

Charge and size matters—How to formulate organomodified silicones for textile applications



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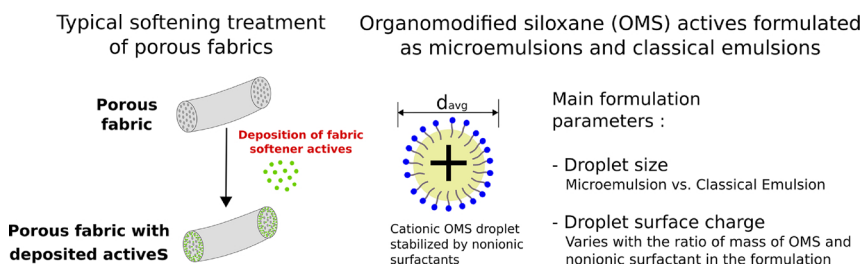
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GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Fabric softener
Organomodified silicones
Silicone quats
Deposition
Streaming potential
Formulation

ABSTRACT

Organomodified silicones (OMS), which conventionally find use in textile finishing processes, have recently become a very interesting prospect in the field of fabric softeners. Here, we present OMS-based fabric softener formulations in the form of classical emulsion (droplet size $\approx 0.1 \mu\text{m} - 10 \mu\text{m}$) and microemulsions (droplet size $\approx 5 \text{ nm} - 50 \text{ nm}$) using nonionic surfactants (NIS) as emulsifier. Streaming potential measurements are used to obtain a measure of droplet surface charge, and it was found to be related to the ratio of masses of OMS and NIS present in the formulations. In this work, it is investigated how the performance of these formulations is influenced by properties such as droplet size and streaming potential. Panel tests were carried out to evaluate the sensory properties of fabric treated by these formulations, and they reveal that the classical emulsion performs better than the microemulsions. For the microemulsions, it is found that softening performance increases with streaming potential. The observed trends in softening performance are explained by considering the difference in location or penetration of softening actives on or into fabric. Two different experimental approaches are implemented to gain insights into the underlying phenomena. In the first approach, batch deposition experiments are carried out to characterize OMS deposition on fabric. In the second approach, a chromatographic technique is used to compare the deposition kinetics of different softening formulations. The findings of the experiments provide insights into the reasons underlying the contrasting softening performance. The final results are discussed with respect to existing literature.

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<https://doi.org/10.1016/j.colsurfa.2018.09.080>

Received 29 March 2018; Received in revised form 25 September 2018; Accepted 27 September 2018

Available online 29 September 2018

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1. Introduction

The sensory properties of textiles can be enhanced via deposition of molecules with good antifriction and lubricating properties such as siloxanes and siloxane derivatives [1,2]. Among such molecules, organomodified silicones (OMS) such as aminosiloxanes and silicone quats are typical candidates used in the industry for conditioning textiles and hair [3,4]. Due to the presence of cationic quaternary ammonium groups bound to the siloxane polymer, such OMS molecules have an overall positive charge. Therefore, their deposition on textiles is facilitated by the attractive electrostatic interactions existing between cationic OMS molecules and the negatively charged surfaces of textiles such as cotton, viscose, and polyester etc. [5–7].

Organomodified silicones, in their pure state, exist in the form of viscous liquids at ambient temperature conditions, and are, due to their incompatibility with water unsuitable for direct usage. Therefore, they need to be converted into suitable formulations which could be used for treating textiles. The most common technique used for preparing OMS formulations involves dispersing the OMS as droplets in water to form aqueous emulsions. The OMS droplets dispersed in water often need to be stabilized using emulsifiers. The stabilization can be achieved by adding both nonionic as well as charged surfactants as emulsifiers. [8] For cationic OMS molecules like aminosiloxanes and silicone quats, the use of nonionic emulsifiers is preferred [9].

The final performance of a softening formulation is typically dependent on (i) the total quantity of actives deposited per unit fabric mass, and (ii) the location of deposited actives on the fabric. However, in a normal softening treatment, the quantity of actives administered to a batch of textiles is much lower than the maximum active uptake capacity of textiles [10,11]. Both, the rate of deposition as well as the distribution of the active on / in the fabric are the two parameters distinguishing the formulations performance-wise and small changes / additions in the formulation can have large effects on the performance [12].

Due to the porous structure of fabrics, the actives in a softening formulation can deposit on both external as well as internal surfaces of fabrics. While deposition on external fabric surfaces is considered favorable for obtaining an enhancement in directly perceived softness [13], deposition on internal fabric surfaces contributes to the so-called bulk softness of textiles as well as reduced wrinkles. Furthermore, the location of deposited actives could determine how the softening treatment influences other important fabric properties such as the imbibition rate and absorption capacity of water [14].

In earlier works dealing with esterquat vesicles as the active, it has been shown that the distribution of deposited actives on fabric could be influenced by formulation properties such as vesicle size and lipid bilayer phase behavior [15,16]. As far as siloxane-based softeners are concerned, a few earlier works have considered the effect of droplet size on location of deposited actives [17–20]. A common finding in these works has been that large sized droplets deposit predominantly on external fabric surfaces, whereas smaller actives can penetrate and deposit on internal fabric surfaces. Thus, droplet size was found to directly influence the softening performance of formulations.

In the case of OMS-based fabric softeners, which are typically formulated as emulsions using nonionic emulsifiers, several factors could influence the location of deposited actives, such as (i) size of OMS droplets, (ii), degree of quaternization of OMS molecules, and (iii) quantity of nonionic emulsifier used for preparing the emulsion. The size of OMS droplets is typically determined by the method being employed for emulsion preparation, and depending upon the obtained droplet sizes the emulsions are classified into different categories, namely macro-, micro-, and nano- emulsions. Classic emulsions need to be formed by applying high shear energies to create new liquid-liquid interface. This interface is then stabilized by emulsifiers and / or thickener which slow down coarsening, ripening and creaming of the emulsion. Microemulsions however are thermodynamically stable

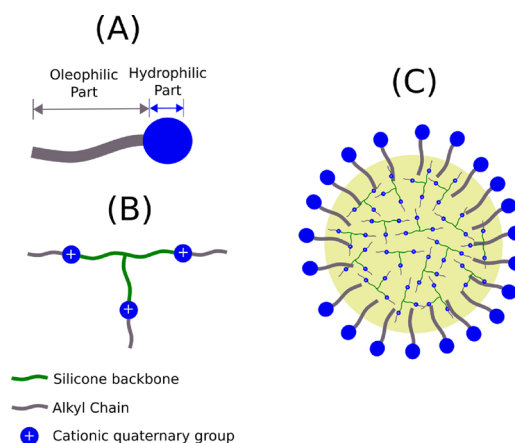


Fig. 1. Schematic representation of the nonionic emulsifier (A), cationic T-quat OMS molecule (B) and OMS droplets stabilized by nonionic emulsifier (C) (not drawn to scale).

mixtures of a hydrophilic, a hydrophobic and an amphiphilic component; therefore, they are formed spontaneously without applying high shear energies. [21,22] Microemulsions are macroscopically homogeneous but microscopically nanostructured [23]. On the other hand, the degree of quaternization of OMS molecules, and the quantity of used nonionic emulsifier together determine the surface charge of OMS droplets. The main objective of the present work is to investigate how the above factors affect the softening performance of OMS formulations.

The OMS used for preparing the softening formulations in this work is a T-shaped cationic siloxane which is presently used commercially in hair care applications (schematic chemical structure shown in Fig. 1(A)) [4,24]. This OMS contains permanently cationic (quat) groups, and it was chosen instead of an aminosiloxane so as to avoid the effects of pH variation on the degree of protonation of the siloxane [1,25]. Three different microemulsions (droplet size \approx 5 nm–50 nm) and one classical emulsion (droplet size \approx 0.1 μ m–10 μ m) were prepared using this OMS.

The surface charge of OMS droplets in the formulations was measured by determining their streaming potential. As the streaming potential of a dispersed object determines its interaction with an oppositely charged substrate, it is expected to influence the deposition of OMS droplets on fabric and as a consequence the softening performance [8,26].

The performance of the prepared formulations as fabric softeners was evaluated via panel tests as described in section 2.3. The interaction of the formulations with fabric was subsequently investigated using two different experimental approaches in order to understand the reasons underlying the observed differences in softening performance.

In the first experimental approach, the batch deposition of OMS droplets on fabric was characterized using the scheme described in section 2.5.1. This experiment provides insights into the kinetics of deposition of OMS droplets on fabric as well as the maximum quantity of quat that could be deposited on fabric for each formulation. The second experimental approach follows a chromatographic approach, as described in section 2.5.2, to obtain to more advanced insights into the deposition process. This experiment allows us to investigate not only the deposition rate of OMS droplets on fabric in different formulations but also the transport characteristics of OMS droplets through the porous fabric.

Our findings indicate that as the OMS droplets in the classical emulsion are large, they cannot transport freely through textile pores, and hence most of the OMS is deposited on the external surfaces of fabric. In the case of microemulsions, where droplet sizes are much smaller than pore sizes, it is droplet surface charge which is found to be

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