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# Adsorption of poly(diallyldimethylammonium chloride)—sodium methyl-cocoyl-aurate complexes onto solid surfaces

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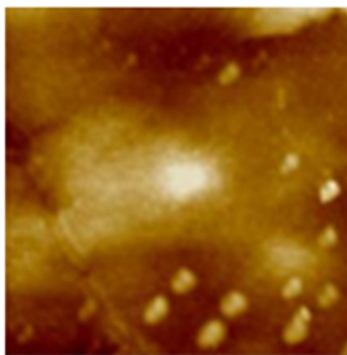
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### HIGHLIGHTS

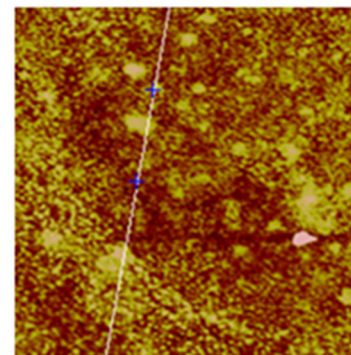
- Oppositely charged polymer–surfactant mixtures are used in capillary cosmetic.
- Conditioning is related to adsorption onto negatively charged surfaces.
- Adsorption onto solid surfaces depends on the bulk behavior of the mixtures.
- PDADMAC–SCMT mixtures are useful in the development of cosmetic formulations.
- Adsorption of mixtures onto solid surfaces governed by diffusion.

### GRAPHICAL ABSTRACT

#### Polymer-Surfactant Systems Adsorbed Onto Hair Fibers and Model Surfaces



Hair Fibers



Model Surfaces

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### ABSTRACT

The study of the adsorption of supramolecular complexes formed by a highly charged cationic homopolymer (poly(diallyldimethylammonium chloride), PDADMAC), and a sulfate-free anionic surfactant, sodium methyl cocoyl taurate (SMCT), on liquid/solid interfaces was performed by dissipative quartz crystal microbalance, ellipsometry and atomic force microscopy. Moreover, turbidimetry, dynamic light scattering and  $\zeta$ -potential measurements were carried out to characterize the bulk solutions. The complexes adsorb onto the solid surfaces, leading to inhomogeneous films formed by isolated aggregates randomly distributed through the surface. Additionally, the adsorption kinetics of the bulk complexes onto the negatively charged surface was studied. This work aims to understand the physico-chemical mechanisms involved in the adsorption of polycation–negatively charged surfactant mixtures, interesting for the design of conditioner cosmetic formulations, onto negatively charged surfaces. It is expected that the results here obtained may provide a better understanding of the conditioning effect of cosmetic products on keratinic (hair) substrates.

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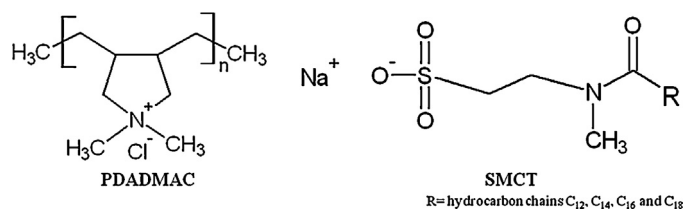
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## 1. Introduction

Mixtures formed by polyelectrolytes and oppositely charged surfactants play a key role in many technological applications ranging from thin films, foams and emulsion stabilization to fabrication of coatings and encapsulation [1]. This has stimulated important research efforts to understand the behavior of polymer – surfactant mixtures in the bulk and at interfaces [2–5]. Mixtures of polyelectrolyte and surfactant bearing opposite charges present a special relevance since they are used in tertiary oil recovery, fabrication of functional materials or formulation of personal care products (mainly cationic polymers) [6,7]. However, many physico – chemical aspects involved in their technological applications remain unclear, in part due to the absence of comprehensive studies [2,7] on the adsorption of the polyelectrolyte – surfactant complexes to interfaces in similar conditions than those occurring during real applications [3]. Furthermore in the last years several studies have been devoted of the correlation between the layer adsorption and the bulk properties of the solutions [8–13]. Understanding the behavior of these systems requires to consider electrostatic and hydrophobic interactions [14] that lead to very complex phase diagrams [12,15,16]. The adsorption strongly depends on the region of the phase diagram considered for the sample studied; for instance, the adsorption at the solid/liquid interfaces usually increases near the coacervation/precipitation region [2,3,17–20]. In recent years, several studies have also tried to correlate the adsorption behavior of polyelectrolyte – surfactant at the air/water interface to the bulk properties of the complexes [21–25]. Furthermore, the mechanical response to shear or/and dilation stresses of the interfacial layers has been also described [26], as well as the adsorption of these systems onto solid surfaces [3,27–29]. Results on the morphology and internal organization of polyelectrolyte – surfactant complexes onto solid surfaces has been reported [30], pointing out a broad variety of possible structures of polyelectrolyte – surfactant complexes, that range from disordered layers to lamellar structures [30]. It is worth mentioning that this type of polyelectrolyte – surfactant systems form complexes that usually are non-equilibrium structures. Therefore, the results are highly sensitive to the procedure used to prepare the solutions [2,3,17–20]. Another characteristic of these systems is their ability to modify the tribological properties of surfaces, which is important for industrial applications [31], e.g., cosmetic formulations [32].

Here, we report a study of the bulk properties and the adsorption onto negatively charged solid substrates of a frequently studied polycation [33–38], poly(diallyldimethylammonium chloride) (PDADMAC), mixed with an anionic surfactant, sodium methyl cocoyl taurate (SMCT). SMCT is an example of sulfate-free surfactant frequently used in shampoos and hair conditioning formulations products [32]. Therefore, the study of the adsorption of this type of mixtures onto solid surfaces can help to understand the physico-chemical phenomena involved in the conditioning process of cosmetic formulations upon adsorption onto complex cosmetic substrates such as hair fibers. Hair fibers after weathering or bleaching treatments present sulfonate groups,  $\text{SO}_3^-$  on their surface (density about  $2.2 \text{ SO}_3^-/\text{nm}^2$  on a bleached hair), thus negatively charged solids substrates become good models for carrying out studies to correlate with the real performance of commercial formulations [39,40]. The choice of the system have been done on the basis of the interest of cosmetic industry for replacing the sulfate based surfactant from shampoo and conditioner formulations by surfactants less aggressive from a dermatological point of view. Thus, a sulfate-free surfactant from a natural source, such as the SMCT (derived from the coconut oil), can be a good candidate.



Scheme 1. Molecular structures of PDADMAC and SLES.

## 2. Experimental section

### 2.1. Chemicals

All chemicals were used without further purification. The ultrapure deionized water used for cleaning and solubilization presents a resistivity higher than  $18 \text{ M}\Omega\text{cm}$  and a total organic content lower than 6 ppm (Younglin 370 Series, South Korea). Poly(diallyldimethylammonium chloride) (PDADMAC) was purchased from Sigma-Aldrich (Germany), having an average molecular weight in the 100–200 kDa range. Sodium methyl cocoyl taurate (SMCT) from L'Oréal was used as anionic surfactant. In house characterization of SMCT shows that the surfactant was a polydisperse mixture with molecules containing hydrocarbon chain (R group or surfactant tail) of various lengths, i.e.,  $\text{C}_{12}$  (55 wt%),  $\text{C}_{14}$  (20 wt%) and  $\text{C}_{16}/\text{C}_{18}$  (25 wt%). In order to test the role of the surfactant purity on the adsorption of the mixtures studied, we have studied several solutions in which the SMCT was recrystallized from ethanol following the procedure described by Song et al. [41], and the results obtained agreed with those obtained with the surfactant before crystallization within the experimental repeatability. This composition corresponds to that of SMCT to be used in real cosmetic formulation [42,43]. The polydispersity of PDADMAC was not considered as a problem for our experiments, in our studied it is possible to consider that the adsorption times used are long enough to obtain films mainly formed by the largest chain, being the shortest displaced to the solution [44]. Scheme 1 shows the molecular structures of PDADMAC and SMCT.

The pH of all solutions was adjusted at 5.6 using glacial acetic acid (purity >99%) and the ionic strength was kept constant by adding 0.3 wt% of KCl (purity >99.9%) to match the conditions of most hair care formulations to decrease the bleaching of hair surface. These conditions minimize the skin irritation [43]. All experiments were carried out at  $25.0 \pm 0.1 \text{ }^\circ\text{C}$ . The SMCT concentration,  $c'$ , is defined as the ratio between the mass of surfactant,  $m_{\text{SMCT}}$ , to the total mass of the solution,  $m_{\text{T}}$ . The PDADMAC concentration used for all the solutions was 0.5 wt%, that corresponds to the dilution of a typical shampoo, when applied onto wet hair [43].

Since the polyelectrolyte-surfactant solutions frequently lead to kinetically arrested non-equilibrium structures [10,45,46], the same protocol has been followed to prepare all the solutions prior their use. Thus, all solutions were used within one week of their preparation.

### 2.2. Techniques

#### 2.2.1. Bulk Characterization

The characterization of the size and charge of the species in solution/dispersion was performed through dynamic light scattering (DLS) and  $\xi$ -potential measurements, respectively. For this purpose, a Zetasizer Nano ZS from Malvern Instruments (United Kingdom) was used.

The hydrodynamic diameter,  $d_{\text{H}}$ , was estimated from DLS measurements, using the red line ( $\lambda = 632 \text{ nm}$ ) of a He-Ne laser in a quasi-backscattering configuration ( $\theta = 173^\circ$ ). The  $\xi$ -potential was

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