

STABILITY AND PHYSICAL PROPERTIES OF DIHYDROXYSTEARIC ACID BASED ORGANOGELS IN DIFFERENT TYPES OF VEGETABLE OIL

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ABSTRACT

Dihydroxystearic acid (DHSA) was obtained from epoxidation and hydroxylation of oleic acid from palm oil. Unlike monohydroxystearic acid, only a few publications described DHSA in the gelation of oils. This study aimed to investigate the stability and physical properties of DHSA organogels in different vegetable oils. Five vegetable oils; refined, bleached and deodourised palm olein (RBDPOo); refined, bleached and deodourised super olein (RBDSPo); refined, bleached and deodourised palm kernel olein (RBDPKOo); medium-chain triglyceride (MCT); and soybean oil (SBO) were used to prepare DHSA organogels. The stability and critical gel concentration (CGC) of organogels were determined at concentrations of DHSA in the range of 0.2%-20.0% (w/w). DHSA organogels were characterised using a polarised light microscope, texture analyser and rheometer. DHSA organogel with RBDSPo showed a more stable gel with the lowest CGC value at 9.9% than other oils. In microscopic characterisation, DHSA organogel with RBDPOo showed a high density of small needle-like crystals. The hardness and rheological measurements revealed that DHSA organogels with RBDPOo and RBDSPo showed the highest elasticity, which is more than 10 000 Pa compared to other oils. However, all DHSA organogels were classified as soft gels and have the potential to be used in cosmetic products.

Keywords: dihydroxystearic acid, organogel, vegetable oils.

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INTRODUCTION

Organogels are defined as semisolid viscoelastic systems having gelator and oil or organic liquid, immobilised within the space of a three-dimensional network structure of gelator molecules. Gelation can be formed with two different interactions, *i.e.*, between organic liquid-gelator and gelator-gelator interactions. Non-covalent interactions within molecular gels include hydrogen bonding, π - π stacking, and van der Waals interactions (Lan and Rogers, 2015; Silva *et al.*, 2021). Gelling ability is related to the balance

between soluble and insoluble fractions of gelator in oil. Gelator must be relatively insoluble to self-assemble into anisotropic structures. Instead, it must have a soluble fraction to interact with the oil moieties (Co and Marangoni, 2012). The organogels formulations have several advantages, *i.e.*, long shelf-life and thermo-reversible behaviour. In addition, the preparation of organogels involves a simple process by dissolving a gelator in an organic liquid at sufficiently high temperatures and then cooling down the solution until there is no flow for elongated periods, normally between 24 hr and several days to allow the development of gel (Flöter *et al.*, 2021).

Mineral and vegetable oils are widely used as solvent phases in cosmetic organogels. The low cost and properties of mineral oil, which is transparent, thermostable and safe have made it

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a popular solvent used in cosmetics (Martinez *et al.*, 2019). Recently, researchers have shown an increased interest in developing edible organogels in several areas, such as in the pharmaceuticals and cosmetics industries to achieve 'green' claims and eco-friendly alternatives to replace mineral oil. The edible organogel is of interest because it has unique physical and functional properties as well as is safe for human use. In the edible organogel formulations, vegetable oils are used as the organic liquid, and gelators can be chosen from different groups of gelators, *i.e.*, crystalline waxes, low molecular weight gelators (LMWGs), polymeric or inorganic gelators. Generally, vegetable oils are composed of triglycerides containing saturated, monounsaturated, polyunsaturated fatty acids and several minor components. In cosmetics, especially lip care products, edible organogels play an important role in replacing petrolatum. Less purified petrolatum may contain impurities, such as polycyclic aromatic hydrocarbons, which has been reported as a potentially carcinogenic substance (Chen *et al.*, 2020). The safety concern of using petrolatum on human health has driven researchers to explore other renewable alternatives to replace this material (Stortz and Marangoni, 2014).

Several attempts have been made to explore the role of oil and the chemical structure of LMWGs that influences the formation of organogels. One of the best LMWGs that was thoroughly studied for their ability to form organogels is 12-hydroxystearic acid (HSA) (Asaro *et al.*, 2020). The HSA is a fatty acid derivative of castor oil, and it was chosen due to its simple structure, inexpensive and superior gelation capacity. It was reported that the gel formation using HSA involved interactions between carboxylic acid groups and the hydroxyl group, which result in the formation of multiple hydrogen-bond (H-bond) sequences (Zhang and Weiss, 2016).

Another potential LMWG is 9,10-dihydroxystearic acid (DHSA). The presence of carboxyl and hydroxyl groups of the DHSA is similar to HSA except for one additional hydroxyl group adjacent to each other. It is derived by converting the unsaturation of oleic acid to an epoxide ring via peracetic acid or performic acid routes, followed by hydrolysis with hydrogen donors, such as water (Koay *et al.*, 2011). The two hydroxyl groups are located at position-9 and position-10 of an 18-carbon aliphatic chain

(Figure 1). The *in vitro* and *in vivo* safety evaluation studies revealed that the DHSA is safe to be applied on skin and areas around the eyes. Thus, it can potentially be used as an ingredient in cosmetic formulations. DHSA was reported to be effective in coating pigments for colour cosmetic products. In addition, the use of DHSA has been further studied in sodium soap and transparent soap. The DHSA soap exhibited good corrosion inhibition as well as showed better foamability and detergency than stearic acid soap. Moreover, DHSA incorporated in transparent soap formulation helps to improve the transparency of transparent soap (Ismail *et al.*, 2015). Although extensive research has been carried out on the application of DHSA in cosmetics and personal care products, there has been little discussion about DHSA for the gelation of oils. Zhang and Weiss (2016) reported that DHSA was capable of gelling sunflower and lavender oils but at 2% to 8% concentration.

It is important to notice that there are some challenges of organogels in cosmetics, such as difficulty to predict gelation process between gelator and solvent as well as the stability of the organogel (Martinez *et al.*, 2019). Therefore, this article aimed to study the effect of DHSA on the stability and physical properties of DHSA-based organogels in different vegetable oils, whereby, this basic organogel formulation can be used to formulate lip care products. For this purpose, refined, bleached and deodourised palm olein (RBDPOo); refined, bleached and deodourised super olein (RBDSPo); refined, bleached and deodourised palm kernel olein (RBDPKOo); medium-chain triglyceride (MCT); and soybean oil (SBO) were chosen due to their differences in the compositions of fatty acids. The stability, critical gel concentration (CGC) and physical properties, such as crystal morphology, hardness, and rheology of DHSA-based organogels were studied.

MATERIALS AND METHODS

Materials

DHSA (appearance: off-white powder, purity: 95%, hydroxyl value: 299 mg KOH g⁻¹, melting point: 91°C and moisture content: 0.6%) and RBDSPo were obtained from the Malaysian Palm Oil Board.

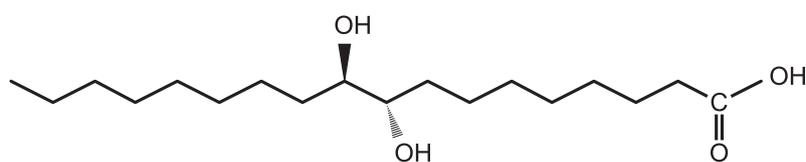


Figure 1. Structural representation of 9,10-dihydroxystearic acid.

RBDPOo and SBO were purchased from Yee Lee Edible Oils Sdn. Bhd., and Kempas Edible Oil Sdn. Bhd., respectively. RBDPKOo was purchased from Southern Edible Oil Industries (M) Sdn. Bhd., Malaysia. MCT or caprylic/capric triglyceride 60/40 was purchased from InterMed Ester Sdn. Bhd., Malaysia. The fatty acid compositions were determined via gas chromatography (GC) analysis.

Determination of Fatty Acid Compositions of Oils

The fatty acid compositions of the oils were determined by GC analysis using Agilent 7890B equipped with a 7693 Series autoinjector from Agilent Technologies, USA. Chromatography separation was achieved with the aid of the HP88 column (60 m x 0.25 mm x 0.20 μm). The GC was operated in the split injection mode using a 100:1 split ratio while maintaining the injector temperature of 250°C. The oil sample was prepared according to a standard method, AOCS Ce 2-66 (AOCS, 2012). Then, 1 μL of the mixture was injected into the GC column with an initial oven temperature of 150°C. The oven temperature was programmed at 3°C min^{-1} until 210°C. The flow rate of the carrier gas helium was 0.8 mL min^{-1} . The temperature of the flame ionisation detector was set at 250°C.

Determination of Viscosity of Oil

The viscosity of oils was measured using a rheometer Physica MCR300 from Anton Paar, Germany. The measurement was performed by a concentric cylinder geometry (CC17) at 25°C. The shear rate was increased step-wise from 1 to 100 s^{-1} .

Preparation of the Organogels

Organogels were prepared in sample tubes (15 cm height x 1.6 cm diameter) with screw caps. The mixtures containing an oil phase (5.00 g), *i.e.*, RBDPOo, RBDSPo, RBDPKOo, MCT or SBO, respectively. DHSa (0.01-1.25 g) was heated to 95°C \pm 2°C and stirred vigorously for 3 min using a vortex. Afterwards, the samples were allowed to cool down to room temperature (25°C) for 24 hr to form gels.

Qualitative Assessment of Gelation Behaviour

The equilibrated organogel in sample tubes were inverted, and the self-standing ability of the samples was assessed visually. The gelation behaviour of organogel samples was monitored up to 12 weeks at 25°C (Burkhardt *et al.*, 2009). The appearance of the samples can be described as two-phase (fluid/gel-like), a mixture of gel and liquid (gel + liquid), and stable gel. Samples that

immediately flowed were categorised as a liquid. Samples in which the gel and liquid were clearly separated from the system and slowly flowed were categorised as two-phase. Lastly, samples that did not flow were categorised as a stable gel. However, gels that were unstable upon storage (whereby oil come out from the system) were categorised as gel + liquid. CGC was determined as the lowest concentration of DHSa at which a stable gel was formed at room temperature.

Assessment of Organogel Crystal Morphology with Respect to Different Oils

The crystalline structures of the organogel samples were examined at 25°C under a polarised light microscope via the Olympus System Microscope Model BX53 (Olympus Corporation, Japan). A small sample containing crystals was placed on a glass microscope slide and gently covered with a glass coverslip. Digital images of specimens were acquired using an Olympus microscope digital camera model Olympus DP73 (Olympus Corporation, Japan).

Determination of Organogel Hardness

The hardness of organogel samples was evaluated according to the method described by Onacik Gur *et al.* (2017). The compression measurement was conducted using a texture analyser TA.XT plus (Stable Micro Systems, United Kingdom). A cylindrical probe (P/0.5R) with a diameter of 12.7 mm was used for the measurement. Each organogel sample (approximately 30 mm height) was conditioned in a 20 mL vial. The internal diameter and depth of the vial were 28 mm and 60 mm. The probe was dipped 5 mm into an organogel sample at 1 mm s^{-1} . The test was carried out at room temperature. The measurements were performed in triplicates.

Characterisation of Rheological Property of Organogel

The rheological properties of organogel samples were performed using a rheometer, MCR300 from Anton Paar GmbH, Germany. The measurement was performed using a parallel-plate geometry of 25 mm wide with a 1 mm gap at 25°C. The organogel samples were handled gently to avoid structural damage. The viscoelastic properties of organogel samples were investigated via oscillation tests. First, amplitude sweep was carried out with an increasing strain from 0.01% to 100.00% at a fixed angular frequency of 10 rad s^{-1} to determine the linear viscoelastic region (LVER) of the organogel samples. Then, a frequency sweep was determined from 0.01 to 10.00 Hz at a constant

strain of 0.50%. Storage modulus (G') and loss modulus (G'') were evaluated. The flow properties were characterised using the shear rate range of 0.1 to 100.0 s^{-1} to evaluate the thixotropic behaviours of the organogel samples.

Statistical Analysis

The data obtained from the above studies were analysed statistically using the analysis of variance (ANOVA) single factor and the Student's t-Test. The differences were considered significant if the probability was $p < 0.05$.

RESULTS AND DISCUSSION

Fatty Acid Compositions of Oils

The compositions of fatty acid and viscosity of the RBDPOo, RBDSP0o, SBO, RBDPKOo and MCT oils, are presented in *Table 1*. It was observed that RBDPOo and RBDSP0o contained an almost equal amount of saturated ($C_{16:0}$) and unsaturated ($C_{18:1}$) fatty acids. SBO contained the highest amount of unsaturated fatty acid ($C_{18:2}$), RBDPKOo contained the highest amount of saturated fatty acid (C_{12}) and

MCT contained medium chain fatty acids C_8 and C_{10} . The viscosity of oil was determined at the plateau value from 30 to 100 shear rate (s^{-1}), and the result follows the reducing viscosity order: RBDPOo = RBDSP0o > SBO > RBDPKOo > MCT.

Stability of DHSA Organogels in Various Oils

The relationship between fatty acid compositions and the degree of gelation for DHSA organogels in various oils is depicted in *Figure 2*. Three phases were identified according to the visual appearance of the samples: two-phase, gel + liquid and gel. The gel + liquid was classified as partial syneresis. Syneresis occurred when gels could not hold the oil and resulted in oil flowing out from the system. It is due to the crystal's structure which formed large aggregates and agglomerates into thick bundles (Daman Huri *et al.*, 2013). Our study showed that DHSA could form gels in all studied oils. However, different amounts of DHSA are required to initiate the formation of organogel on different oil. A minimum amount of DHSA to form organogels was observed in RBDSP0o and RBDPOo, followed by SBO, RBDPKOo and MCT. However, syneresis was observed after seven days of storage at 25°C and the lowest CGC of

TABLE 1. FATTY ACID COMPOSITIONS AND VISCOSITY OF OILS

Fatty acids (%)	Oil				
	RBDPOo	RBDSP0o	SBO	RBDPKOo	MCT
$C_{6:0}$	n.d.	n.d.	n.d.	0.2	n.d.
$C_{8:0}$	n.d.	n.d.	n.d.	3.4	54.6
$C_{10:0}$	n.d.	n.d.	n.d.	3.3	45.2
$C_{12:0}$	0.3	0.3	n.d.	42.0	0.2
$C_{14:0}$	0.9	1.0	0.1	13.6	n.d.
$C_{15:0}$	n.d.	n.d.	n.d.	n.d.	n.d.
$C_{16:0}$	38.6	37.6	10.8	9.1	n.d.
$C_{16:1}$	0.2	0.2	0.1	n.d.	n.d.
$C_{17:0}$	0.1	0.1	0.1	n.d.	n.d.
$C_{17:1}$	n.d.	n.d.	0.1	n.d.	n.d.
$C_{18:0}$	4.1	3.9	4.9	2.7	n.d.
$C_{18:1}$	44.2	45.5	24.2	22.0	n.d.
$C_{18:2}$	10.7	10.5	52.3	3.3	n.d.
$C_{18:3}$	0.2	0.2	6.4	n.d.	n.d.
$C_{20:0}$	0.4	0.4	0.4	0.2	n.d.
$C_{20:1}$	0.2	0.2	0.2	0.1	n.d.
$C_{22:0}$	0.1	0.1	0.4	n.d.	n.d.
Σ SAFA	44.6 ± 0.1	43.4 ± 0.1	16.7 ± 0.0	74.4 ± 0.1	100 ± 0.1
Σ MUFA	44.6 ± 0.1	45.9 ± 0.1	24.6 ± 0.1	22.2 ± 0.1	n.d.
Σ PUFA	10.9 ± 0.1	10.7 ± 0.1	58.6 ± 0.1	3.3 ± 0.1	n.d.
Viscosity (mPa.s)	69.0 ± 0.0 ^a	69.0 ± 0.0 ^a	66.0 ± 0.0 ^b	51.0 ± 0.0 ^c	23.0 ± 0.0 ^d

Note: SAFA - saturated fatty acids; MUFA - monounsaturated fatty acids; PUFA - polyunsaturated fatty acids; n.d. - not detected; means with different letters in the same row are significantly different ($p < 0.05$).

DHSA was determined in RBDSP0o followed with RBDPOo, SBO, RBDPKOo and MCT at CGC of 9.90%, 10.70%, 13.00%, 16.00% and 17.40%, respectively. This behaviour could be correlated with the hypothesis that oil polarity and degree of unsaturation provide some effect on its ability to form H-bonds with DHSA molecules (Gravelle *et al.*, 2016). It was found that MUFA helps in stabilising the organogel structure (Rocha-Amador *et al.*, 2014). According to a report by Burkhardt *et al.* (2009), HSA formed gel in MCT at CGC of 0.75%. Thus, HSA is a more efficient gelator than DHSA. The ability of HSA to form self-assembled fibrillar structures occurs most efficiently when the position of the carboxylic headgroup forms cyclic dimers between adjacent HSA molecules and the hydroxyl group at C₁₂ interconnected with neighbouring molecules leading to unidirectional hydrogen bonding along the fibre axis (Fameau and Rogers, 2020). In contrast, it was anticipated that DHSA can utilise only one hydroxyl group per molecule to create intermolecular hydroxyl H-bonds per molecule and the other hydroxyl group for intramolecular H-bonding interactions. Due to this interaction, fibrillar structures could not be formed and a higher amount of DHSA is required for the stabilisation of organogel (Zhang *et al.*, 2015). All studied oils contain MUFA except MCT oil. The presence of MUFA in RBDSP0o, RBDPOo, RBDPKOo and SBO helps to stabilise the DHSA based organogel, possibly through the formation of H-bonds between DHSA and MUFA. The RBDSP0o has the highest MUFA, thus, showing a stable gel with the lowest CGC value than the other oils.

Effect of DHSA on Organogel Crystal Morphology

According to a study by Cerqueira *et al.* (2017), there are three steps involved in the process to form an organogel: Crystal nucleation, crystalline

branching and crystal growth. Prior to gel formation (at low gelator concentrations), a two-dimensional structure was formed, which led to nucleation and growth of the crystal surface. Then, with increasing gelator concentration, the process continued until a three-dimensional structure was formed, in which the crystal cross-linked or entangled to trap or immobilise the oil molecules. *Figure 3* shows the crystal morphologies of the organogels at 8% (w/w) as well as 16% (w/w) DHSA in RBDSP0o, RBDPOo, SBO, RBDPKOo and MCT, respectively, after 24 hr at room temperature. It was observed that DHSA organogels with RBDSP0o, RBDPOo and SBO have similar crystalline structures, which are in the form of needles. However, DHSA organogels with RBDPKOo and MCT resulted in platelet crystalline structures. This showed that the use of different oils affects the type of crystal formation. Increasing the DHSA concentration, the crystal structure network was more compact, which can entrap more oil and stabilise the organogels.

Effect of DHSA on Hardness of Organogel

The addition of DHSA as a gelator in vegetable oils is to give structure to the gel and immobilise the oils at room temperature. *Figure 4* shows the hardness of organogels at 8% (w/w) and 16% (w/w) DHSA in different oils viscosity, in which the high hardness values indicate a rigid organogel structure. The results showed that the hardness of DHSA organogels was significantly different in the studied oils. It was observed that at 16% (w/w) DHSA could form stable gels with all oils, and its hardness can be determined in all samples. However, 8% (w/w) DHSA formed stable gel in all oils except RBDPKOo and MCT. The maximum hardness achieved for organogel at 16% (w/w) DHSA was more than eight times higher than organogels at 8% (w/w) DHSA. It was observed that increasing SAFA content, especially C₁₆ and

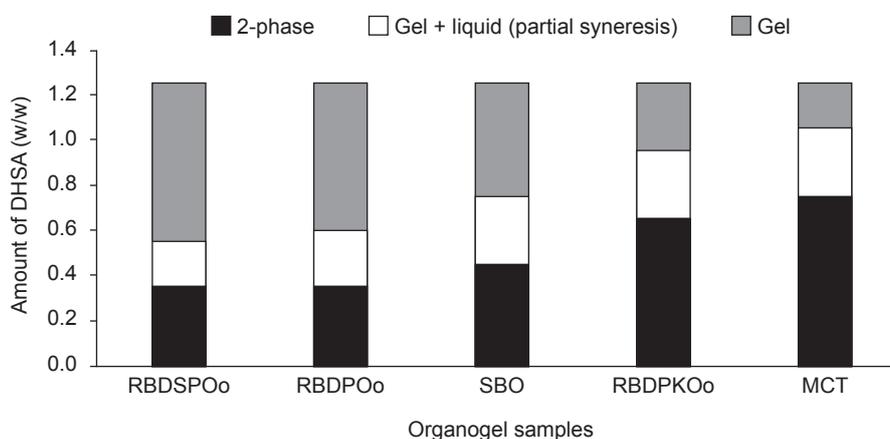


Figure 2. Gelation behaviour of organogels in various oils after 12 weeks at 25°C as a function of added DHSA.

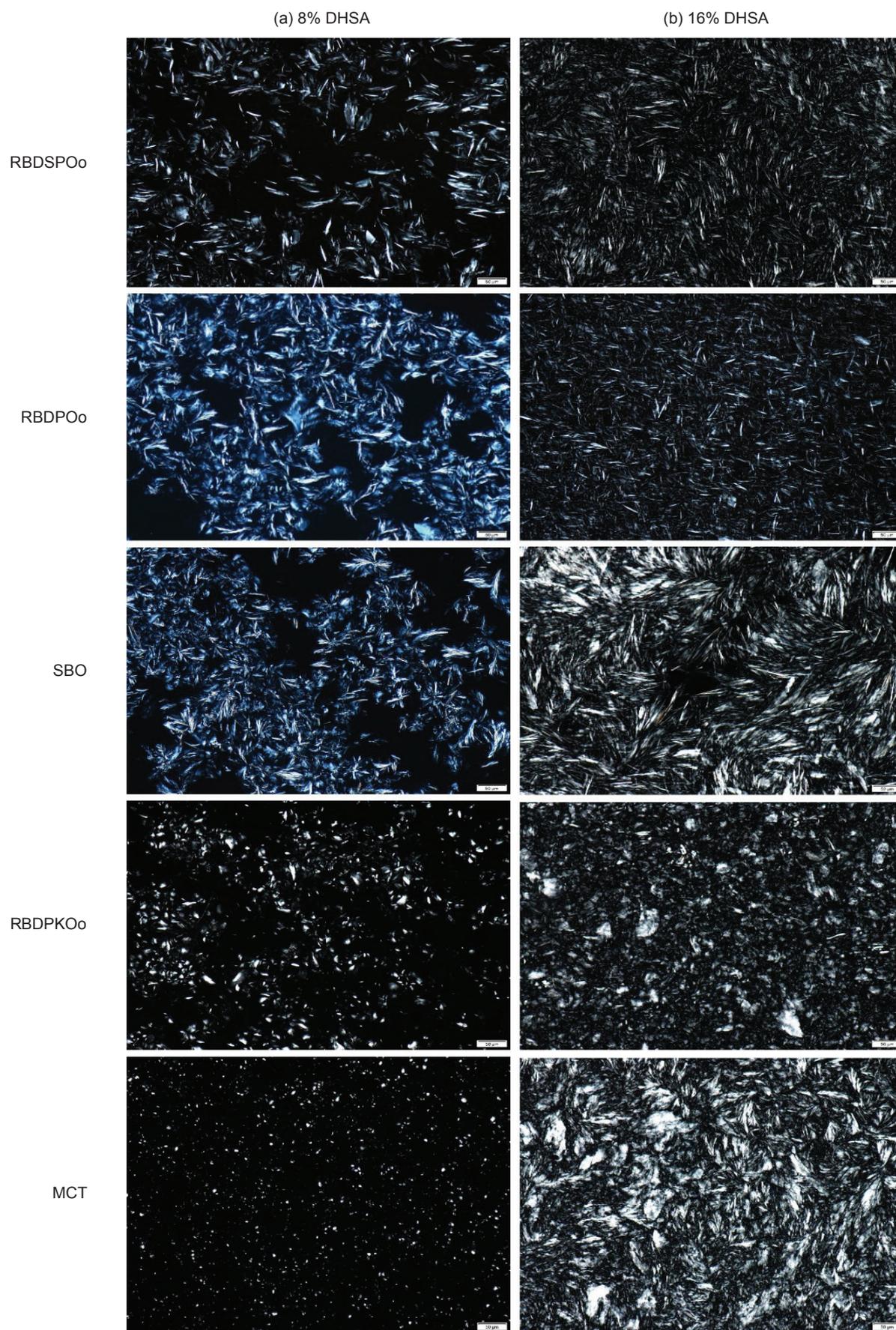


Figure 3. Polarised light microscopy images of organogels at (a) 8% (w/w) DHSA and (b) 16% (w/w) DHSA in oils i.e., RBDPOo, RBDSP0o, SBO, RBDPKOo and MCT, respectively. The images were analysed at room temperature under objective magnification 20x with a scale bar of 50 μm .

C_{18} in the oils (RBDPOo, RBDSPo and SBO) also helped to increase the hardness of organogels. In contrast, the SAFA content in RBDPKOo and MCT did not help to increase the organogel hardness as these two oils had a high content of C_{12} (42.0%) and C_8 (54.6%) with low melting points compared to C_{16} and C_{18} . It was reported by Knothe and Dunn (2009) that melting points of C_{18} , C_{16} , C_{12} and C_8 were 69.29°C, 62.20°C, 43.29°C and 15.41°C, respectively. These findings are in line with Pehlivanoglu *et al.* (2018) and Dassanayake *et al.* (2012), which reported that vegetable oils with a higher level of saturated fatty acids and high melting fatty acids produced hard organogels. In addition, it was also observed that oils with higher viscosities resulted in higher organogel hardness. These results agree with Valoppi *et al.* (2017) findings, which showed that there is a correlation between hardness and viscosity. Furthermore, the small needle-like crystal with dense distribution observed in DHSA organogel with RBDPOo has also contributed to the highest hardness of the organogel. In addition, the hardness of the organogels could also be affected by several other factors, such as thermal kinetics, the ratio of crystal and oil surfaces, as well as its solubility in oil (Dassanayake *et al.*, 2012; Onacik Gur *et al.*, 2017). Although DHSA organogels in MCT and RBDPKOo have the same crystal platelet structure, the crystalline structures of DHSA organogel in RBDPKOo were less compact, resulting in lower hardness values. Hardness is one of the essential criteria to determine the potential application of organogel in cosmetic products, especially lip products. The hardness for commercial solid lipstick was reported at least 0.29 N (Esposito and Kirilov, 2021) and commercial

lip balm was within 0.49-0.64 N (Azmin *et al.*, 2020). Thus, hardness values of all DHSA organogels with the studied oils except for DHSA organogel with SBO were considered acceptable to formulate lip care products.

Rheological Characterisation

The elastic properties such as storage modulus (G') and loss modulus (G'') of organogels containing 16% (w/w) DHSA in RBDPOo, RBDSPo, SBO, MCT and RBDPKOo were characterised using oscillating rheological experiments. The viscoelastic behaviour involves structural changes within the organogel, either solid-like (G') or liquid-like (G'') properties. Figure 5 shows the frequency sweep of DHSA organogels in various oils at a varied angular frequency and constant strain (0.5%). The results showed that all organogel samples have gel-like behaviour with $G' > G''$ over a broad angular frequency range. DHSA organogel containing RBDPOo and RBDSPo showed the highest elastic modulus compared to other oils. This showed that DHSA formed a better crystal network and gelation in RBDPOo and RBDSPo. However, DHSA organogel with RBDPKOo showed G' and G'' very close to each other at high angular frequency, indicating that this gel has liquid-like behaviour.

In order to determine the gel strength, a graph of $\tan \delta$ as a function of angular frequency for DHSA organogels in various oils was plotted (Figure 6). In general, organogels can be classified into three types, which are strong gels ($G''/G' \leq 0.1$), weak gels ($0.1 \leq G''/G' < 1$) and viscous sols ($G''/G' \geq 1$) (Tavernier *et al.*, 2017). All DHSA organogels

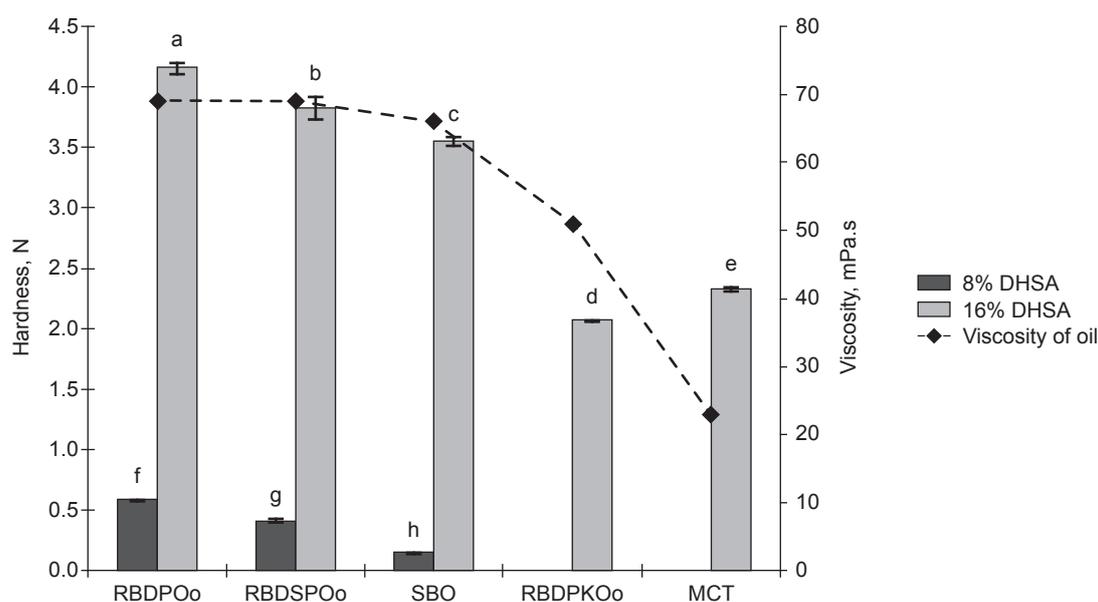


Figure 4. Hardness of organogel samples at 8% (w/w) and 16% (w/w) DHSA in different oils viscosity after seven days of storage at 25°C. Mean values with different letters are significantly different ($p < 0.05$).

were classified as weak gels ($0.1 \leq G''/G' < 1$). At high angular frequency, $\tan \delta$ or G''/G' of DHSA organogel with RBDPKOo is close to unity, which could not be considered as a gel. This is probably due to low oil viscosity. These results agree with Valoppi *et al.* (2017) findings, which showed a correlation between the rheology properties and oil viscosity. However, these correlations are not observed in MCT due to MCT's ability to make the dipole-dipole rotation, which affects the crystallisation behaviour (Cerqueira *et al.*, 2017). MCT containing 100% SAFA can form a more structured crystal than RBDPKOo, which contains mixtures of SAFA, MUFA and PUFA.

CONCLUSION

The main objective of this study was to evaluate the effect of DHSA as a gelling agent for vegetable oils on stability, crystal morphology, hardness and rheological properties. This study has shown that DHSA organogel with RBDSP0o showed a stable gel with the lowest CGC value, followed by RBDPOo, SBO, RBDPKOo and MCT. It was found that a higher percentage of MUFA in the oil contributed to a more stable gel. In addition, needle-shaped crystals and higher DHSA concentrations also help to entrap more oil and thus, improve organogel stability. Regarding texture

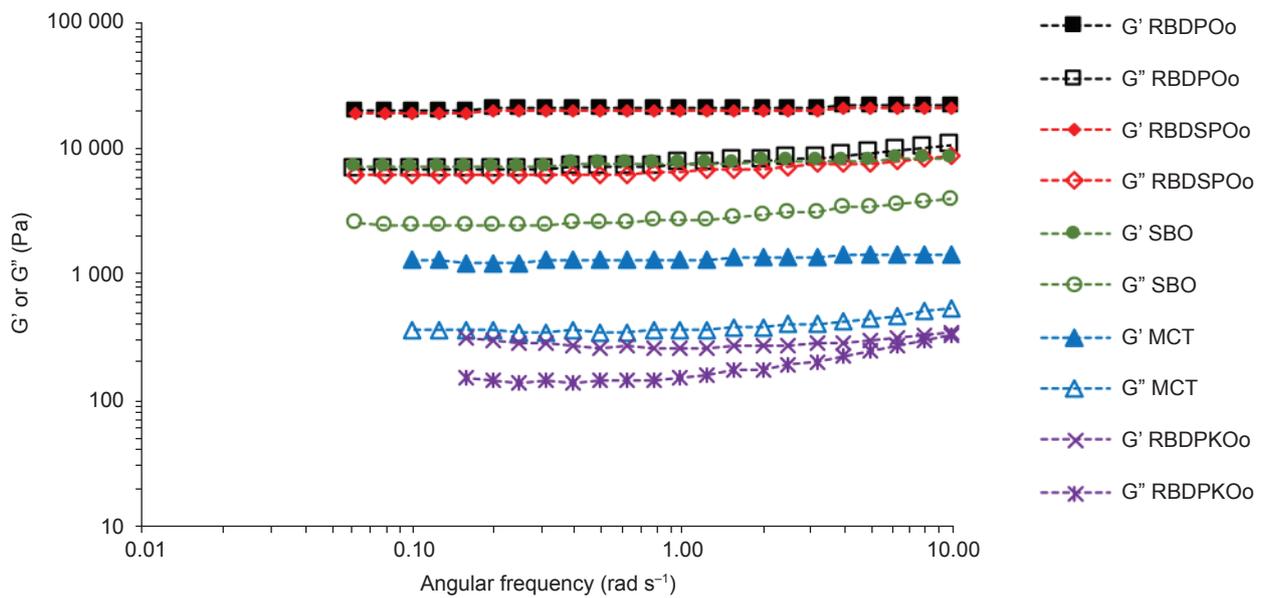


Figure 5. Frequency sweep of DHSA organogel samples as a function of angular frequency at 16% (w/w) DHSA in RBDPOo, RBDSP0o, SBO, MCT and RBDPKOo after seven days of storage at 25°C.

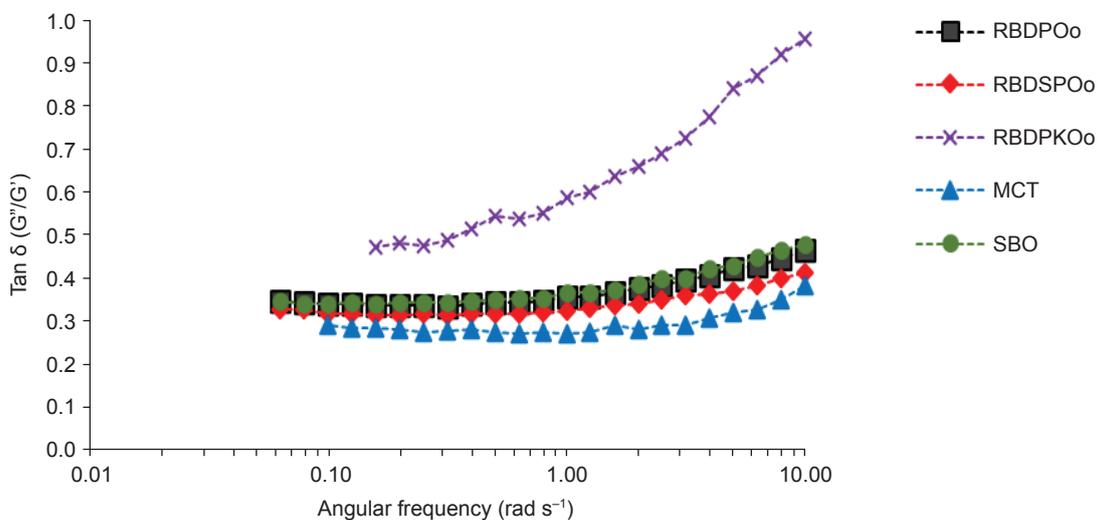


Figure 6. $\tan \delta$ value as a function of angular frequency for DHSA organogels at 16% (w/w) DHSA in RBDPOo, RBDSP0o, SBO, MCT and RBDPKOo after seven days of storage at 25°C.

characteristics, the hardness value of the DHSA organogels was improved with the formation of compact crystals and high melting fatty acids of the oil. Finally, it was found that elasticity and strength of the DHSA organogels were correlated with viscosity and degree of unsaturation of the oil as well as crystal network in the DHSA organogel. Considering the hardness value, DHSA organogel has the potential to be formulated in lip care products.

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