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Short communication

Inverse toroidal vesicles: precursors of tubules in sorbitan monostearate organogels

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Abstract

Sorbitan monostearate organogels are opaque, thermoreversible semi-solids whose microstructure consists of surfactant tubules dispersed in the organic continuous phase. Inverse toroidal vesicles are the precursors of the surfactant tubules. The gelation process was observed as an isotropic sol phase of sorbitan monostearate in isopropyl myristate was cooled using hot-stage light microscopy. At the gelation temperature, inverse toroidal vesicular structures were seen to grow in the organic phase. These toroids are thought to be analogous to other well-known vesicles, liposomes and niosomes, except for their toroidal (rather than spherical) shape and their inverse nature. They are rather short-lived structures: on further cooling of the sol phase, tubules form in the organic medium: it is speculated that the toroids elongate into tubular shapes or split into rod-shaped segments.

Keywords: Organogel; Inverse vesicles; Toroids

We have previously reported the gelation of a number of organic solvents, such as hexadecane, isopropyl myristate and vegetable oils, by the non-ionic surfactant sorbitan monostearate (Yoshioka and Florence, 1994; Murdan et al.,

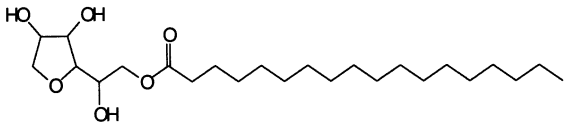

Fig. 1. Molecular structure of the sorbitan monostearate organogelator.

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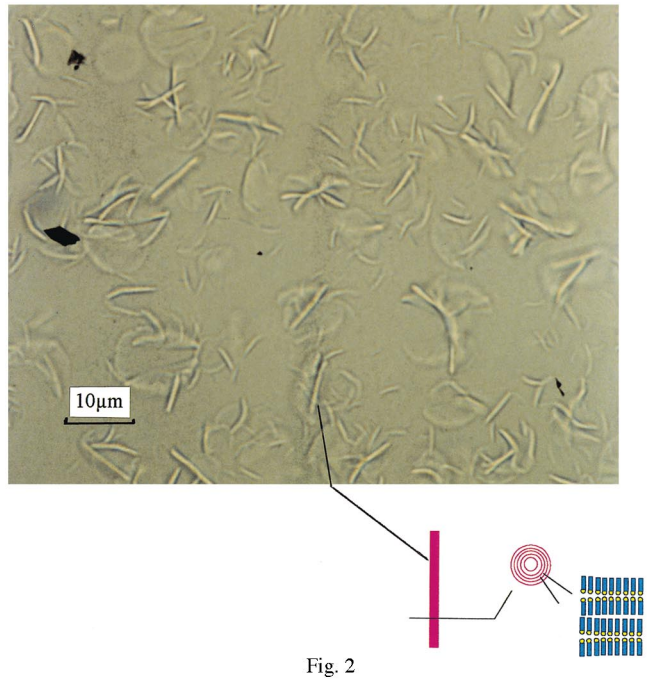

Fig. 2

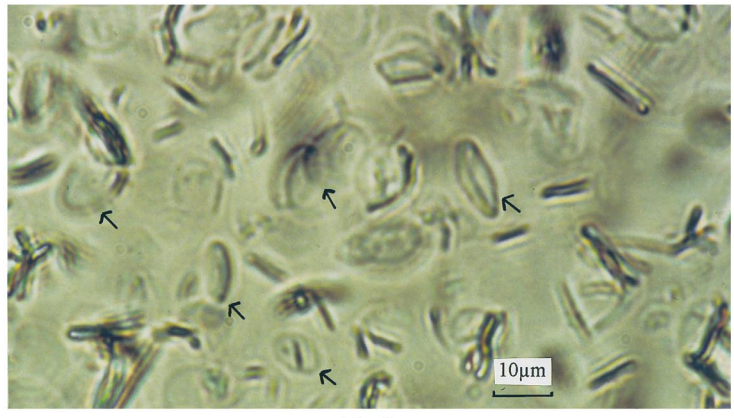

Fig. 3

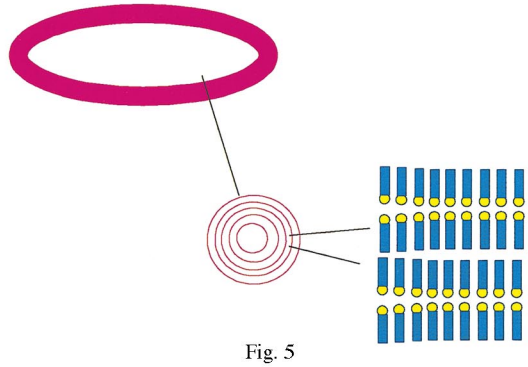

Fig. 5

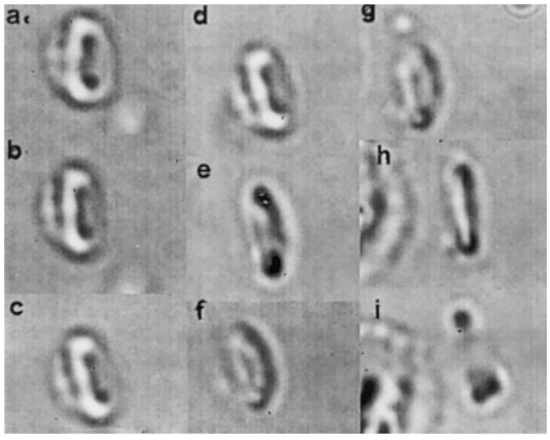

Fig. 4. One toroidal vesicle was photographed in rapid succession as it rotated about its axis. These pictures confirm the toroidal shape of the inverse vesicles.

1996) which is a small (MW 431), hydrophobic (HLB 4.1) molecule (Fig. 1). The organogels are prepared by dissolving/dispersing the sorbitan monostearate (10%w/v) as gelator in the organic solvent at 60°C and allowing the resulting solution/suspension to cool. Cooling the sol phase causes a reduction in the solubility of the gelator with consequent lowered solvent–surfactant affinities. As a result, the surfactant molecules self-assemble into aggregates, which interact forming a 3-dimensional network which immobilises the solvent and thus causes gelation. The resulting gels are opaque, thermoreversible semi-solids, with a smooth 'silky' feel while their microstructures (as revealed by light microscopy) consist of tubular aggregates dispersed in the fluid phase (Fig. 2). The tubules are believed to be composed of surfactant molecules arranged in inverse bilayers, as illustrated in Fig. 2.

In an attempt to understand the sol to gel transformation, we have observed using hot-stage microscopy the gelation process as a sorbitan monostearate/isopropyl myristate suspension was cooled from 60°C to room temperature. As the sol phase (transparent, isotropic phase) was cooled, inverse toroidal vesicular structures (Fig. 3) are formed at the gelation temperature (44–41°C). The toroids are thought to be analogous to other well-known vesicles, liposomes and niosomes, except for their toroidal (rather than spherical) shape and their inverse nature. At the gelation temperature, the toroids rotate rapidly in the organic medium. Using a high-speed camera, one of these toroids was photographed in rapid succession as it rotated about its axis and the toroidal shape was confirmed (Fig. 4). It is possible that the toroidal shape is favoured over the spherical ones upon surfactant self-assembly due to the lower curvatures required in the toroidal aggregates. The ultrastructure of the toroids is believed to be similar to that of the tubular aggregates and to consist of a number of inverse surfactant bilayers, as shown in Fig. 5.

The toroidal vesicles are rather short-lived structures and only exist at the sol to gel transition, in this case, from 44–41°C. Upon further cooling of the formulation, tubules are found in the organic medium. It seems that the toroidal vesicles are the precursors of the tubules. The toroids may elongate into tubular shapes on cooling, or they may split into rod-shaped cylindrical tubular segments. This transition has not been observed so far, possibly due to the rapidity of the events. Further cooling of the formulation results in the newly formed tubules joining with one another and forming a 3-dimensional network which immobilises the solvent, resulting in the gel.

References

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Fig. 2. Light micrograph of a sorbitan monostearate–hexadecane gel at room temperature. The gel microstructure consists of surfactant tubular aggregates dispersed in the organic medium. The tubules are believed to consist of a number of inverse surfactant bilayers.

Fig. 3. A photomicrograph of an isopropyl myristate formulation at the transition temperature between sol and gel phases. A number of toroidal vesicular structures can be seen.

Fig. 5. The toroid, like the resultant tubular aggregate, is thought to consist of a number of inverted surfactant bilayers.