Colloids and Surfaces A: Physicochem. Eng. Aspects 502 (2016) 19–25

Contents lists available at ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: www.elsevier.com/locate/colsurfa

Physical characterization and antimicrobial evaluation of glycerol monolaurate organogels

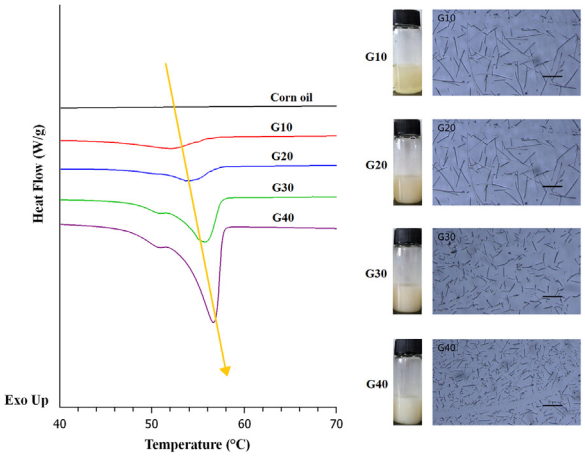
Haoxue Zheng[a], Lingli Deng[a], Fei Que[b], Fengqin Feng[a], Hui Zhang[a,*]

[a] Zhejiang University, College of Biosystems Engineering and Food Science, Fuli Institute of Food Science, Zhejiang Key Laboratory for Agro-Food Processing, Zhejiang R&D Center for Food Technology and Equipment, Hangzhou 310058, China
[b] Department of Applied Engineering, Zhejiang Economic and Trade Polytechinc, Hangzhou 310018, China

HIGHLIGHTS

- Organogels based on glycerol monolaurate and corn oil were prepared.
- More compact gel networks were induced by increasing gelator concentration.
- The prepared organogels showed strong antimicrobial activity against bacteria.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 15 January 2016
Received in revised form 20 April 2016
Accepted 1 May 2016
Available online 3 May 2016

Keywords:
Organogel
Glycerol monolaurate
Physical characterization
Antimicrobial activity

ABSTRACT

Organogels have been prepared with corn oil as organic solvent and glycerol monolaurate (GML) as organogelator for antimicrobial purpose. The critical gelation concentration (CGC) was found to be 3% (w/w) by the inverted tube method. The physical properties of the organogels containing 3%, 10%, 20%, 30%, 40% GML were studied by light microscopy, differential scanning calorimetry, and rheological analysis. The results showed that with the increasing organogelator concentration, the gels had a more compact three-dimensional network as the GML molecules crystallized and grew into a stronger backbone network as the gel skeleton, leading to higher thermal stability, higher resistance to deformations and lower spreadability. The antimicrobial assay using zone of inhibition indicated that the prepared organogels possessed strong antimicrobial activities against *Staphylococcus aureus* and *Escherichia coli*. The successful development of GML-based organogels suggests the potential application as new gelled capsules or spreadable products for controlled drug delivery.

1. Introduction

Recently, organogel-based formulations have been increasingly used in various industries, as they are easy to prepare and

* Corresponding author.
E-mail address: hubert0513@zju.edu.cn (H. Zhang).

http://dx.doi.org/10.1016/j.colsurfa.2016.05.001
0927-7757/

have inherent long-term stability [1,2]. Organogels are composed of organogelators and organic solvents. The gelator molecules self-assemble by physical or chemical interactions, forming a three-dimensional network which immobilizes the continuous solvent phase [3–5]. Organogels may be categorized as either fluid matrix or solid matrix organogels [6]. The properties of organogels have been found to be dependent on the composition of gelators and solvents used [7]. Commonly used organogelators include lecithin, sterols, cholesteryl anthraquinone derivatives, and fatty acid esters [8].

To date, a limited number of organogels with different organogelators and organic solvents have been developed for antimicrobial purpose. Organogels based on castor oil and sorbitan monopalmitate (Span 40) showed strong intramolecular/intermolecular hydrogen bonding amongst the gel components, and exhibited a shear thinning behavior as a topical formulation for controlled drug delivery [9]. Emulsion gels prepared by lanolin and Span 80 for topical drug delivery showed absence of lanolin-drug chemical interactions, non-Newtonian and thixotropic flow behavior, sufficient spreadability and biocompatibility, and good antimicrobial efficacy against *Bacillus subtilis* [10]. Sunflower oil and lecithin based organogels developed for controlled drug delivery revealed the presence of spherical reverse micellar structures, an increase in the intermolecular hydrogen bonding among the gel components and crystallinity upon incorporation of metronidazole into the organogels, and showed a good antimicrobial activity against *B. subtilis* [11]. Organogels based on sunflower oil and Span 40 exhibited the clusters of rod-shaped tubules responsible for the formation of network in the presence of intermolecular hydrogen bonding, showing a pseudoplastic flow behavior, and the ciprofloxacin-loaded gels possessed excellent antimicrobial properties against *B. subtilis* [12].

Glycerol monolaurate (GML), as a nonionic surfactant with a HLB value of approximately 3.5 to promote water-in-oil emulsions, is a generally recognized as safe (GRAS) food additive with a wide range of antimicrobial activities [13]. Recently, some food-grade emulsions based on glycerol monolaurate have been established by our group for antimicrobial purpose [14].

In the present work, attempts were made to develop food-grade organogels with GML as an organogelator and corn oil as an organic solvent for potential antimicrobial applications. The physical properties of the prepared organogels were investigated by light microscopy, differential scanning calorimetry, and rheological analysis. The antimicrobial activities were studied against *Staphylococcus aureus* and *Escherichia coli*.

2. Materials and methods

2.1. Materials

2.1.1. Chemicals

Glycerol monolaurate (GML) (purity > 90%) was provided from Hangzhou Kangyuan Food Science and Technology Co., Ltd., China. Corn oil was purchased from a local market. Double distilled water was used throughout all experiments.

2.1.2. Microorganisms

Microbial cultures of *S. aureus* (CMCC 26003) and *E. coli* (ATCC 25922) were obtained from Qingdao Hope Bio-Technology Co., Ltd, and preserved at the Department of Food Science and Nutrition, Zhejiang University. The strains were cultured in NA (Nutrient Agar, Hangzhou Microbiological Agents Co., Ltd, China) broth at pH 7.0 and transferred every 20–24 h with incubation at 37 °C.

2.2. Preparation of organogels

Accurately weighted GML was dissolved in a specified amount of corn oil. The mixture was constantly stirred at 70 °C, and then cooled to room temperature. The concentration of GML was varied from 0% (w/w) to 40% (w/w) to find the critical gelation concentration (CGC). The phenomenon of gelation was confirmed by the inverted tube method [12].

2.3. Light microscopy

The organogels were observed under light microscope (Eclipse E600w Pol, Nikon, Japan) with the attached camera (QImaging MicroPublisher 3.3 RTV, Canada), and analyzed using image processing software (Linksys32). The magnification of microscope was 100× for observation.

2.4. Rheological measurements

The rheological measurements of organogels were performed in a rheometer (MCR302, Anton Paar, Austria). The gels were analyzed using parallel-plate (PP-25) geometry with a gap size of 1 mm due to their highly viscous nature. Viscoelasticity was investigated by dynamic oscillatory rheometry. The amplitude sweep of the storage modulus (G′) and loss modulus (G″) was collected for strain values from 0.001% to 100%. The composition dependence of the storage modulus (G′) and loss modulus (G″) for organogels were measured by applying a frequency sweep varied from 0.1 Hz to 10 Hz at a constant 0.01% strain within the linear viscoelastic region. For temperature dependence test, organogels were investigated with a heating rate of 1 °C/min from 10 °C to 70 °C with a frequency of 1 Hz.

2.5. Differential scanning calorimetry

The thermal properties of organogels were studied using differential scanning calorimeter (DSC1, Mettler Toledo, Switzerland). The DSC was purged with dry nitrogen gas at a flow rate of 30 mL/min. Calibration was done with a sapphire disk for baseline and with indium for temperature. Accurately weighed samples (10–15 mg) were encapsulated in aluminum (Al) crucibles and hermetically sealed with a pierced Al lid. The gels were analyzed at a heating rate of 1 °C/min from 20 °C to 100 °C.

2.6. Spreadability

The spreadability values of organogels were determined as reported previously [10]. A sample of 0.2 g gel was pressed between two glass slides, having equal weight and area. The initial diameter (D1) was recorded. Thereafter, a known weight of 200 g and 500 g was put over the upper slide. The final diameter (D2) was recorded after a period of 60 s. The spreadability of the samples was calculated using the following equation:

$$\text{Spreadability} = \frac{D2}{D1}$$

2.7. Antimicrobial evaluation

The gram-positive *S. aureus* and the gram-negative *E. coli* bacteria were used as model microbes to analyze the antimicrobial efficacy of the organogels. About 100 μL of the inoculums (containing approximately 10^6 cfu/mL) was spread on the solidified NA plate with a sterilized spreader. A known weight of 0.2 g sample was accommodated. The plates were then incubated at 37 °C for 24 h.