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Bile salts and derivatives: Rigid unconventional amphiphiles as dispersants, carriers and superstructure building blocks

Luciano Galantini [a,*], M. Chiara di Gregorio [a], Marta Gubitosi [a], Leana Travaglini [a], José Vázquez Tato [b], Aida Jover [b], Francisco Meijide [b], Victor H. Soto Tellini [c], Nicolae V. Pavel [a]

[a] Dipartimento di Chimica, Università di Roma "La Sapienza", P. le A. Moro 5, 00185 Roma, Italy
[b] Departamento de Química Física, Facultad de Ciencias, Universidad de Santiago de Compostela, Avda. Alfonso X El Sabio s/n, 27002 Lugo, Spain
[c] Escuela de Química, Centro de Investigación en Electroquímica y Energía Química (CELEQ), Universidad de Costa Rica, San José, Costa Rica

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ABSTRACT

Steroidal surfactants like bile salts have a rigid amphiphilic structure significantly different from the typical head–tail one. As a result, these molecules show peculiar features in their self-assembly behavior and solubilization and permeation abilities. Bile salts are widely used as starting materials in the preparation of synthetic derivatives by changing their amphiphilic structure and by introducing specific functionalities. Due to the steroid rigidity and the peculiar distribution of hydrophobic and hydrophilic domains, these molecules self-organize in ordered supramolecular assemblies and are particularly attractive for the bottom up construction of complex nanostructures. They often self-assemble in 1D structures such as tubes or fibers and show low molecular weight gelator features. Their tubular nanoscale structures have cross section diameters spanning a wide range of values (inner diameter 3–450 nm) and are sometimes formed through appealing pH or temperature responsive aggregations. Moreover, mixtures of these surfactants allow in some cases the preparation of mixed tubes with tunable composition and related features such as charge and sizes. The unconventional amphiphilic molecular structure of BSs dictates also remarkable abilities as carriers across tissues and membranes of many compounds (e.g. drugs, carbohydrates and ions). Therefore they are often employed as encapsulators, dispersants and transporters in complex systems. Chemical modifications can also be used to provide derivatives with improved performances.
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1. Introduction

Conventional surfactants are generally depicted with a head–tail structure, where the head represents the hydrophilic moiety and the tail is a flexible hydrophobic chain [1]. Steroidal surfactants are significantly different as they are rigid and have a rather complex distribution of polar and non-polar regions [2]. As a result, these molecules show peculiar features in their self-assembly behavior, solubilization and permeation abilities. Bile salts (BSs) are anionic natural steroidal surfactants, found predominantly in bile of mammals and widely involved in various biological processes and applications. Moreover, BSs are widely used as starting materials in the preparation of synthetic derivatives (Table 1). By properly choosing the BS and substituents, it is possible to tune the distribution of hydrophobic/hydrophilic region on the molecule and to introduce specific functionalities, thus preparing new molecules with uncommon features. Due to the steroid rigidity and the peculiar distribution of hydrophobic and hydrophilic domains, these molecules self-organize in ordered supramolecular assemblies in solution and are particularly attractive for the bottom up construction of complex nanostructures. It was demonstrated, for example, that BS-based molecules are particularly versatile building blocks for the preparation of 1D nanostructures like supramolecular tubules or fibers. Tubular nanoscale structures are very attractive in the field of nanotechnology, where they can be used in a wide range of applications with specific roles according to their dimensions. With this respect, BSs and BS derivatives (BSDs) were proved to be able to form nanochannels spanning a wide range of sizes (cross section diameters ranging from 3 to about 450 nm and lengths up to 100 μm), in some cases through appealing stimuli dependent aggregations (pH [3, 4•] or temperature responsiveness [4•–6•]) or with tunable features such as charge [7••] or size [8••]. Moreover, they exhibit remarkable features as gelators due to their ability to form networks of interconnected fibers. Beside the peculiar aggregation, a number of BSDs show other interesting features as efficient receptors, carriers and protein, drug or carbon nanotube solubilizers and stabilizers, thank to specific recognition, dispersion and permeation abilities. This review was focused on recent developments in fundamental research and applications of BSs and BSDs. Some general features of natural BS self-assembly were described in the first section to provide background information and BSDs obtained by modification of a single BS molecule (mono-steroidal BSD) were illustrated in the second section. Subsections were created to separate

* Corresponding author.
E-mail address: luciano.galantini@uniroma1.it (L. Galantini).

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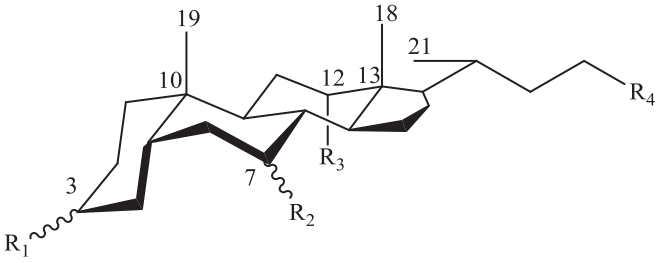
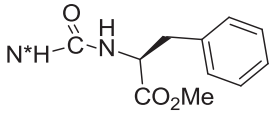
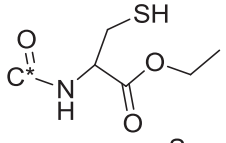
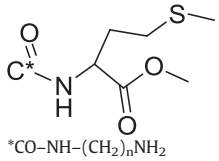
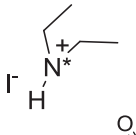
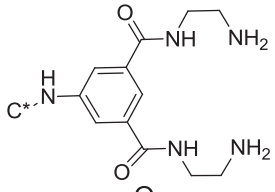
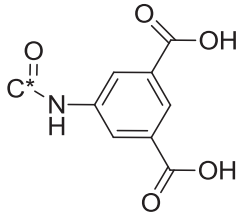
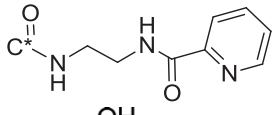
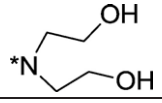
Table 1
Structure, acronyms and identification numbers of natural BSs and their mono-steroidal derivatives. All substituents have an α-orientation; exceptions are indicated. Linking atoms are indicated with an asterisk where needed.



R_1	R_2	R_3	R_4	Acr./Id. num.
OH	OH	OH	$^*CO_2^-\ Na^+$	NaC
OH	OH	H	$^*CO_2^-\ Na^+$	NaCDC
OH	H	OH	$^*CO_2^-\ Na^+$	NaDC
H	H	H	$^*CO_2^-\ Na^+$	NaLC
OH	H	OH	$^*CO–NH–CH_2–CO_2^-\ Na^+$	NaGDC
OH	H	OH	$^*CO–NH–CH_2–CH_2\ SO_3^-\ Na^+$	NaTDC
OH	OH	OH	$^*CO–NH–CH_2–CH_2\ SO_3^-\ Na^+$	NaTC
OH	β-OH	H	$^*CO_2^-\ Na^+$	NaUDC
OH	OH	OH	$^*CO–NH–NH–CO–(CH_2)_9CO_2H$	**1**
OH	OH	OH	$^*CO–NH–NH–CO–(CH_2)_5CO_2H$	**2**
OH	OH	OH	*CO–NH–NH–CO–4-chlorobenzoic	**3**
OH	OH	OH		**4**
OH	OH	OH	$^*NH–(CH_2)_2–SH$	**5**
OH	OH	OH		**6**
OH	OH	OH		**7**
OH	H, OH	H, OH	$^*CO–NH–(CH_2)_nNH_2$	**8**
OH	H, OH	H	$^*CO–NH–CH_2–CH(OH)–CH_2OH$	**9**
OH	H	H		**10**
OH	OH	OH		**11**
OH	OH	OH		**12**
OH	H	OH		**13**
OH	H	OH		**14**

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