interest for their potential applications in biomaterials and electro-optics. It was demonstrated that 1-alkyl-3-methylimidazolium bromides ($C_n$mimBr) could spontaneous self-organize and form liquid-crystalline gel in aqueous solutions [27–29]. Herein, in this article, the effects of L-Lysine (L-Lys) and L-Arginine (L-Arg) on the aggregation behavior of C_{14}mimBr were investigated, and then the driving forces that govern evolution of the phase behavior of C_{14}mimBr were speculated. Various techniques were used to study and verify the phase-transition processes in these systems, including polarized optical microscopy (POM), small angle X-ray scattering (SAXS), field emission-scanning electron microscopy (FE-SEM), Fourier transform infrared (FT-IR) spectroscopy and rheological measurements. Our results indicated that after the addition of L-Lys and excess L-Arg, the LLC behaviors of C_{14}mimBr were weakened, and L-Lys can even cause the system transforming from LLC phase to worm-like micelles (WLMs). Although varieties of addition responsive systems have been reported previously, it is very rare for observation of the formation of WLMs with imidazolium-type SAILs and amino acids. This research may open an avenue to gain a better knowledge about the behavior of such systems in the fields of drug delivery, oilfields, biochemistry, etc.

2. Experimental section

2.1. Material

Ionic liquid-type imidazolium surfactant (C_{14}mimBr) was purchased from Lanzhou institute of chemical physics with the purity greater than 99%. L-lysine (L-Lys) and L-arginine (L-Arg) were purchased from Sinopharm Chemical Reagent. Ultrapure water with a resistivity of 18.25 MΩ cm was obtained using a UPH-IV ultrapure water purifier (China). The structures of C_{14}mimBr, L-Lys and L-Arg are shown in Fig. 1.

2.2. Sample preparation

The desired amount of C_{14}mimBr was added to a certain concentration of amino acids solution. The mixtures were homogenized by stirring for about 10 min. Then the mixture was allowed to equilibrate at 20.0 ± 0.1 °C for two weeks before any tests.

2.3. Methods and characterization

For field-emission scanning electron microscopy (FE-SEM) observations, a drop of hydrogel was freeze-dried in vacuum at −55 °C. Then the freeze-dried powder was placed on conductive tapes to form a thin film. A layer of gold was sputtered on top to make the conducting surface. The sample was observed on a JSM-6700F FE-SEM.

The obtained sample phases were characterized by an HMBG-SAX X-ray small-angle scattering system (Austria) with Ni-filtered Cu Kα radiation (0.154 nm) operating at 50 kV and 40 mA. The distance between the sample and the detector was 27.8 cm. Fourier transform infrared (FTIR) spectra were recorded on a VERTEX-70/70v spectrometer (Bruker Optics, Germany). XRD patterns were obtained on a Rigaku D/Max 2200-PC diffractometer

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