

Performance of Cellulose and Gum-based Oleogelators to Form **Oleogels in Various Oil Systems**

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ABSTRACT

A type of structured fats that is attracting attention as an alternative to transfat is oleogel. Oleogels can be formed from edible and non-edible vegetable oils, depending on the desired application. In this study, we used an emulsiontemplate approach with three oil systems: canola oil, sunflower oil, and castor oil, combined with hydroxypropyl methylcellulose (HPMC) and xanthan gum (XG) as oleogelators in specific ratios. Stable oleogels were formed when both oleogelators were present in the system. The oil binding capacity of the oleogel with respect to the amount of oil loss varied from less than 5% to more than 30%, depending on the saturated fatty acid composition of the oil and HPMC: XG ratio. Higher saturated fatty acid concentration in the oil strengthens the oleogel matrix and reduces the oil loss rate. High HPMC composition and low XG composition also reduce the oil binding ability of the oleogels. It was found that in oleogel systems, HPMC acts as an emulsifier and XG acts as a thickener, resulting in a lower-than-average XG concentration and a thinner gel. Although the structure and color of oleogels are very stable over 28 days of storage, the oxidation rate still needs to be minimized. Overall, oleogel heating as well as the presence of aqueous phase can accelerate the oxidation rate of the oleogel, which can shorten its shelf-life and limit its applications, especially in the food industry.

KEYWORDS

Vegetable oil Gum Oleogel Cellulose

INTRODUCTION

Cardiovascular diseases come as the leading cause of death, especially stroke and coronary heart disease. One of the main causes of cardiovascular disease is the intake of fats, mainly trans- or saturated-fat [1]. These trans- and saturated-fats are generally found in hard stock fats, such as shortening, margarine, and confectionary products. To substitute these types of fats, researchers

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have actively studied alternative sources to produce hard stock fats that are free of trans- and saturated fats. One type of structured fats that is gaining attention as an alternative to trans-fats is oleogel. In oleogel systems, fats are entrapped in a solid-like matrix formed using gelling agents with low or high molecular weights [2]. Oleogel has similar system with hydrogel, both are type of hydrocolloids with their own unique properties and applications. Hydrogel can be used to dehumidify drying air that can reduce drying time and enhance the product quality [3], [4].

Edible oils and fats are plant-derived biological mixtures composed of ester mixtures derived from glycerol and fatty acid chains. The type and composition of fatty acids in the edible oils have a significant influence on the physical and chemical properties of oils and fats [5]. Various types of edible oils have been studied to form oleogel, including canola oil [6], soybean oil [7], virgin olive oil [8], camellia oil [9], hazelnut oil [10], sunflower oil [11], [12], [13], [14], and linseed oil [15]. In contrast to hydrogels, which have a water-based liquid phase, oleogel has an oil-based liquid phase. It has numerous potential applications in wide range of industries especially food, pharmaceutical, cosmetic, and petrochemical industries. In addition, oleogel has great potential in the food industry, such as reducing oil movement in cream-containing multi- constituent foods, such as chocolate, providing matrix for vegetable oils, hence lowering the requirement for saturated- and trans-fatty acids [16]. Furthermore, oleogel can be used as a good alternative to lubricant products as a bio-based lubricating grease. Many studies have already investigated the potential of oleogels for this application from various non-edible oils, especially castor oil [17], [18], [19], [20], [21].

Indonesia is a biologically diverse country, with pulp as one of the abundant forestry products. Other than used as a raw material for paper production, it can also be processed into more valuable derivative products, such as cellulose. The pulp has a significantly high cellulose content exceeding 90% [22]. Many derivative products with higher economic value and functionality can be produced from this cellulose [23]. Hydroxypropyl methyl cellulose (HPMC) is a derivative product of cellulose that can be utilized as an oleogelator [24]. Many studies have already applied cellulose derivatives as gelling agents to form oleogels, especially for food applications. Beside cellulose, gum-based gelling agents such as xanthan gum are also commonly used as oleogelators. Several studies have investigated the potential of xanthan gum in oleogel formation [2], [25].

In recent years, research on the formation of oleogel from various vegetables oil and oleogelators has been carried out, such as manipulating the mechanical strength of oleogels prepared using ethyl cellulose [26], formation and properties of soybean oil-based oleogel and various gelling agents [27], effect of temperature on oleogel formation using ethylcellulose [28]. Therefore, this present study focuses on the performance of cellulose- and gum-based emulsifiers for forming oleogels in various oil systems.

LITERATURE REVIEW

Oleogel formation

Oleogelation is one of the promising methods for structuring liquid oils into solid-like substances, and the resulting oleogels can be applied as alternatives of structured fats to reduce the consumption of unhealthy fats such as saturated- and trans-fat. Various types of oleogelators have

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been utilize to structure liquid oils to form oleogels. There are several methods or approaches that have been applied to fom ologels, such as direct dispersion, heating and cooling, and indirect methods, such as the emulsion-template approach. Table 1 summarizes previous studies on oleogel formation.

Table 1. Studies on oleogel formation					
Edible Oils	Oleogelator	Methods	Ref.		
Canola oil	HPMC, XG	Emulsion-Template Approach	[6]		
Soybean oil	HPMC, XG	Emulsion-Template Approach	[7]		
	Rice bran wax	Direct dispersion	[29]		
Virgin olive oil	Carnauba wax and monoglyceride	Heating and Cooling	[8]		
Camellia oil	Tea polyphenol-palmitate	Emulsion-Template Approach	[9]		
	particles and citrus pectin				
Hazelnut oil	Sunflower and Carnauba Waxes	Heating and Cooling	[10]		
Sunflower oil	Myverol (monoglyceride)	Heating and Cooling	[12]		
Linseed Oil	Beeswax and	Heating and Cooling	[15]		
	a mixture with 60:40 (w/w)	Direct dispersion			
	γ -oryzanol and β -sitosterol				

Oleogel applications

Oleogels are versatile products that have been used to structure edible oils for various food products such as margarine and shortening-like products, emulsion-based products, and other processed food, such as bakery, processed meat, ice cream and dairy, confectionary, and edible films [8]. The reports on the application of oleogel for food products are summaries in Table 2.

Table 2. Oleogel application for food products				
Vegetables Oil	Oleogelator	Application	Ref.	
Sunflower Oil	Monoglyceride	Cake/muffin	[12]	
Coconut Oil	Rice bran wax	Dairy and cream	[30]	
		cheese products		
Linseed Oil	Shellac wax	Chocolate	[15]	
Soybean Oil	Rice bran wax	Cream cheese	[29]	
Canola Oil	HPMC, MC	Bakery Products	[31]	
		Cookie fillings		
Canola Oil, Soybean	Ethyl cellulose	Processed meat	[32]	
Oil, and Linseed Oil		products		

Many studies also investigated the potential applications of oleogel-structured oils in non-food products, mainly as bio-based lubricating greases. A summary of related studies is shown in Table 3.

Table 3. Related studies for oleogel as biobased lubricating greases				
Vegetables Oleogelator Main Results			Ref.	
Oil				
Castor Oil	Cellulosic	The linear viscoelasticity behavior of gel-like dispersions are	[17]	
	Derivatives	unaffected by oleogelator molecular weights.		

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Vegetables Oil	Oleogelator	Main Results	Ref.
		 Temperature has little effect on the linear viscoelasticity behavior of oleogels. This is different from the behavior observed with commercial lubricating greases. 	
Castor Oil		 The type and concentration of the oleogelator and the vegetable oil have a significant impact on the linear viscoelastic properties and structure stability of the oleogel. The cooling profile used during gelification has a significant impact on the rheological behavior of SMS-based oleogels but has no effect on the GMS-based oleogels. Glyceryl monostearate (GMS) form mechanically stronger oleogel compared to sorbitan monostearate (SMS). GMS-based oleogels have significantly higher mechanical strength than SMS-based oleogels, and comparable to that of standard lubricating greases. 	[18]
Castor Oil	Isocyanate- Functionalized Chitin and Chitosan	The producing oleogels had adequate thermal resistance.The oleogels produced achieve the typical rheological behavior found in conventional lubricating greases.	[19]
		 The oleogels demonstrated outstanding frictional and wear behaviors, which comparable to commercial lubricating greases. Cellulose pulps can be convincingly recommended as effective oleogelator for the preparation of lubricant based oleogels and has potential as replacement for non-renewable thickener in lubricating grease preparations. 	[20]

Storage stability

Oleogel can oxidize slower than liquid oil caused by the restriction of oil movement in the gel system. It should be noted, however, that the primary goal of using oleogel is to replace saturated fatty acid-rich-oils with unsaturated fatty acid-rich oils, which are more susceptible to oxidation than saturated fatty acids. If necessary, special precautions, such as the addition of antioxidants, should be taken to reduce the oxidation rate and negative health effects of oleogel.

Synthetic antioxidants are now found in the majority of foods and pharmaceutical products. The additives are added to extend the product life by inhibiting the oxidation of unsaturated double bonds in fatty acids. Two conventional synthetic antioxidants utilized currently are butylated hydroxyanisole (BHA) and butylated hydroxyl-toluene (BHT). Every year, however, new evidence emerges that synthetic antioxidants utilized in industry may be carcinogenic to human cells, encouraging an intense exploration for new, natural, and efficient antioxidants. A brief review focuses on the oxidative stability of oleogel is presented in Table 4.

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Table 1. Oleogel oxidative stability					
Edible Oils	Oleogelator	Storage Condition	Oxidative Stability	Ref.	
High-oleic soybean oil (HOSO)	Rice bran wax	T = 5°C, 14 days	Oxidation analysis revealed that oleogel samples contained more volatile compounds than oil.Minimum oleogel degradation due to thermal processing and storage	[29]	
High-oleic sunflower oil	Monoglycerides (MG)	$T = 5^{\circ}C$, 8 weeks	 Structured through MG self-assembly improved the oxidative stability of the oil, implying that the matrix properties of MG oleogel may interrupt the occurrence of the beginning phases of oxidation process. The rate of peroxide formation is determined by the amount of oxygen available in the system. The existence of the MG crystalline structure would act as a barrier, preventing oxygen from entering the reaction sites. 	[33]	
Camellia Oil	glycerol mono- laurate (GML)	T = 40°C, 14 days	 Lower oxidation rates were observed in oleogel, indicating improved oxidative stability. Rising GML concentration, causing the oxidation rate of the emulsion declined but remained higher than that of the respective oleogel, which could be attributed to increased interfacial surface induced by the smaller particle size in the emulsions. 	[34]	
Canola Oil	Candelilla, carnauba, and beeswax	T = 60°C, 18 days	 Compared to the oleogel, the peroxide value of fresh oil raised considerably quicker. Candelilla wax oleogel, carnauba wax oleogel, and beeswax oleogel had the lowest to highest peroxide values. These peroxide value trends were related to the hardness of the oleogel. The lower the peroxide value, the harder the oleogel. 	[35]	
Pine nut oil (PNO)	Soy protein-tannic acid	$T = 50^{\circ}C, 16 \text{ days}$	• Oleogel had lower values for peroxide, thiobarbituric acid, and conjugated diene than PNO, implying outstanding oxidation stability.	[36]	
Canola oil	Sunflower wax, Sitosterol-oryzanol and ethyl cellulose	$T = 20^{\circ}C, 6$ months	The characteristics of the bouillon cubes can be adjusted through gelator selection and inclusion level.If antioxidants are used, the risk of intolerable oxidation effects during storage is reduced.	[37]	
Fish oil	Sorghum wax	$T = 50^{\circ}C, 6 \text{ days}$	The oleogel formulations protected the fish oil from oxidation.The secondary oxidation products in fish oil occurred much faster than in oleogel.	[38]	

RESEARCH METHOD

Experiment was carried out by generating oleogel from cellulose and gum-based oleogelators in three types of oils: canola oil, sunflower oil, and castor oil. Experiments were carried out in three stages: oleogel formation, oil loss measurement, and storage stability.

Materials and equipment

Canola oil and sunflower oil are used as commercial cooking oil, and the non-edible oil is represented by castor oil. The main compositions of the fatty acids from these vegetable oils are summarized in Table 5. The HPMC and xanthan gum are commercially available. Oleogel is formed by varying the amount and type of gelling agent. Distilled water is used to dissolve powder samples. All concentration parameters are expressed as percent by mass (% w/w).

Table 5. Fatty acids main compositions of vegetable oil					
	Sunflower Oil	Castor Oil			
Saturated	Palmitic Acid (C16:0)	3.9	6.2	14.2	
	Stearic Acid (C18:0)	1.1	2.8	7.0	
Unsaturated	Oleic Acid (C18:1-cis)	64.4	28.0	44.7	
	Linoleic Acid (C18:2-cis)	20.4	62.2	32.8	
	Linolenic Acid (C18:3)	9.6	na	0.2	

Oleogel formation

The method of oleogel formation refers to [9] with some modifications. The oleogel formation is performed by mixing vegetable oil with a gelling agent according to the specified variations. The stirring process takes place at a temperature of 12oC under atmospheric pressure for approximately 45 minutes. HPMC and XG powders were prepared by weighing the exact amount of the powder and dissolving it in distilled water. After that, the vegetable oil is slowly poured under stirring. The resulting products are subsequently characterized.

Determination of oil loss

The centrifugation method can be implemented to determine the ability of the oleogel to bind oil [28]. Calculation of oil loss percentage is done by comparing the weight of released oil to the initial weight of the oleogel sample. The amount of oil released is calculated using Equation (1), where w1 is the initial mass of the oleogel sample and w2 is the final mass of the oleogel sample after being separated from the released oil. For this process, 30 g of oleogel sample (w1) was put into a 50 mL centrifugation bottle. The sample was then centrifuged at 5000 rpm for 30 minutes until some of the oil was released. The released oil is then separated from the oleogel by removing it from the plastic bottle. The oleogel remained in the bottle after the centrifugation process and separated from the loose oil was then weighed (w2).

Oil loss = $(w_1-w_2) / w_1 \times 100\% [1]$

Storage stability

The stability of oleogel products produced during the storage period is observed by monitoring the changes in oleogel products stored at room temperature for a period of 28 days. The

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properties include visual appearances, such as colour, structure, and smell. From the observations, the stability of the oleogel during the storage period is determined and the parameters of the product that need to be improved are considered.

DISCUSSION

Oleogel formation

Both HPMC and XG are used as food additives, each of which is used primarily as a stabilizer and thickener. XG is acquired naturally as product from *Xanthomonas campestris* fermentation, while HPMC is synthesized from natural cellulose by substituting several hydroxyl groups with methoxy hydrophobic groups. HPMC shows a high surface activity and is therefore used to stabilize colloidal dispersions such as emulsions and foams. XG, on the other hand, has a non-active surface but can still be used to assist colloidal stabilization (when used in conjunction with a surface-active stabilizer) by extending kinetic stability through increased viscosity of the bulk phase [15]. In our work, 60% (w/w) of oil in a water emulsion is prepared by varying oleogelator using only HPMC, while only XG and the combination of MC-XG as a stabilizer. It appears that oleogel is formed when the gelling agent used is a combination of HPMC and XG.

Table 6. Gelling agent effect in oleogel formation					
Oil : Water Ratio	Gelling agent	% w/w	Result		
1:0	HPMC	1	Oleogel not formed		
1:0	XG	1	Oleogel not formed		
3:2	HPMC	1	Oleogel not formed		
3:2	XG	1	Oleogel not formed		
3:2	HPMC and XG	1	Oleogel formed		

According to Daniel and Rajasekaran [39], oleogel formation at room temperature can be prepared with saturated fatty acids having carbon chain lengths of 10 to 31. The higher the amount of fatty acids, the higher the ability of gel formed, consistent with the increase of chain length. The melting point of oleogel increases from 30 to 70°C with an increase in the length of the fatty acid chain. Studies related to the carboxyl group show that the position of the hydroxyl group and the length of the acyl chain portray an essential role in the formation of oleogel.

Gandolfo et al [40], also investigated the possibility in the preparation of oleogel disclosed using a series of fatty acids varying from carbon chains of 16 to 22 (palmitic acid, stearic, arachid and behenic acids). It was discovered that fatty acids can structure sunflower oil and other vegetable oils as low as 2% (w/w) at 5° C. Furthermore, oleogel hardness also increases linearly with increasing concentrations of fatty acids, due to the rise in the amount of solids. At a constant temperature, the solubility will also be constant.

Oleogel is used in a variety of food products that contain conventional solid fats. Oleogel can be applied as spread products, hence recent research has been directed to determine their viability to replace conventional spreads. Several characteristics, e.g., firmness, stickiness, oxidative stability, spreadability, and other sensory properties, such as grassy, milky, rancid, fatty, sweet, salty, waxy, grittiness, and mouth coating were evaluated in comparison to traditional spreads [35].

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No.	Oil	Sample Code	Ratio of HPMC : XG	Gelling Agent %w/w
1		CX11-CO	1:1	2
2	Canola oil	CX21-CO	2:1	2
3		CX31-CO	3:1	2
4		CX11-SO	1:1	2
5	Sunflower Oil	CX21-SO	2:1	2
6		CX31-SO	3:1	2
7		CX11-JO	1:1	2
8	Jatropha Oil	CX21-JO	2:1	2
9		CX31-JO	3:1	2

Table 7. Gelling agent composition

Determination of Oil Loss

Oil losses from oleogel obtained using different ratio combination of gelling agents and oil are shown in Table 8. The loss of oil in oleogel indicates the relative binding capacity of oleogel to oil. Before centrifugation, the oleogel samples contained more than 75-97% (w/w) of liquid oil. After centrifugation, the oleogel in variation 3 loses more oil compared to variation 4, which shows a weaker oil binding capacity. The binding capacity of oil-oleogel can be associated with the mechanical strength of emulsions and oleogel by considering observed values of oil loss. The best oleogel is produced with a variation of HPMC: XG = 1: 1, in an amount of 2 gr (2% w / w), where the oil binding capacity is better than other variations.

Table 8. Oil binding capacity				
No.	Sample Code	Oil Loss (%)		
1	CX11-CO	15.46		
2	CX21-CO	22.06		
3	СХ31-СО	33.31		
4	CX11-SO	3.99		
5	CX21-SO	8.83		
6	CX31-SO	30.00		
7	CX11-JO	11.50		
8	CX21-JO	16.10		
9	CX31-JO	11.73		

The interaction between oleogelator compositions and saturated fatty acid compositions (%) in oil is analyzed using response surface methodology. The RSM analysis resulted in the following quadratic model.

Oil Loss = 15,1 - 2,76 Saturated FA + 8,9 HPMC:XG + 0,1372 Saturated FA*Saturated FA + 1,77 HPMC:XG*HPMC:XG - 0,752 Saturated FA*HPMC:XG

The accuracy of the model is determined by the coefficient of determination test (R2). The R2 value obtained in this study was 88.42%, where the R2 value > 70% indicates that the experimental and prediction values are quite accurate. The determination of the variables that influence the percentage of oil loss can be determined using the Pareto Chart presented in Figure 1.

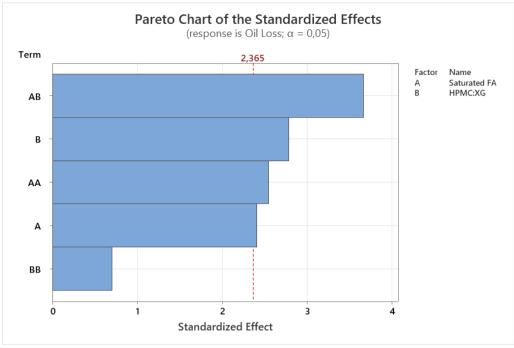


Figure 1. Pareto chart of the standardized effects

Variables that have a significant effect are determined from the length of the bar that exceeds the reference line (2.365). From Figure 1 it can be seen that AB, B, AA, and A have a significant effect on the percentage of oil loss.

The contour plot for interaction of oleogelator composition and saturated fatty acids composition of oil is illustrated in Figure 2. From the contour plot, it can be seen that a higher saturated fatty acids concentration in oil will result in a stronger oleogel matrix that is measured by the percentage of oil loss. Higher HPMC composition linear with less XG composition will decrease the oil binding capacity of oleogel. HPMC acts as an emulsifier while XG as thickener, that implies less XG will produce less thick oleogel. Emulsions with stronger mechanical strength tended to produce oleogel with stronger gel structure and better oil binding capacity [15]. The gel strength and oil binding capacity of oleogel is related to emulsion mechanical strength as well as the network of soft solids. The binding of liquid oil to polysaccharides resulted in the formation of semi-crystalline oleogel, which were stabilized by intramolecular or intermolecular hydrogen bonds between polysaccharides [27].

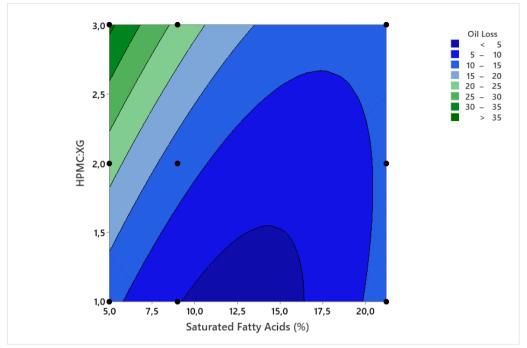


Figure 2. Contour plot for interaction of oleogelator composition and saturated fatty acids composition of oil

Storage Stability

The storage stability of the oleogel was observed at room temperature for 28 days. The product of oleogel from the gelling agent combination which produces structurally stable oleogel did not change significantly during the storage period at room temperature for 28 days. However, because the water content in the oleogel is still quite high and has not gone through the drying process, the surface of the oleogel has been overgrown with fungus. The oleogel drying process was not successful because the oleogel product was unstable, that make the oil could be separated from the gel. In general, it was found that during the storage period of the oleogel product, there was no change in the structure and color of the oleogel. As a result of some of the oil being oxidized during the heating process, the resulting oleogel product has a stink aroma that can affect the application of oleogel, especially in food products. For this reason, it is recommended to add antioxidants in the oleogel formation process, in order to reduce the high rate of oil oxidation. In addition, oil oxidation can also be reduced by optimizing the oleogel stirring time and temperature of formation.

CONCLUSION

Canola oil, sunflower oil, and castor oil-based oleogels were formed by combining HPMC as the main emulsifier and XG as the thickener. All oleogel samples formed are not stable enough to be dried to remove moisture. The mechanical strength of the obtained oleogel was tested by centrifugation, and the oil binding capacity was measured. Oleogels with stronger mechanical properties exhibit a denser soft solid structure. The mechanical strength of stronger oleogels is proportional to the better oil retention ability of oleogels. Higher mechanical strength of the emulsion indicates a tighter soft-solid structure, stronger gel, and higher oleogel capacity. HPMC and XG thickeners produce oleogels with time-dependent behaviour and structural recovery. It is

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also stable over a wide temperature range. Although structured oleogels exhibit fairly good stability during storage at room temperature, the oxidation rate requires further improvement.

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