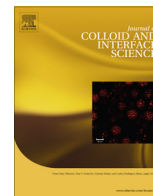




Contents lists available at ScienceDirect

Journal of Colloid and Interface Science

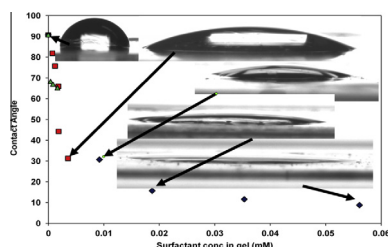
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Incorporation of polymerizable surfactants in hydroxyethyl methacrylate lenses for improving wettability and lubricity

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GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 2 October 2014

Accepted 16 December 2014

Available online 29 December 2014

Keywords:

Wettability

Lubricity

Contact lenses

Friction

Contact angle

Polymerizable surfactants

P-HEMA

ABSTRACT

Dryness and discomfort are the main reasons for dropouts in contact lens wearers. Incorporating surfactants in lens formulations could improve wettability and lubricity, which can improve comfort. We have focused on incorporating polymerizable surfactants in hydroxyethyl methacrylate lenses to improve comfort, while minimizing the potential for surfactant release into the tears. The surfactants were added to the polymerization mixture, followed by UV curing and extraction of leachables in hot water. Wettability and lubricity were characterized by measuring the contact angle and coefficient of friction. Lenses were also characterized by measuring transmittance, loss and storage moduli and ion permeability. Incorporation of surfactants significantly reduced contact angle from 90° for p-HEMA gels to about 10° for 2.43% (w/w) surfactant loading in hydrated gel. The coefficient of friction also decreased from about 0.16 for HEMA gels to 0.05 for the gels with 2.43% surfactant loading. There was a good correlation between the contact angle and coefficient of friction suggesting that both effects can be related to the stretching of the surfactant tails near the surface into the aqueous phase. The water content was also correlated with the surfactant loading but the contact angle was more sensitive suggesting that the observed improvements in wettability and lubricity arise from the protrusion of the surfactant tails in into the liquid, and not purely from the increase in the water content. The gels were clear and certain compositions also have the capability to block UVC and UVB radiation. The results suggest that incorporation of polymerizable surfactants could be useful in improving surface properties without significantly impacting any bulk property.

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1. Introduction

Almost 17% of new contact lens wearers drop out in the first year of wear, which is a major concern for the contact lens industry [1]. Several reasons cause the high drop rates but in all studies,

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discomfort and dryness are the leading causes, sometimes accounting for almost 50% of the drop outs [2–4]. In spite of the continuous improvements in lens design, a very large fraction of the contact lens users experience at least some form of dryness and discomfort, particularly towards the end of the day [5]. The mechanisms that cause the dryness and discomfort are complex and not completely understood so a number of researchers have focused on exploring correlations between various lens properties and discomfort and dryness. A recent study showed that excessive lens movement, inferior lens decentration, poor surface wettability and deposits, inferior corneal staining, and Asian ethnicity are associated with dryness and discomfort [6]. In another study [7] analysis of end of day comfort data for several different lens types showed a significant correlation with the coefficient of friction data [8]. The analysis also showed a lack of correlation between comfort and oxygen permeability, modulus and water content. The same researchers had earlier shown a correlation between comfort and friction coefficient measured with a different technique [9], which suggests that the friction coefficient is the key lens property associated with the end of day comfort, irrespective of the technique used in the measurement. Though detailed mechanisms are not understood, the correlation of dryness and discomfort to lubricity can be related to the rapid motion of the upper eyelid over the contact lens surface during a blink. Irrespective of the mechanisms, it is believed that lubricity and wettability impact dryness and discomfort, and so significant efforts have been made to improve both of these properties.

Efforts to improve wettability have focused on incorporation of highly hydrophilic monomers, plasma surface treatments and addition of internal wetting agents such as polyvinyl alcohol (PVA) or polyvinyl pyrrolidone (PVP) [10]. Extended release of PVA incorporated in Focus DAILIES® (CIBA Vision) into the tears improves comfort [11,12]. Incorporation of PVP in Acuvue® Oasys™ with Hydraclear™ Plus lenses (Johnson and Johnson) increases wettability eliminating the need for surface treatment typically required in silicone hydrogel contact lenses. Contrary to the PVA incorporation, the PVP loaded in lenses remains trapped due to the high molecular weight but it is believed to partially diffuse to the surface to increase the wettability [13]. Hyaluronic acid (HA) has also been explored as a potential biocompatible comfort enhancer [14] and lenses loaded with sodium hyaluronate (Hyaluronate Gel) have been commercialized (Safigel™).

The incorporation of wetting and comfort agents in contact lenses has improved comfort but yet a large number of patients experience discomfort and dryness. For achieving improvements in comfort, lens modifications have been complemented with efforts for designing better formulations of rewetting drops and lens care solutions to improve the comfort. Both PVA and PVP and also HA have been used in artificial tears and rewetting drops for contact lenses [5]. Addition of surfactants in rewetting drops and lens care solutions has been found to be beneficial most likely due to improved wettability of the surface due to surfactant adsorption [15,16]. The adsorbed and absorbed surfactant however desorbs with time, possibly leading to the reduced end of day comfort [17]. Also, the preservatives included in the formulations can absorb into the lenses and then desorb during lens wear which could lead to patient discomfort and potential toxicity issues [18–21].

The prior research clearly shows the benefits of incorporation of wettability enhancers and the surfactants in the lens or in the lens care solutions. The disadvantage though is that the degree of contact angle reduction is not very large likely due to low packing at the interface and the beneficial effects are only transient due to desorption and drainage of the surfactant. We propose that a densely packed thin layer of surfactant at the interface could achieve very large improvements in wettability and lubricity, and this

effect could be rendered irreversible by crosslinking the surfactant at the interface. Here we propose to covalently attach surfactants to the lens matrix to increase wettability and lubricity while eliminating desorption and release into the eye. The covalent attachment is achieved by adding polymerizable surfactants with an ethylenically unsaturated bond to the monomer mixture, followed by UV based lens curing. The surfactants are chosen to have long hydrophilic tails and we hypothesize that the hydrophilic groups located at the surface will likely extend into the water to form a structure similar to polymer brushes. Several studies have shown that grafted polymer brushes can reduce friction though the molecular reason for this effect are still uncertain. To explore the potential of our approach, we focus only on the hydroxyethyl methacrylate (HEMA) gels, which can be used as contact lens materials.

2. Materials and methods

2.1. Materials

2-Hydroxyethyl methacrylate (HEMA) monomer and crosslinker ethylene glycol dimethacrylate (EGDMA) were purchased from Sigma–Aldrich Chemicals (St Louis, MO). Sodium chloride (99.9+%) was purchased from Fisher Scientific (Fairlawn, NJ). 2,4,6-trimethylbenzoyl-diphenyl-phosphineoxide (Darocur® TPO) was kindly provided by Ciba (Tarrytown, NY). Noigen RN-10™ (N10), Noigen RN30™ (N30) and Hitenol BC30™ (H30) were kindly provided by Montello Inc (Tulsa, OK). All the chemicals were reagent grade and were used without further purification.

2.2. Preparation *p*-HEMA gels with polymerizable surfactants

The structures of the three polymerizable surfactants used in this study are shown in Fig. 1. These surfactants were chosen because of the vinyl group which allows copolymerization with the HEMA monomer and due to similarities in structure to the Brij surfactants which have been explored in several ophthalmic applications. The N10 and N30 belong to the Noigen series with values of 10 and 30 for n , respectively, while H30 belongs to the Hitenol series with $n = 30$. The molecular weights of N10, N30 and H30 are 700, 1580 and 1677 respectively. The surfactant loaded gels were prepared by photo-polymerization of an aqueous solution of the surfactants, monomer (HEMA), crosslinker (EGDMA) and the initiator Darocur® TPO. The exact compositions of the solutions are included in Table 1. The solution was degassed to reduce the concentration of dissolved oxygen by bubbling nitrogen for 15 min. 12 mg of the photoinitiator (Darocur® TPO) was then added, accompanied by stirring at 750 rpm for 5 min to ensure complete dissolution of the initiator. The solution was then poured in between two 5 mm thick glass plates that were separated by spacers approximately 780, 240 and 100 μm in thickness. The mold was then placed on Ultraviolet transilluminator UVB-10 (UltraLum, Inc.) and the gel was cured by irradiating UVB light (305 nm) for 40 min. The gels were removed from the glass plates and cut into rectangles pieces of the desired size. The unreacted

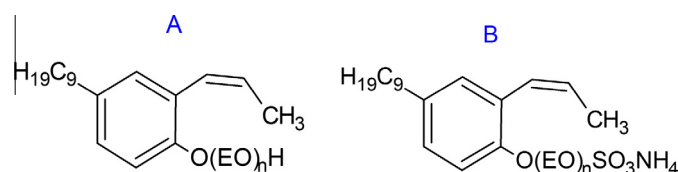


Fig. 1. Structure of the polymerizable surfaces (A) Noigen series and (B) Hitenol series.

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