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Improvement of the surface wettability of silicone hydrogel films by self-assembled hydroxypropyltrimethyl ammonium chloride chitosan mixed colloids

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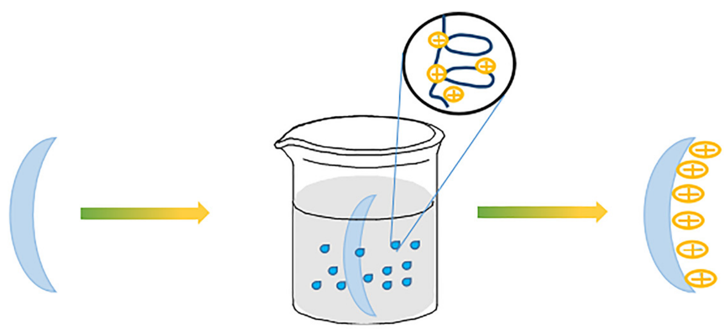
GRAPHICAL ABSTRACT



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ABSTRACT

A novel kind of hydroxypropyltrimethyl ammonium chloride chitosan (HACC) mixed colloid was developed to improve the hydrophilicity of silicone hydrogel films (SHFs) via self-assembly. The contact angle of the modified SHF decreased from 33.24° to 12.98°, whereas the atomic force microscopy image verified that the roughness of the modified films decreased and indicated that the HACC mixed colloids filled in the interstices on the surface of the SHF. A new method of capacitance was developed to characterize the electric charge value, the groups, and the microscopic conditions of the self-assembled SHF surface for the first time. Cyclic voltammetry and chronopotentiometry exhibited that the capacitance of the SHF increased to 0.0196 F/g, which indicated the enhanced positively charged surface and increased the number of hydrophilic quaternary ammonium groups on the surface. Lipid adsorption analysis also revealed that the modified SHF possessed a weaker lipid adhesion ability and stronger lipid removal ability than those of the original SHF and hence showed promising features for application in ophthalmological products.

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1. Introduction

The special wetting behavior of hydrophilicand hydrophobic solid surfaces have attracted great interest in recent years and played an important role in fundamental research and practical applications [1]. To date, various general techniques have been developed to fabricate hydrophilic and hydrophobic films. These techniques include mechanical processing [2], physical and chemical vapor deposition [3–5], sol-gel formation [6–8], and layer-by-layer deposition [9–13]. Many studies relating to self-assembled nanoparticles, including AuNPs and AgNPs [14,15], have been conducted, whereas research on mixed colloid self-assembly and characterization, which is highly essential, is rare. Zhao et al. [16] applied the surface grafting technique to attach functional monomers or polymers onto the surface of base film to improve the surface's hydrophilicity, whereas Asatekin et al. [17] mixed the hydrophilic polymer with film material to enhance the film's hydrophilicity. In addition, Lin et al. [18] enhanced the surface wettability of silicone hydrogel contact lenses by layer-by-layer self-assembly techniques.

Hydrogel with superior performance is composed of polymeric networks with a high level of hydration and large specific surface area besides possessing 3D microstructures similarly to natural tissues [19]. Since the first-time establishment of synthetic hydrogels by Wichterle and Lim in 1954 [20], hydrogel technologies have played crucial roles in numerous fields including agriculture [21], drug delivery systems [22], wastewater treatment [23,24], biomedical applications [25], regenerative medicines [26], wound dressing [27], separation of biomolecules or cells [28] and bionic fields [29,30]. Hydrogels possess numberous physical and chemical characteristics that can help fulfill requirements in different situations. These attributes include high absorption capacity, the low soluble content and residual monomer, photostability, rewetting capability, notch insensitivity, and low lipid adhesion [31,32]. Given the hydrophobic feature, soft siloxane groups in the silicon-based hydrogel are easily migrate to the surface of the low free-energy material, followed by rearrangement, this behavior substantially impedes the development of silicone hydrogel. Furthermore, hydrophobicity can lead to decreased surface wettability and increased lipid interactions [33–35] that severely limit the applications of the silicone hydrogel in certain fields, such as commercial contact lens material. Much work has been carried out to improve the preparation and self-assembly modification of silicone hydrogel film (SHF). Zhao et al. [36] prepared novel porous hydrogel films based on glycerophosphate and chitosan. However, the poor solubility of chitosan in water considerably limits chitosan's practical applications [37].

Recently, quaternary ammonium chitosan has received extensive attention due to its excellent properties. Hydroxypropyltrimethyl ammonium chloride chitosan (HACC), a quaternary ammonium salt of chitosan with good film-forming ability, moisture-retention capacity, high antibacterial activity, high biocompatibility, and high biodegradability, has been applied in textile- and industrial water treatments, paper handling and the medical fields [38–42]. Instead of sole usage, HACC is generally combined with other molecules and consequently demonstrates its complex performance after interaction. In our previous studies, the behavior of HACC in aqueous solutions was discussed. In addition to van der Waals force, the positively charged HACC and the uncharged poloxamer form a positively charged bead string core-shell microsphere mixed colloid through hydrophobic interaction and hydrogen bonding [43,44]. Herein, we modified the SHF with these colloids. Unlike the self-assembly of a single colloid, the characterization of the self-assembled SHF was a challenge due to the complexity of the HACC mixed colloids. To solve this problem, we first used AC impedance (EIS) to characterize the self-assembled SHF.

In this paper, to improve the hydrophobicity and anti-lipid adhesion ability of SHF, a new surface was obtained via the self-assembly of SHF with HACC mixed colloids. The newly formed surfaces were investigated by static contact angle measurements and atomic force microscopy (AFM) measurements. In addition, for the first time, AC impedance (EIS), cyclic voltammetry (CV) and chronopotentiometry (CP) were applied to study the HACC mixed colloids self-assembled SHF. Furthermore, the lipid adsorption and removal properties of modified SHF have also been observed by light transmittance measurements. Protein adsorption was also investigated. The newly developed method can effectively characterize polymer colloid self-assembly interface and also be widely applied to various systems.

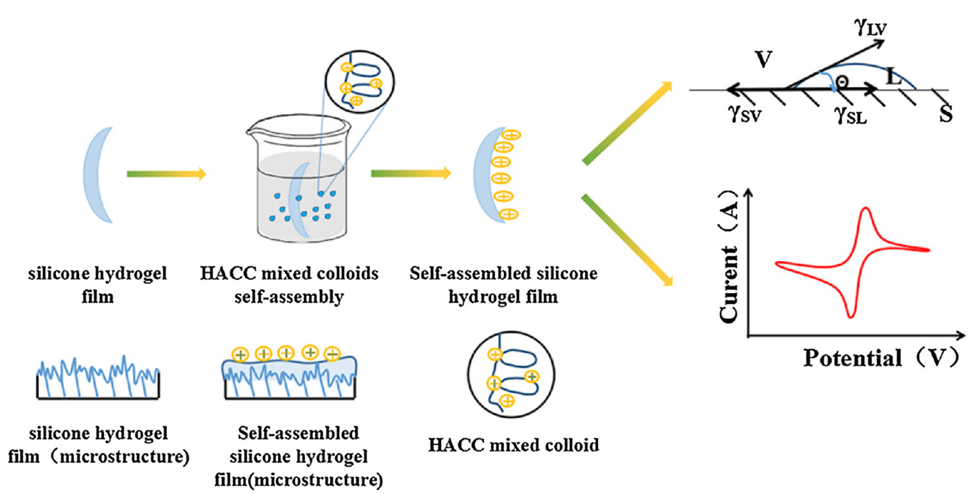
2. Material and methods

2.1. Material

Hydroxypropyl trimethyl ammonium chloride chitosan (HACC) was purchased from Jiaxing Korui Technical co., Ltd. (with deacetylation degree of 83.6% and a molecular weight of 800 kDa). Poloxamer 407 (USP) was purchased from BASF. Polyhexamethylene biguanide (PHMB) and Dimethylthetin (DMT) were purchased from Wuhan HongRuiKang Reagent co., Ltd. Sodium chloride, boric acid, sodium borate, cholesterol, lysozyme, bovine serum albumin, γ-globulin and acetic acid were purchased from Sinopharm Chemical Reagent co., Ltd. Ultrapure water was used throughout.

2.2. Preparation of test solution and SHF

0.25 g/L HACC, 1.3 mg/L PHMB, 25 mg/L DMT, 1.75 g/L poloxamer and borate buffered saline (BBS) were dissolved in ultrapure



Scheme 1. Schematic of the modification and characterization of SHF.