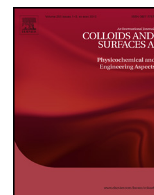
Colloids and Surfaces A: Physicochem. Eng. Aspects 471 (2015) 148–152

Contents lists available at ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: www.elsevier.com/locate/colsurfa



Lag-burst kinetics of surfactant displacement from the liquid crystal/aqueous interface by bile acids

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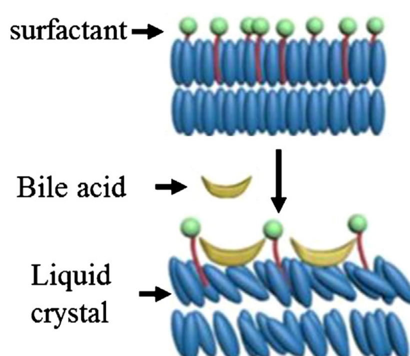
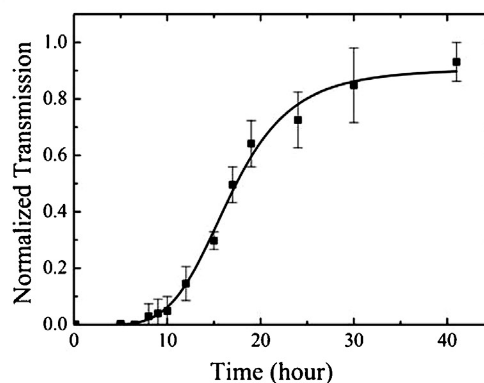
HIGHLIGHTS

- Sodium dodecyl sulfate can be displaced from the liquid crystal/aqueous interface by the competitive adsorption of bile acids.
- The displacement exhibits lag-burst kinetics.
- The lag time and burst rate depends on the number and position of the hydroxyl groups of bile acids.

GRAPHICAL ABSTRACT





ARTICLE INFO

Article history:
Received 29 October 2014
Received in revised form 30 January 2015
Accepted 4 February 2015
Available online 16 February 2015

Keywords:
Bile acids
Surfactants
Liquid crystals
Displacement
Kinetics

ABSTRACT

Bile acids play an important role in fat digestion by displacing surfactants from the oil–water interface through emulsification. In this paper, we study the time course of the displacement of sodium dodecyl sulfate (SDS) from the liquid crystal (LC)/aqueous interface by four unconjugated bile acids, which differ in the number and position of hydroxyl groups on their steroid backbones. The competitive adsorption of bile acids displaces the SDS from the LC/aqueous interface and consequently triggers a homeotropic-to-tilted anchoring transition of the LC at the interface, which allows the displacement kinetics to be monitored by a polarizing optical microscope. The microscopy image analysis reveals that the displacement exhibits lag-burst kinetics, where a lag phase is followed by a burst phase. We find that the number and position of the hydroxyl groups of bile acids have significant impact on the lag time and burst rate of the displacement kinetics.

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1. Introduction

Bile acids are important biological surfactants formed by the enzymatic catabolism of cholesterol in liver [1]. They have a rigid, quasi-planar steroid backbone with hydroxyl groups on the concave α face and methyl groups on the convex β face. The facial amphiphilic nature of bile acids makes them extremely surface active in displacing surfactants from the oil/water interface during fat digestion [2]. The interaction of bile acids with surfactants at the oil/water and air/water interfaces has been studied with several experimental methods, including surface tension [3–5], zeta potential [5], and atomic force microscopy [6]. These studies have suggested that the competitive adsorption of bile acids can disrupt

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http://dx.doi.org/10.1016/j.colsurfa.2015.02.026
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the packing of the surfactants and eventually displace them from the interfaces. However, the effect of the nature of bile acids on the displacement kinetic has not been fully understood.

Liquid crystals (LCs) are a sensitive material with long-range orientational order [7]. The orientation of LCs is sensitive to the change of the surface which they are in contact with. This surface-induced local order can be amplified over several tens of micrometers in LC bulk due to the long-range interaction of LCs. The optical amplification of LCs makes them a unique optical probe for imaging the molecular ordering [8,9] and chemical patterns [10–12] of organic layers and sensing the chemical reactions including enzymatic reactions [13,14], DNA hybridization [15,16], ligand-receptor bindings [17,18], and peptide–lipid interactions [19,20] at the LC/aqueous interface.

In a previous publication [21], we showed that the competitive adsorption of cholic acid (CA) could displace surfactants from the LC/aqueous interface and consequently trigger a homeotropic-to-planar anchoring transition of the LC at the interface, which allowed the displacement of the surfactants to be monitored by a polarizing optical microscope. The critical concentration of CA required to displace the surfactants from the LC/aqueous interface was found to be affected by the nature of LCs. There are four unconjugated bile acids in human body. Cholic acid (CA) and chenodeoxycholic acid (CDCA) are primary bile acids, which are directly converted from cholesterol by liver. Deoxycholic acid (DCA) and lithocholic acid (LCA) are secondary bile acids, which are converted from CA and CDCA by bacterial enzymes in colon, respectively [1]. These unconjugated bile acids differ in the number and position of hydroxyl groups on their steroid backbones. In this paper, we study the time course of the displacement of sodium dodecyl sulfate from the LC/aqueous interface by LCA, DCA, CDCA and CA by observing the anchoring transition of the LC at the interface, which is triggered by the displacement. We find that the displacement shows lag-burst kinetics: a lag phase is followed by a burst phase. The lag time and the burst rate are associated with the number and position of the hydroxyl groups of bile acids.

2. Experimental

2.1. Materials

Liquid crystals (LCs) used in our experiments are 4-cyano-4′-pentylbiphenyl (5CB, 98% purity) and 4-(4-pentylcyclohexyl) benzonitrile (5PCH, 99% purity) from Sigma–Aldrich (St. Louis, MO). Sodium dodecyl sulfate (SDS), cholic acid (CA), deoxycholic acid (DCA), chenodeoxycholic acid (CDCA), and lithocholic acid (LCA) were obtained from Sigma–Aldrich (St. Louis, MO). Cholyl-lysyl-fluorescein (CLF) was purchased from BD Biosciences (Woburn, MA). All chemicals were used without further purification. Water used in our experiments was purified using an Easypure II system (18.2 MΩ cm and pH 5.7). Phosphate buffered saline solution (PBS) with 1.19 mM phosphates, 13.7 mM sodium chloride, 0.27 mM potassium chloride, and pH 7.4 was from Fisher Scientific (Fair Lawn, NJ). Total ionic strength of PBS is 171.88 mM. Polyimide coated glass substrates used for inducing homeotropic anchoring of liquid crystals were purchased from AWAT PPW (Warsaw, Poland). Glass microscopy slides were from Fisher Scientific. Copper TEM grids (18 μm thickness, 285 μm grid spacing, and 55 μm bar width) were obtained from Electron Microscopy Sciences.

2.2. Preparation of liquid crystal films

Copper TEM grids were cleaned with ethanol and then dried. The cleaned TEM grids were placed on a polyimide-coated glass substrate. LCs we used were a mixture of 19 wt% of 5PCH and 81 wt% of 5CB. The mixture was more sensitive in detecting the interaction between SDS and CA, compared to pure 5CB [21]. One microliter of the LC mixture was filled in the pores of the TEM grid supported by the polyimide-coated glass substrate. The excess LC was removed from the grid by dipping the LC filled grid into water, which led to the formation of a thin LC film confined in the pores of the grids. The confined LC film was then immersed in PBS solution containing 50 μM SDS. The adsorption of SDS led to the formation of a SDS-laden LC/aqueous interface.

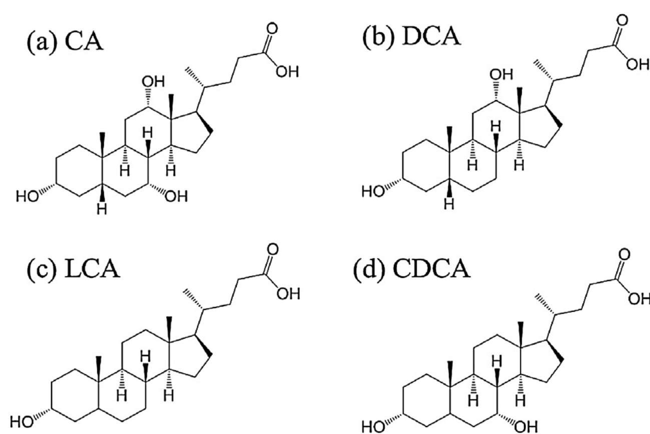

Fig. 1. Chemical structures of CA (a), DCA (b), LCA (c), and CDCA (d).

2.3. Optical observation

The optical texture of the LC films confined in the pores of the TEM grids was examined by using a polarizing optical microscope (BX 40, Olympus) in transmission mode at 25 °C. All optical microscopy images were taken with a digital camera (C2020 Zoom, Olympus) mounted on the polarizing optical microscope and then analyzed with NIH Image J. Fluorescence microscopy images were acquired with a confocal fluorescence microscope (Zeiss TCS SP5MP) with 488 nm excitation from an Ar^+ laser.

3. Results and discussion

Four unconjugated bile acids (CA, CDCA, DCA, and LCA) were chosen for studying the comparative kinetics of the displacement of SDS from the LC/aqueous interface. Their chemical structures are shown in Fig. 1. LCA has only one hydroxyl group at C-3 position. DCA has two hydroxyl groups at C-3 and C-12 positions. CDCA has two hydroxyl groups at C-3 and C-7 positions. CA has three hydroxyl groups at C-3, C-7, and C-12 positions. The hydrophobicity of these bile acids increases with the increase of the number of hydroxyl groups on their steroid backbones. It has been shown that the order of hydrophobicity is LCA > DCA > CDCA > CA [22]. The critical micelle concentration (CMC) of LCA, CDCA, DCA, and CA is 0.9, 9, 10, and 18 mM, respectively [23]. 5PCH and 5CB are widely used cyano-containing LCs. As compared to 5CB, 5PCH has a more flexible and bulky core containing one phenyl ring and one cyclohexane ring. The flexible and bulky 5PCH requires higher anchoring energy to achieve homeotropic anchoring, compared to the rigid 5CB [24]. The adsorption of SDS at the 5PCH/aqueous interface is unable to induce a homeotropic anchoring of the 5PCH at the interface. It has been shown that 5PCH and 5CB can form nematic mixtures over a wide range of mixed ratios [25]. The adsorption of SDS at the 5PCH/5CB mixture-aqueous interface can induce a homeotropic anchoring of the 5PCH/5CB mixtures with the mixed ratio up to 19 wt% of 5PCH. We have shown that the LC mixture of 19 wt% of

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