

Ultrafiltration for homogenization of wheat germ oil:water system: Droplet size distribution and stability of emulsion

Quoc Dat Lai (✉ lqdat@hcmut.edu.vn)

Ho Chi Minh City University of Technology (HCMUT)

Ngoc Thuc Trinh Doan

Ho Chi Minh City University of Technology (HCMUT)

Hoang Dung Nguyen

Ho Chi Minh City University of Technology (HCMUT)

Thi Thuy Loan Huynh

Ho Chi Minh City University of Technology (HCMUT)

Research Article

Keywords: Wheat germ oil, ultrafiltration, homogenization, particles diameter, stability of emulsion

Posted Date: March 23rd, 2022

DOI: <https://doi.org/10.21203/rs.3.rs-1475675/v1>

License:  This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Abstract

Wheat germ oil (WGO) in water emulsion is increasingly being used in a variety of fields due to its outstanding nutritional and health benefits. To enhance WGO composability, emulsification should be performed to allow them to dissolve more easily in the food matrix or active packaging. Membrane emulsification is a promising advanced homogenization technology that has recently been developed. This research focused on application of membrane emulsification for homogenization of WGO-in-water emulsion polyether sulfone (PESU) membrane with 0.45 μm of pore size, operating pressure from 5 to 9 bar, with WGO fraction in range 10 – 20% w/w and lecithin ratio in range 0 – 0.2% w/v were evaluated on the mean of particle diameter, distribution of particles diameter, stability of emulsion, and permeate flux. Higher operating pressure led to smaller mean particle diameters, higher homogenization efficiency and permeate flux. However, increasing the WGO fraction showed the opposite trend. Notably, increasing lecithin ratio from 0 to 0.2%, the mean particle diameter increased from 3.36 to 7.72 μm , homogenization efficiency decreased from 98.54 to 97.68%, and permeate flux decreased from 105.95 to 77.45 $\text{L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$. The results showed the advantage of using PESU membrane in WGO-in-water homogenization, that produce emulsions with small particle size (50% of particle volume was less than 5 μm), high emulsion stability (greater than 95%) and lower emulsifier usage. Results imply premix membrane emulsification is potential to apply homogenization WGO-in-water emulsion.

1. Introduction

Wheat germ, which is part of the wheat grain, a by-product derived from the wheat milling industry, containing about 10–15% of oil. Wheat germ oil (WGO) is widely recognized as a good vegetable oil with high nutritional value (Brandolini and Hidalgo 2012). WGO contains numerous beneficial bioactive compounds that are good for health. WGO has a higher tocopherol content than other vegetable oils, up to about 2,500 mg/kg, with α -tocopherol is predominant accounting for 60% of the total content (Ghafoor et al. 2017). In particular, it is worth noting that vitamin E has up to 500 ppm (Türkoğlu et al. 2021). WGO contains a high concentration of unsaturated fatty acids, specifically linoleic and linolenic acids, which are important compounds in human metabolism but cannot be produced by organisms' method. WGO is rich in high-nutritional-value compounds, including sterols, unsaponifiable matter, pigments, phytosterols, policosanol, octacosanol, ceramide, etc. WGO is also a valuable source of micronutrients, which are necessary for many different bodily functions, such as A, B complex vitamins, and many minerals and fibers (Brandolini and Hidalgo 2012; Šramková et al. 2009). Due to its high concentration of bioactive compounds, WGO has the potential to provide numerous health benefits to humans when used on a regular basis. It functions as an antioxidant, preventing lipid oxidation and thus assisting in the cell protection from free radicals, as well as lowering plasma and blood cholesterol levels (Ghafoor et al. 2017; Jha et al. 2013; Zhu et al. 2011). It increases the efficiency of microcirculation in venous and arterial, blood flow in vessels. Therefore, it has the potential to prevent cardiovascular disease (Ghafoor et al. 2017). WGO contains linoleic and linolenic acids, which are precursors to prostaglandins, a group of hormones involved in muscle contraction and anti-inflammatory activity (Harrabi et al. 2021; Köse 2021).

WGO also helps to lower cholesterol levels in the liver, provide precursor of cell membrane phospholipids, improve physical endurance, decongest the body, and slow the effects of aging (Koba and Yanagita 2014). It is considered a good food for athletes because it helps to improve exercise performance. Besides, it has the ability to improve platelet aggregation, and plasma cholesterol levels, as well as reduce the risk of obesity (Brandolini and Hidalgo 2012; Ghafoor et al. 2017). WGO can reduce free radicals and the immunosuppressive effects of lipid peroxidation in UV-exposed skin, as well as slow skin aging by moisturizing and smoothing skin (Dunford 2009). WGO also has strong antimicrobial properties against a variety of pathogenic bacteria (Al-Rimawi et al. 2020; B.-S. Choi and Kang 2009). Because WGO has a wide range of potential health benefits, it is becoming increasingly popular to develop in food production, formulation products, pharmaceuticals, cosmetics and even agriculture (Boukid et al. 2018; B.-S. Choi and Kang 2009; Türkoğlu et al. 2021; Wang et al. 2021).

WGO-in-water emulsion is a temporarily stable mixture of two immiscible liquids, the dispersed phase has WGO droplets and the dispersion medium is an aqueous phase. WGO-in-water emulsion are formed when two non-soluble (i.e. WGO and water) liquids with surfactant additions, are agitated together to disperse one liquid into the other, in the form of drops. WGO-in-water emulsion assists in overcoming difficulties in WGO incorporation in some products, particularly liquid products, caused by differences in rheological properties and solubility. The use of simple delivery systems such as oil-in-water, or water-in-oil emulsions is one of the most effective and widely used methods for combining oil into foods (S. J. Choi and McClements 2020; Galanakis 2019). It enables the mixing of components with different rheological properties, particularly solubility, and forms a physical barrier (i.e. interfacial layer) that helps prevent fat oxidation due to contact between oil and oxygen or prooxidants. Furthermore, because water soluble digestive/lipolytic enzymes are more readily active at the interface between oil/water and the surface area of the emulsion is increased, WGO-in-water emulsion improves fat absorption efficiency in the human body (Bodewes et al. 2015). WGO-in-water emulsions also combine with wall materials more effectively during spray drying, leading to greater encapsulation efficiency and easier WGO use and maintenance (Karadeniz et al. 2018). Therefore, WGO-in-water emulsion facilitates the addition of WGO into products such as food, cosmetic, and pharmaceutical. WGO-in-water emulsions are now used to product spray drying products, micro and nano WGO capsules; as well as to preserve the oil stability of cooked fish fillets; and use in cosmetics; or substitute animal fat in beef burgers (Barros et al. 2021; Ceylan et al. 2020; Dunford 2009).

Membrane emulsification, also known as membrane homogenization, a relatively new emulsification technology (Spyropoulos et al. 2014a). Membrane emulsification employs low energy inputs to press coarse emulsion through membrane pores, resulting in the formation of droplets at the membrane. Droplets form at pore openings and detach when reaching a certain size (Alliod et al. 2018). This is the result of a balance between four major forces that govern the membrane emulsification process: shear, pressure/inertia, interfacial tension, and buoyancy forces (Joscelyne and Trägårdh 2000). Membrane emulsification enables the development of emulsions with a uniform droplet size distribution over a wide range of mean droplet sizes ranging from less than 1 to more than 100 μ m (Vladisavljević 2019a). The advantage of this method over other emulsification methods is that it is low-pressure and does not

significantly raise temperature or shear stresses during the process, which can prevent damage to emulsion components such as proteins, starches, etc. (C Charcosset et al. 2004; Vladisavljević 2019a). At low energy inputs, it still can be efficient in preparing droplets with very narrow particle size distributions. Even so, it easily can produce emulsions with higher droplet concentration (Vladisavljević 2019b). Beside that, the technique is highly appealing due to its simplicity, low surfactant requirement, suitability for large-scale production, and continuous or semicontinuous operation (Emma Piacentini and Giorno 2016). It has been used to produce monodispersed multiple emulsions (oil-in-water (o/w) and water-in-oil (w/o) emulsions), as well as o/w/o and w/o/w emulsions, especially those with shear sensitive components (Catherine Charcosset 2021; Consoli et al. 2020; E Piacentini et al. 2020). Applications contribute to the creation of high value in pharmaceuticals, chromatography beads, luxury cosmetics, and food industry (Jiang et al. 2020; Spyropoulos et al. 2014b; Vladisavljević 2019b).

In conventional direct ME, fine droplets are formed on the interface between membrane and continuous phase by pressing pure the dispersed phase through the membrane (Aserin 2007). However, there are several drawbacks to direct ME, including a relatively low dispersed phase flux, low productivity, a long production time, suitability for emulsions with a dispersed phase of up to 30%, and difficulty in preparation and operation. Premix ME was developed to overcome the drawbacks of direct ME by forcing a preliminarily emulsified coarse emulsion (rather than a single pure dispersed phase) through the membrane (Vladisavljević and Williams 2005). This is accomplished by first mixing the two immiscible liquids together with a conventional stirrer mixer, then passing the preliminarily emulsified emulsion through the membrane (SUZUKI et al. 1996). Premix ME was used to homogenize an oil-in-water emulsion in food. There are several studies that have evaluated the influence of membrane and emulsifier type and demonstrated the effectiveness of premix ME compared to other homogenization methods (Berendsen et al. 2014; Ramakrishnan et al. 2013). However, the most important factors influencing membrane emulsification include not only membrane and emulsifier parameters, but also dispersed phase and process parameters. It influences filtration process efficiency as well as properties of the emulsion system, such as particle size distribution and emulsion stability (Catherine Charcosset 2009; Jiang et al. 2020). So far, there are no studies on using premix ME to emulsify WGO-in-water emulsion. Therefore, the present study aims to investigate the feasibility of production of WGO-in-water emulsion by premix membrane emulsification, using ultrafiltration membrane, specifically, polyether sulfone (PESU) membrane with cellulose acetate active layer. The influences of operating pressure, content of WGO phase, and lecithin ratio on the droplet mean diameter, particle size distribution, emulsion stability, and permeate flux were investigated. The study's findings will reveal the feasibility of using premix ME in WGO-in-water emulsification in particular, and other emulsions in general.

2. Experimental

2.1. Material and chemical

Wheat germ oil was extracted from wheat germ with a moisture of 13–15% w/w, and oil concentration of about 8–12% w/w.

WGO preparation was carried out in accordance with Megahed, 2011, with slight modifications. Wheat germ that had been milled in day was lipid-deactivated by autoclaving at 121°C for 15 minutes. Following that, wheat germ was dried at 60 °C to a moisture content of 8%. Wheat germ oil was extracted by hexane as a solvent. The wheat germ: solvent ratio was 1:3. The extraction was conducted at ambient temperature for 12 hours. The crude oil was recovered after distilling the micelle mixture of oil and solvent, which was liquefied via condenser in rotary evaporator based on the different evaporating temperature. Using a filter paper No. 4 to remove residues from crude oil that could cause membrane blockage. This step occurred under vacuum condition. The resulting wheat germ oil contained 1,170 ppm of vitamin E, and iodine value of 105 g/100g.

All chemicals were analytical grade and supplied by Sigma Aldrich (USA).

2.2. Membrane apparatus

Hydrophobic polyether sulfone (PESU) membrane with cellulose acetate active layer was provided by Alfa Laval (Denmark). This membrane is plate with a pore diameter of 0.45 µm. Surface active area of membrane is 14.6 cm².

Membrane homogenization was performed in batch concentration mode using a stainless steel HP4750 Stirred Cell. This is a dead-end model with a magnetic stirrer, which supplied by Sterlitech Corporation (Kent, WA, USA). A stirring speed of 300 rpm was set in each run to reduce the concentration polarization and fouling on the membrane surface.

2.3. Experimental design

Firstly, the premix emulsions were prepared, contained WGO dispersed in water, and emulsifier lecithin. The mixture was stirred at 200 rpm for 10 min and then the premix was immediately placed in the feed tank and pumped in the syringe pump. The homogenization process was conducted in Sterlitech HP4750 Stirred Cell. In order to investigate the effect of operating pressure, the operating pressures were held at a fixed value ranging from 5 to 9 bar, with an emulsion containing 10%v/v of WGO and 0.1%w/v lecithin. Besides that, the control sample was prepared without homogenization by membrane. Then, the investigation on WGO fraction was conducted with the emulsion which had oil content of 10 to 25% v/v and lecithin ratio of 0.1% w/v at 9 bar. The effect of lecithin ratio on emulsion system was studied in the range from 0 to 0.2% w/v at 9 bar operating pressure and 10%v/v WGO fraction.

2.4. Analytical methods

2.4.1. Distribution size of particles

Distribution size of particles was measured immediately after preparation at room treatment by laser diffraction particle size analyzer based on measuring the angular variation in intensity of light scattered as laser beam passed through a dispersed particulate sample. The Phase Doppler Particle Analyzer was

used to conduct the measurements (PDPA). Mean particle diameter was calculated according to the Sauter diameter (D_{32}) (Major-Godlewska 2019).

$$D_{32} = \frac{\sum_i z_i \cdot D_i^3}{\sum_i z_i \cdot D_i^2}$$

In where, Z_i is the number of dispersed particles of size D_i

2.4.2. Homogenization efficiency (Stability of emulsion)

The homogenization efficiency was used to evaluate the stability of emulsion, that determined by the NIZO centrifugation method. Homogenization efficiency was determined by the fat content of the bottom 25 ml is divided by the fat content of the whole sample prior to centrifuging at 1000 rpm 30 minutes (Ransmark et al. 2019). The fat content was determined by Adam – Rose – Gottlieb. Diethyl ether and petroleum ether were used to extract the fat in an ammonia and ethyl alcohol solution. The ether mixture was evaporated, and the resulting residue was weighed (Thiex et al. 2003).

2.5. Statistical treatment

Experiments were triplicated. The results were presented as means \pm standards deviation. The data were analyzed using by 1-way analysis of variance and means of each pair were compared at 5% significance level by using the least significant difference (LSD) test in R software (version 3.5.3).

3. Results And Discussion

3.1. Influence of pressure of membrane homogenization of WGO-in-water emulsion

3.1.1. Distribution of size particle

Table 1 and Fig. 1, respectively, presented the mean particle diameter and particle size in diameter of WGO-in-water system after membrane homogenization at different operating pressure, 10%v/v of oil fraction and 0.1%w/v of lecithin. The results showed that increasing the operating pressure range from 5 to 9 bar resulted in a sharp decrease in mean particle diameter from 8.14 to 3.37 μm . There was a significant difference between the pressures, except at 7 and 8 bar, where there was none. Coalescence was most likely a contributing factor to the relatively large droplet sizes under these processing conditions, as these mean particle diameters greater than the pore diameter by 7.5 to 18.1 times (Catherine Charcosset 2009). The particle size in emulsion was the result of the balance between droplet disruption and re-coalescence at adjacent pores. As the pressure raised, the particle size shifted to the left, where the particle size was smaller. Unlike high pressure homogenization, the mechanism of membrane homogenization did not involve turbulence, compression, or impact to produce small dispersed particles (Joscelyne and Trägårdh 2000). In membrane homogenization, there are main forces

impact on droplet detachment: shear, pressure/inertia, interfacial tension. Each of the forces plays a role in droplet detachment from the surface of the membrane (Spyropoulos et al. 2014b). Therefore, the force acting on the detaching droplet varies depending on the pressure condition, resulting in a difference in particle size. In there, when the increasing operating pressure increased the pressure difference on both sides of membrane, allowing the continuous phase to pass through the membrane and form foam, which attracted the dispersed phase (Nazir et al. 2015). The higher the pressure, the greater the inertia force and the easier it was to overcome the binding force between the membrane and the droplets, breaking the surface tension and resulting in the formation of small sized particles (Spyropoulos et al. 2014b). Besides, similar to high pressure homogenization, high pressure also increases the shear stress on the droplet (Schultz et al. 2004). In dead-end membrane homogenization, the impact of shear stresses occurred inside the pores (Vladislavljević et al. 2004). Hence, the droplet breakdown was more intense, and particle size was smaller at higher pressures. These behaviors were consistent with previous observations of Suzuki et al., 1998, and Vladislavljević et al., 2004s.

Table 1
Effect of homogenization pressure on mean particle diameter of emulsions containing 10%v/v WGO, 0.1% w/v lecithin

Homogenization pressure (bar)	5	6	7	8	9
D ₃₂ (µm)	8.14 ± 0.21	6.21 ± 0.05	5.09 ± 0.14	5.07 ± 0.37	3.37 ± 0.18

An increase in operating pressure resulted in insignificant change in mean particle size at 8 bar. This could be influenced by the type of emulsifier or rather the adsorption kinetics and droplet formation times (Schröder et al. 1998). If the time it took to form particles was longer than the time to reduce the surface tension enough for them to be detached by drag forces, interfacial tension dynamics had little or no effect on droplet size. As a result, the size did not change as the flux or pressure increased (Joscelyne and Trägårdh 2000). The rate of particle formation and detachment from the membrane both increased as increasing pressure, but the processes of stable particle formation were also affected by emulsifier, porosity, membrane hydrophobicity and concentration polarization (Joscelyne and Trägårdh 2000). In the case of the high surface expansion rate of droplet did not allow for adequate coverage by particles or emulsifiers, it could lead to recoalescence at the membrane surface and make the emulsion unstable (Van der Graaf et al. 2004). High transmembrane pressure could also produce oil droplets in a 'jetting' regime, at which droplets at adjacent pores could coalesce, particularly when the emulsifier concentration was low and steric hindrance between the droplets had not been formed (Manga and York 2017). Hence, when the pressure was increased from 7 to 8 bar, the mean particle diameter did not change. But in the range of size less than 1.5µm, the volume distribution of particle at 8 bar was still higher than at 7 bar.

3.1.2. Homogenization efficiency

The change of homogenization efficiency by operating pressure in ultrafiltration homogenization of WGO-in-water emulsion containing 10%v/v of oil fraction and 0.1%w/v of lecithin was shown in Fig. 2.

Increasing the operating pressure from 5 to 9 bar, the efficiency increased from 93.48 to 98.31%. There was a significant difference between the operating pressures. Compared with the control, which was not homogenized, the membrane homogenized samples had nearly twice greater homogenization efficiency. This demonstrated that membrane homogenization could improve the dispersion of WGO particles in continuous phase water. Furthermore, increasing the operating pressure also improved homogenization efficiency, resulting in smaller dispersed particles. Therefore, the WGO-in-water emulsion was more stable. Although the mean particle size was the same at 7 and 8 bar, the volume distribution of particle in the small size region was larger at 8 bar than at 7 bar. Consequently, the emulsion homogenized at 8 bar was more stable than at 7 bar. The obtained results agreed with the findings of Zanatta et al., 2017, that emulsion stability increased at higher feed pressure when emulsifying sunflower oil in water emulsions using a ceramic membrane.

3.1.3. Permeate flux

Figure 3 presents the effect of operating pressure on permeate flux in homogenization of WGO-in-water emulsion at 10%v/v of oil fraction and 0.1%w/v of lecithin. The permeate flux was not different at 5 and 6 bar. When the pressure is raised from 6 to 9 bar, the flux through the membrane increases dramatically from 67.12 to 100.68 L.h⁻¹.m⁻². As the pressure increased, the driving force pushing the components across the membrane increased, and so did permeate flux. This allowed the droplets to detach from the membrane at a faster rate, limiting the re-coalescence of particles formed in the adjacent pores at the membrane surface. This result implied that droplets breakup did not significantly contribute to the friction loss in the membrane, and there was no appreciable membrane fouling throughout the experiments. It agreed with observations of Nazir et al., 2011, in premix membrane emulsification of oil-in-water with nickel sieves.

3.2. Influence of oil fraction on membrane homogenization of WGO-in-water emulsion

3.2.1. Distribution of size particle

Table 2 and Fig. 4 respectively show the mean particle diameter and particle size distribution at different WGO fraction in homogenization membrane of WGO-in-water emulsion at 9 bar and 0.1%w/v of lecithin. The oil-to-volume fraction was increased from 10–20%, the mean particle diameter increased from 3.37 to 7.72µm. With a smaller oil ratio, the particles in the system were more dispersed in the small particle size region. This result was consistent with experiments of Surh et al., 2008, and Zanatta et al., 2017. This behavior can be explained by the fact that in the absence of re-coalescence the mean particle diameter of homogenized oil droplets was primarily dictated by the mean pore size and the applied shear stress inside the pore (Spasic and Hsu 2005). At lower disperse phase content, the permeate flux was higher and the emulsion viscosity was lower, so that under certain operating condition, the shear stress inside the pores was higher. The droplets passed through the membrane more quickly and thus were disrupted more easily into smaller droplets. Besides that, the particle at the adjacent pores had less opportunity to interact with other (C Charcosset et al. 2004; Vladisavljević et al. 2004). Moreover, the

amount of emulsifier was sufficient to stabilize the interface of the particles coming out of the membrane pores, the droplet could be small in size due to re-coalescence restriction (Nazir et al. 2011). Hence, the mean particle diameter was higher at higher of WGO fraction.

Table 2
Effect of oil fraction on mean particle diameter in homogenization of emulsions containing 0.1% w/v lecithin at 9 bar

Oil fraction (%v/v)	10	12	15	18	20
D_{32} (μm)	3.37 ± 0.26	3.48 ± 0.17	4.35 ± 0.22	5.89 ± 0.09	7.72 ± 0.31

3.2.2. Homogenization efficiency

The effect of oil fraction (%v/v) on homogenization efficiency in membrane homogenization of WGO-in-water at 9 bar and 0.1%w/v of lecithin is shown in Fig. 5. The efficiency of emulsion system increased in range from 98.54 to 93.66% as the oil fraction rose from 10 to 20%. The homogenization efficiency differed significantly between the different oil fractions. This finding demonstrated that the stability of an emulsion decreased as the oil fraction in the mixture increased. It is explained by the fact that the more dispersed the oil particles were in the system, it was easily to accumulate and separate during centrifugation, resulting in a lower stability of emulsion. Moreover, the agglomeration of oil particle in premix emulsion made it difficult for dispersed particles to pass through the membrane because the dynamics remained constant while the binding force in the dispersed phase increased. Furthermore, the high fraction of WGO in the emulsion could cause accumulation on the membrane surface and inside pores, preventing lecithin from passing through the membrane that significantly reduced the efficiency of homogenization process (Spasic and Hsu 2005). Therefore, as increasing oil fraction, the oil availability increased, thereby emulsion stability decreased. Less stable emulsion rapid declined in stability of emulsion over short periods of time, causing the separation of oil and aqueous phase.

3.2.3. Permeate flux

Figure 6 presents the effect of oil fraction on permeate flux in homogenization membrane of WGO-in-water emulsion containing 0.1%w/v of lecithin at 9 bar. Under the same pressure and emulsifier content conditions, increasing the oil fraction in system to 20% reduced the permeate flux from 95.89 to 63.34 $\text{L}\cdot\text{h}^{-1}\cdot\text{m}^{-2}$. The permeate flux peaked at 10% and 12% of the oil fraction, respectively. The explanation for this observation was that an emulsion with a high dispersed phase fraction could not easily pass through the membrane because the disruption of a large number of droplets required a large amount of mechanical energy, resulting in a low permeate flux (Surh et al. 2008). In the low oil fraction, the pressure difference was predominantly used to overcome flow resistance forces, whereas at the higher oil fraction, the higher amount of energy used to overcome interfacial tension forces and droplet break-up, the less energy was available for emulsion flow (Spasic and Hsu 2005). Surh et al., 2008, reported that increasing corn oil content from 10 to 20 wt.% in lecithin-stabilized oil-in-water emulsion decreased the transmembrane flux in premix membrane emulsification by a Shirasu porous glass membrane.

3.3. Influence of lecithin membrane homogenization of WGO-in-water emulsion

3.3.1. Distribution of size particle

The influence of lecithin ratio on the mean particle size and particle size distribution was demonstrated in Table 3 and Fig. 7 in homogenization of WGO-in-water emulsion at 9 bar, and 10%v/v of oil fraction. At the condition of homogenizing pressure of 9 bar and oil fraction of 10%, lecithin ratio increased from 0 to 0.2%, the average particle size in the dispersed phase increased from 3.36 to 7.72 μm . At 0 and 0.05%, the particles were distributed in a narrower region (0–1 μm), while at 0.1, 0.15, and 0.2% of lecithin ratio, particles were gradually distributed in a larger region (1.5–13 μm). When the particle size in the dispersed phase was large, the particles easily coalesced and separated, resulting in a less stable emulsion and reduced homogenization efficiency. The findings contradicted previous research on the effect of lecithin ratio on emulsion properties using a different homogenization method (Floury et al. 2000; Yan et al. 2017). This study found that lecithin had no role in supporting WGO-in-water homogenization by membrane.

Table 3
Effect of lecithin ratio (%w/v) on mean particles diameter in homogenization of emulsions containing 10% v/v WGO at 9 bar

Lecithin ratio (%w/v)	0	0.05	0.10	0.15	0.20
D_{32} (μm)	3.36 ± 0.13	3.48 ± 0.24	4.35 ± 0.08	5.89 ± 0.17	7.72 ± 0.32

This could be explained by reasons as follows, lecithin could increase the viscosity of the continuous phase, causing detached particle to agglomerate near the membrane surface, increasing probability of coalescence and thus larger droplet sizes (Lloyd et al. 2014). In addition, the high viscosity reduced the shear force inside pore and permeate flux. Lecithin could be adsorbed on membrane surface due to electrostatic attraction. It also could not pass through membrane because fouling or accumulation on the membrane surface reduced pore size. As a result, the high lecithin content increased the particle pulling force above the membrane surface, leading to a prolonged particle formation time, larger particle size, and thus increased particle coalescence in adjacent pores. Consequently, the higher the lecithin ratio, the larger the particle size.

3.3.2. Homogenization efficiency

Figure 8 shows the homogenization efficiency of WGO-in-water emulsion containing 10% v/v WGO at 9 bar and difference lecithin ratio. At 0 and 0.1% of lecithin ratio, the efficiency was not significant difference. However, lecithin rose from 0.1–0.2%, the efficiency value reduced from 98.31 to 97.68%. The results revealed that increasing the lecithin ratio reduced homogenization efficiency and thereby emulsion stability, in contrast to high pressure homogenization, increasing the emulsifier ratio increased emulsion stability (Salminen et al. 2020). The finding was completely consistent with the particle size distribution

results. Lecithin is a surfactant that aids in emulsification; however, the lecithin molecule has a positively charged end $-N(CH_3)_3^+$ or $-NH_3^+$, the cellulose acetate membrane are negatively charged ($-OH$, CH_3COO^-). Hence, lecithin was difficult to pass through the membrane, because the electrostatic attraction on the membrane. In addition, it also interfered with water and oil particles in dispersed phase, leading to a reduction in homogenization efficiency.

3.3.3. Permeate flux

The effect of lecithin ration on permeate flux in WGO-in-water homogenization by membrane at 9 bar, and 10%v/v of oil fraction was shown in Fig. 9. The permeate flux dropped dramatically from 105.98 to 77.45 $L.h^{-1}.m^{-2}$ when lecithin ratio increased from 0 to 0.2%. This is explained by an increase in viscosity, in conjunction with the electrostatic attraction caused by lecithin, which reduced the transmembrane dynamics of the components and increased clogging on the membrane surface and inside the capillary. As a result, the filtration efficiency decreased significantly, especially when the lecithin ratio was increased from 0.15 to 0.2%. The obtained results completely agree with the results on particle size distribution and stability of the emulsion (Ezzati et al. 2005)

To summarize, while lecithin is a surfactant that supports conventional homogenization processes, it did not support membrane homogenization of WGO-in-water, even at high ratios, causing membrane clogging and lowering process efficiency. Ezzati et al., 2005, found that increasing emulsifier from 0.2 to 0.8 vol%, the permeate flux also decreased when microfiltration oil-in-water emulsion.

4. Conclusion

The utilization of PESU membrane with pore size 0.45 μm for premix membrane homogenization of WGO-in-water emulsion was investigated in this research. The effect of operating pressure, WGO fraction and lecithin ration on the droplet size, stability of emulsion and permeate flux was researched. Under the same mixture emulsion, the higher operating pressure, the narrower the mean particle diameter and higher homogenization efficiency were obtained. While particle diameter increased and homogenization efficiency reduced as WGO fraction and lecithin ratio increased. The increase lecithin ratio reduced both, the homogenization and permeate flux. Besides that, it increased the diameter of particles in the premix membrane emulsification of WGO-in-water emulsions. The permeate flux dramatically increased with increase in operating pressure or decrease in oil fraction and lecithin ratios. The finding of this research revealed the potential of using a PESU membrane to homogenize WGO-in-water emulsions in particular, as well as the successful application of membrane emulsification in the food processing industry in general.

Declarations

Acknowledgement

We acknowledge the support of time and facilities from Ho Chi Minh City University of Technology (HCMUT), VNU-HCM for this study.

Declaration of interest

None

Ethical approval

Ethics approval was not required for this research.

Data Availability Statement

Data available on request from the authors.

Author Contribution Statement

Q.D.L and H.D.N designed and directed the project. Q.D.L. and N.T.T.D. wrote the main manuscript text. T.T.L.H. processed the experimental data, performed the analysis. All authors reviewed the manuscript.

References

1. Al-Rimawi, F., Alayoubi, M., Elama, Claude, Jazzar, M., & Çakıcı, A. (2020). Use of cinnamon, wheat germ, and eucalyptus oils to improve quality and shelf life of concentrated yogurt (Labneh). *Cogent Food & Agriculture*, *6*(1), 1807810.
2. Alliod, O., Valour, J.-P., Urbaniak, S., Fessi, H., Dupin, D., & Charcosset, C. (2018). Preparation of oil-in-water nanoemulsions at large-scale using premix membrane emulsification and Shirasu Porous Glass (SPG) membranes. *Colloids and surfaces a: physicochemical and engineering aspects*, *557*, 76–84.
3. Aserin, A. (2007). *Multiple emulsion: technology and applications* (Vol. 1). John Wiley & Sons.
4. Barros, J. C., Munekata, P. E. S., de Carvalho, F. A. L., Domínguez, R., Trindade, M. A., Pateiro, M., & Lorenzo, J. M. (2021). Healthy beef burgers: Effect of animal fat replacement by algal and wheat germ oil emulsions. *Meat Science*, *173*, 108396.
5. Berendsen, R., Güell, C., Henry, O., & Ferrando, M. (2014). Premix membrane emulsification to produce oil-in-water emulsions stabilized with various interfacial structures of whey protein and carboxymethyl cellulose. *Food Hydrocolloids*, *38*, 1–10.
6. Bodewes, F. A. J. A., Wouthuyzen-Bakker, M., & Verkade, H. J. (2015). Chapter 41 - Persistent Fat Malabsorption in Cystic Fibrosis. In R. R. B. T.-D. and E. in C. F. Watson (Ed.), (pp. 373–381). Boston: Academic Press. <https://doi.org/10.1016/B978-0-12-800051-9.00041-9>
7. Boukid, F., Folloni, S., Ranieri, R., & Vittadini, E. (2018). A compendium of wheat germ: Separation, stabilization and food applications. *Trends in Food Science & Technology*, *78*, 120–133.

8. Brandolini, A., & Hidalgo, A. (2012). Wheat germ: not only a by-product. *International journal of food sciences and nutrition*, *63*(sup1), 71–74.
9. Ceylan, Z., Meral, R., Kose, Y. E., & Cavidoglu, I. (2020). Wheat germ oil nanoemulsion for oil stability of the cooked fish fillets stored at 4° C. *Journal of food science and technology*, *57*(5), 1798–1806.
10. Charcosset, C, Limayem, I., & Fessi, H. (2004). The membrane emulsification process—a review. *Journal of Chemical Technology & Biotechnology: International Research in Process, Environmental & Clean Technology*, *79*(3), 209–218.
11. Charcosset, Catherine. (2009). Preparation of emulsions and particles by membrane emulsification for the food processing industry. *Journal of Food Engineering*, *92*(3), 241–249.
12. Charcosset, Catherine. (2021). Classical and recent applications of membrane processes in the food industry. *Food Engineering Reviews*, *13*(2), 322–343.
13. Choi, B.-S., & Kang, K.-O. (2009). Studies on the analysis of physiological and antimicrobial activity of wheat germ. *Journal of the East Asian Society of Dietary Life*, *19*(4), 585–592.
14. Choi, S. J., & McClements, D. J. (2020). Nanoemulsions as delivery systems for lipophilic nutraceuticals: Strategies for improving their formulation, stability, functionality and bioavailability. *Food science and biotechnology*, *29*(2), 149–168.
15. Consoli, L., Hubinger, M. D., & Dragosavac, M. M. (2020). Encapsulation of resveratrol using Maillard conjugates and membrane emulsification. *Food Research International*, *137*, 109359. <https://doi.org/https://doi.org/10.1016/j.foodres.2020.109359>
16. Dunford, N. T. (2009). Wheat germ oil. In *Gourmet and Health-Promoting Specialty Oils* (pp. 359–376). Elsevier.
17. Ezzati, A., Gorouhi, E., & Mohammadi, T. (2005). Separation of water in oil emulsions using microfiltration. *Desalination*, *185*(1–3), 371–382.
18. Floury, J., Desrumaux, A., & Lardières, J. (2000). Effect of high-pressure homogenization on droplet size distributions and rheological properties of model oil-in-water emulsions. *Innovative Food Science & Emerging Technologies*, *1*(2), 127–134.
19. Galanakis, C. M. (2019). *Nutraceuticals and natural product pharmaceuticals*. Academic Press.
20. Ghafoor, K., Özcan, M. M., AL-Juhaimi, F., Babiker, E. E., Sarker, Z. I., Ahmed, I. A. M., & Ahmed, M. A. (2017). Nutritional composition, extraction, and utilization of wheat germ oil: a review. *European Journal of Lipid Science and Technology*, *119*(7), 1600160.
21. Harrabi, S., Ferchichi, A., Fellah, H., Feki, M., & Hosseinian, F. (2021). Chemical Composition and in vitro Anti-inflammatory Activity of Wheat Germ Oil Depending on the Extraction Procedure. *Journal of Oleo Science*, *70*(8), 1051–1058.
22. Jha, P. K., Kudachikar, V. B., & Kumar, S. (2013). Lipase inactivation in wheat germ by gamma irradiation. *Radiation Physics and Chemistry*, *86*, 136–139.
23. Jiang, T., Liao, W., & Charcosset, C. (2020). Recent advances in encapsulation of curcumin in nanoemulsions: A review of encapsulation technologies, bioaccessibility and applications. *Food*

- Research International, 132, 109035. <https://doi.org/https://doi.org/10.1016/j.foodres.2020.109035>
24. Joscelyne, S. M., & Trägårdh, G. (2000). Membrane emulsification—a literature review. *Journal of Membrane Science*, 169(1), 107–117.
 25. Karadeniz, M., Sahin, S., & Sumnu, G. (2018). Enhancement of storage stability of wheat germ oil by encapsulation. *Industrial crops and products*, 114, 14–18.
 26. Koba, K., & Yanagita, T. (2014). Health benefits of conjugated linoleic acid (CLA). *Obesity research & clinical practice*, 8(6), e525–e532.
 27. Köse, Y. E. (2021). Degradation kinetic modeling of bioactive compounds and enzyme activity in wheat germ during stabilization. *LWT*, 112501.
 28. Lloyd, D. M., Norton, I. T., & Spyropoulos, F. (2014). Processing effects during rotating membrane emulsification. *Journal of Membrane Science*, 466, 8–17.
 29. Major-Godlewska, M. (2019). Evaluation of drops dimensions in time and rheological properties of the multiple emulsion. *Chemical Papers*, 73(8), 2073–2080.
 30. Manga, M. S., & York, D. W. (2017). Production of concentrated Pickering emulsions with narrow size distributions using stirred cell membrane emulsification. *Langmuir*, 33(36), 9050–9056.
 31. Megahed, M. G. (2011). Study on stability of wheat germ oil and lipase activity of wheat germ during periodical storage. *Agric Biol JN Am*, 2(1), 163–168.
 32. Nazir, A., Maan, A. A., Sahin, S., Boom, R. M., & Schroën, K. (2015). Foam preparation at high-throughput using a novel packed bed system. *Food and Bioproducts Processing*, 94, 561–564.
 33. Nazir, A., Schroën, K., & Boom, R. (2011). High-throughput premix membrane emulsification using nickel sieves having straight-through pores. *Journal of membrane science*, 383(1–2), 116–123.
 34. Piacentini, E, Bazzarelli, F., Drioli, E., & Giorno, L. (2020). Advances in Membrane Emulsification and Membrane Nanoprecipitation Using Membrane Contactors: State-of-the-Art and Perspectives. In *Hollow Fiber Membrane Contactors* (pp. 143–158). CRC Press.
 35. Piacentini, Emma, & Giorno, L. (2016). Emulsification by Membrane Operations BT - Encyclopedia of Membranes. In E. Drioli & L. Giorno (Eds.), (pp. 675–678). Berlin, Heidelberg: Springer Berlin Heidelberg. https://doi.org/10.1007/978-3-662-44324-8_1233
 36. Ramakrishnan, S., Ferrando, M., Aceña-Muñoz, L., De Lamo-Castellví, S., & Güell, C. (2013). Fish oil microcapsules from O/W emulsions produced by premix membrane emulsification. *Food and bioprocess technology*, 6(11), 3088–3101.
 37. Ransmark, E., Svensson, B., Svedberg, I., Göransson, A., & Skoglund, T. (2019). Measurement of homogenisation efficiency of milk by laser diffraction and centrifugation. *International Dairy Journal*, 96, 93–97.
 38. Salminen, H., Bischoff, S., & Weiss, J. (2020). Formation and stability of emulsions stabilized by Quillaja saponin–egg lecithin mixtures. *Journal of food science*, 85(4), 1213–1222.
 39. Schröder, V., Behrend, O., & Schubert, H. (1998). Effect of dynamic interfacial tension on the emulsification process using microporous, ceramic membranes. *Journal of colloid and interface*

- science, 202(2), 334–340.
40. Schultz, S., Wagner, G., Urban, K., & Ulrich, J. (2004). High-pressure homogenization as a process for emulsion formation. *Chemical Engineering & Technology: Industrial Chemistry-Plant Equipment-Process Engineering-Biotechnology*, 27(4), 361–368.
 41. Spasic, A. M., & Hsu, J.-P. (2005). *Finely Dispersed Particles: micro-, nano-, and atto-engineering*. CRC Press.
 42. Spyropoulos, F., Lloyd, D. M., Hancocks, R. D., & Pawlik, A. K. (2014a). Advances in membrane emulsification. Part A: recent developments in processing aspects and microstructural design approaches. *Journal of the Science of Food and Agriculture*, 94(4), 613–627.
 43. Spyropoulos, F., Lloyd, D. M., Hancocks, R. D., & Pawlik, A. K. (2014b). Advances in membrane emulsification. Part A: Recent developments in processing aspects and microstructural design approaches. *Journal of the Science of Food and Agriculture*, 94(4), 613–627.
<https://doi.org/10.1002/jsfa.6444>
 44. Šramková, Z., Gregová, E., & Šturdík, E. (2009). Chemical composition and nutritional quality of wheat grain. *Acta chimica slovacica*, 2(1), 115–138.
 45. Surh, J., Jeong, Y. G., & Vladisavljević, G. T. (2008). On the preparation of lecithin-stabilized oil-in-water emulsions by multi-stage premix membrane emulsification. *Journal of Food Engineering*, 89(2), 164–170. <https://doi.org/10.1016/j.jfoodeng.2008.04.023>
 46. Suzuki, K., Fujiki, I., & Hagura, Y. (1998). Preparation of corn oil/water and water/corn oil emulsions using PTFE membranes. *Food Science and Technology International, Tokyo*, 4(2), 164–167.
 47. SUZUKI, K., SHUTO, I., & HAGURA, Y. (1996). Characteristics of the membrane emulsification method combined with preliminary emulsification for preparing corn oil-in-water emulsions. *Food Science and Technology International, Tokyo*, 2(1), 43–47.
 48. Thiex, N. J., Anderson, S., Gildemeister, B., & M Collaborators: Adcock W., Boedigheimer J., Bogren E., Coffin R., Conway K., DeBaker A., Frankenius E., Gramse M., Hogan P., Knese T., MacDonald J., Möler J., Royle R., Russell M., Shafiee F., Shreve B., Sieh J., Spann M., Töpler E., W. M. (2003). Crude fat, hexanes extraction, in feed, cereal grain, and forage (Randall/Soxtec/submersion method): collaborative study. *Journal of AOAC International*, 86(5), 899–908.
 49. Türkoğlu, G. C., Sarıışık, M., Karavana, S. Y., & Köse, F. A. (2021). Production of wheat germ oil containing multilayer hydrogel dressing. *Carbohydrate Polymers*, 118287.
 50. Van der Graaf, S., Schroën, C., Van der Sman, R. G. M., & Boom, R. M. (2004). Influence of dynamic interfacial tension on droplet formation during membrane emulsification. *Journal of colloid and interface science*, 277(2), 456–463.
 51. Vladisavljević, G. T. (2019a). Preparation of microemulsions and nanoemulsions by membrane emulsification. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 579, 123709.
 52. Vladisavljević, G. T. (2019b). Membrane emulsification in pharmaceuticals and biotechnology. In *Current Trends and Future Developments on (Bio-) Membranes* (pp. 167–222). Elsevier.

53. Vladisavljević, G. T., Shimizu, M., & Nakashima, T. (2004). Preparation of monodisperse multiple emulsions at high production rates by multi-stage premix membrane emulsification. *Journal of Membrane Science*, *244*(1–2), 97–106.
54. Vladisavljević, G. T., & Williams, R. A. (2005). Recent developments in manufacturing emulsions and particulate products using membranes. *Advances in colloid and interface science*, *113*(1), 1–20.
55. Wang, J., Tang, J., Ruan, S., Lv, R., Zhou, J., Tian, J., et al. (2021). A comprehensive review of cereal germ and its lipids: Chemical composition, multi-objective process and functional application. *Food Chemistry*, 130066.
56. Yan, B., Park, S. H., & Balasubramaniam, V. M. (2017). Influence of high pressure homogenization with and without lecithin on particle size and physicochemical properties of whey protein-based emulsions. *Journal of Food Process Engineering*, *40*(6), e12578.
57. Zanatta, V., Rezzadori, K., Penha, F. M., Zin, G., Lemos-Senna, E., Petrus, J. C. C., & Di Luccio, M. (2017). Stability of oil-in-water emulsions produced by membrane emulsification with microporous ceramic membranes. *Journal of Food Engineering*, *195*, 73–84.
58. Zhu, K.-X., Lian, C.-X., Guo, X.-N., Peng, W., & Zhou, H.-M. (2011). Antioxidant activities and total phenolic contents of various extracts from defatted wheat germ. *Food Chemistry*, *126*(3), 1122–1126.

Figures

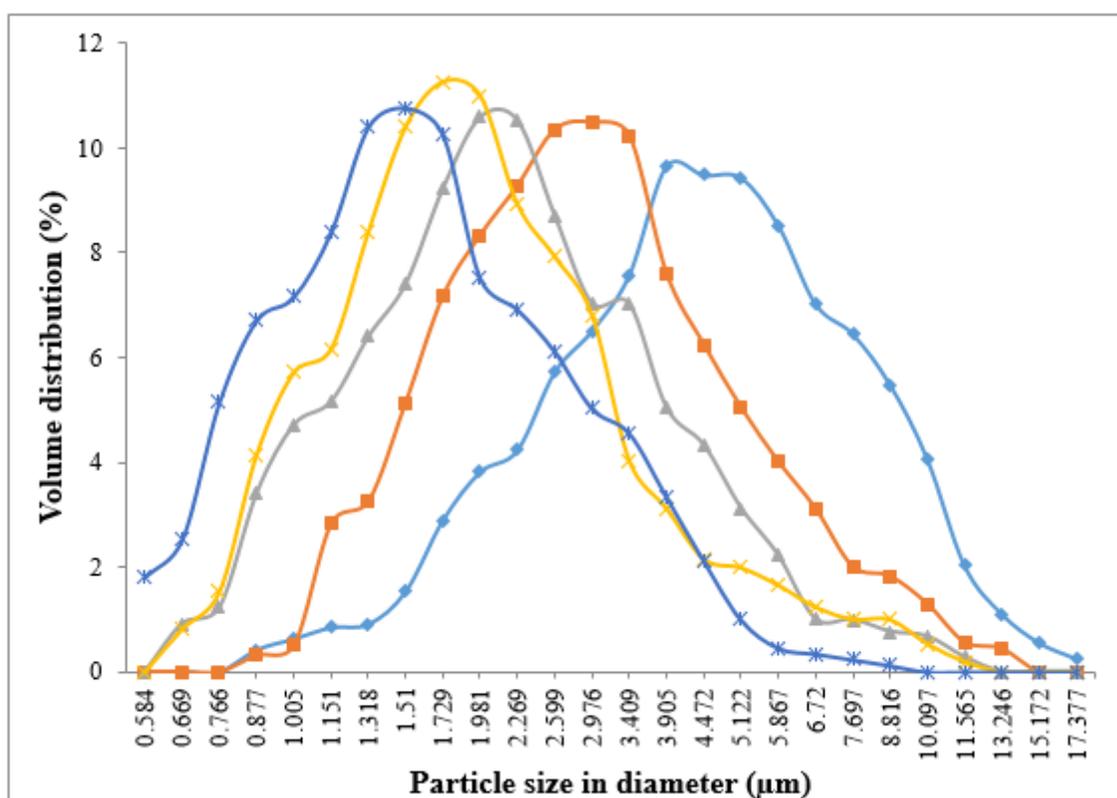


Figure 1

Effect of homogenization pressure on particle size distribution of emulsions containing 10%v/v WGO, 0.1% w/v lecithin

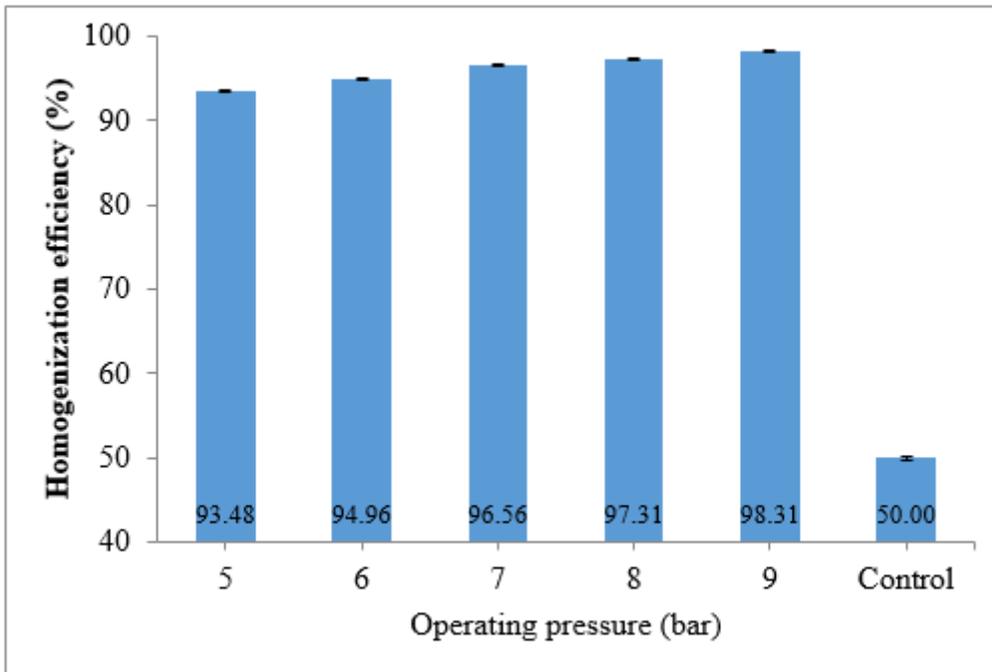


Figure 2

Effect of homogenization pressure on homogenization efficiency of emulsions containing 10%v/v WGO, 0.1% w/v lecithin

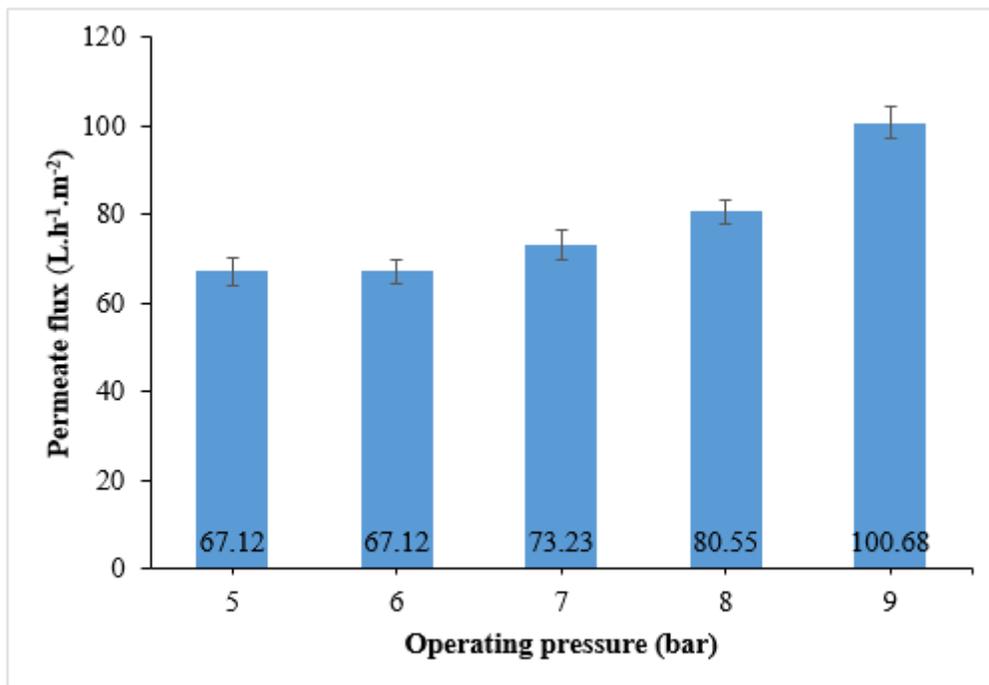


Figure 3

Effect of operating pressure on permeate flux in homogenization of emulsions containing 10%v/v WGO, 0.1%

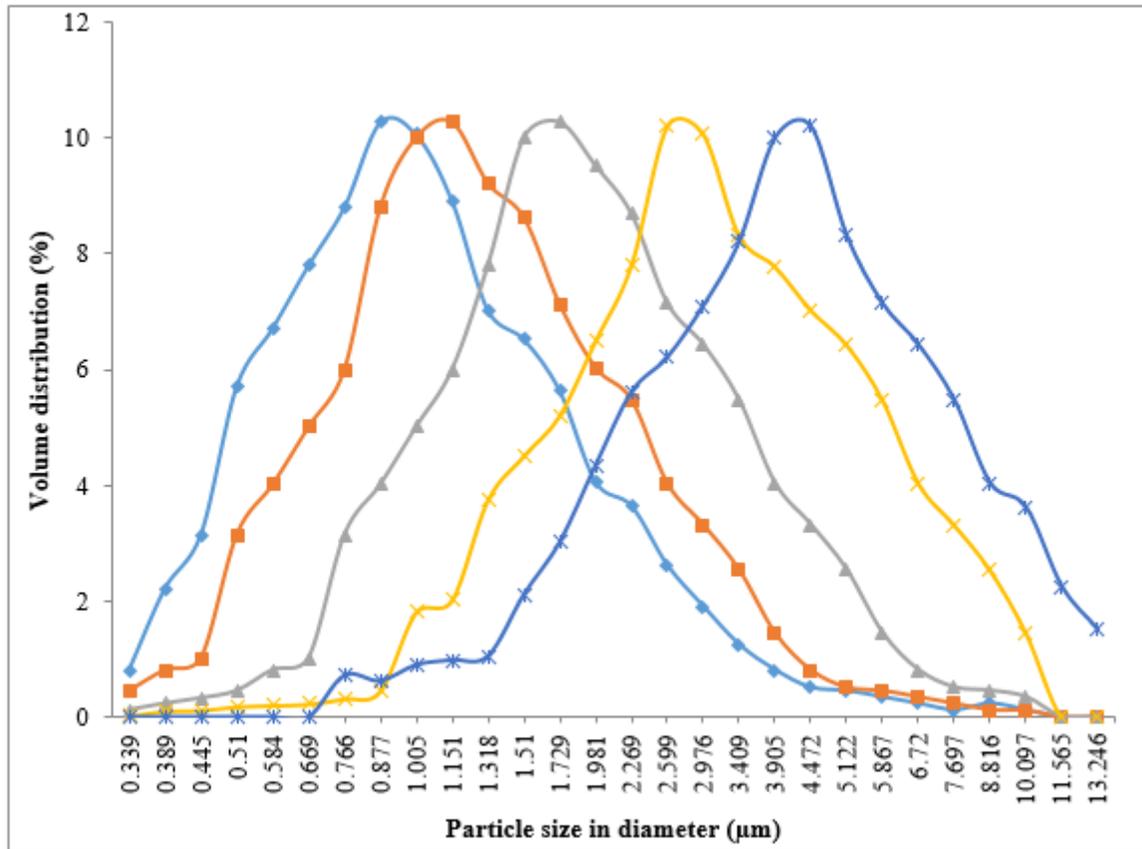


Figure 4

Effect of oil fraction (% v/v) on particle size distribution in homogenization of emulsions containing 0.1% w/v lecithin at 9 bar w/v lecithin

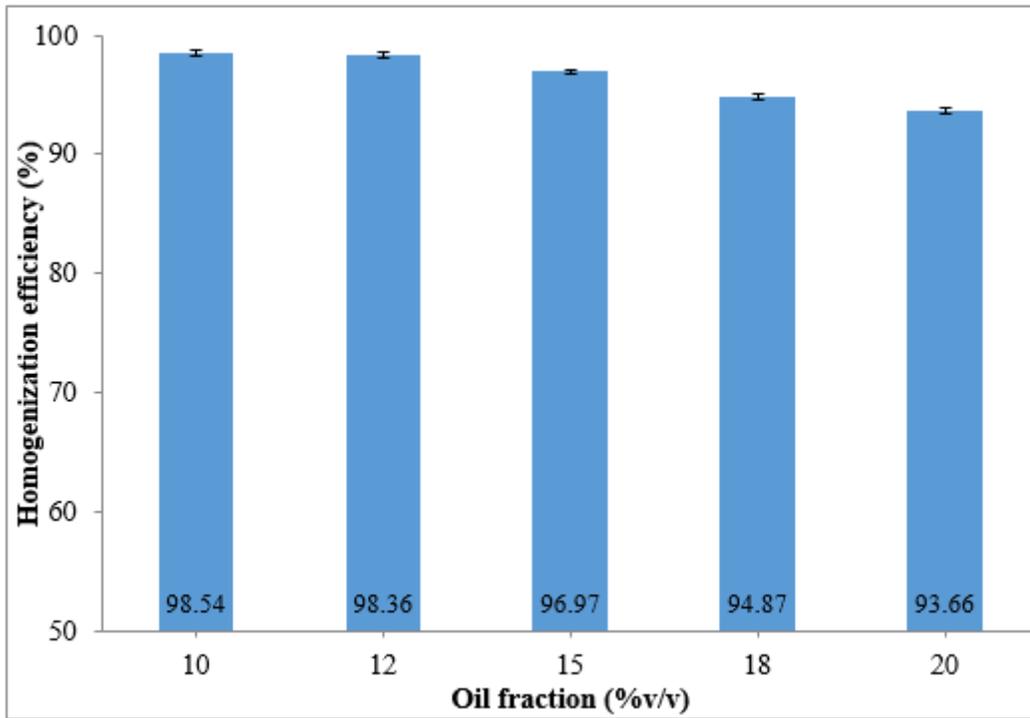


Figure 5

Effect of oil fraction (%v/v) on efficiency in homogenization of emulsions containing 0.1% w/v lecithin at 9 bar

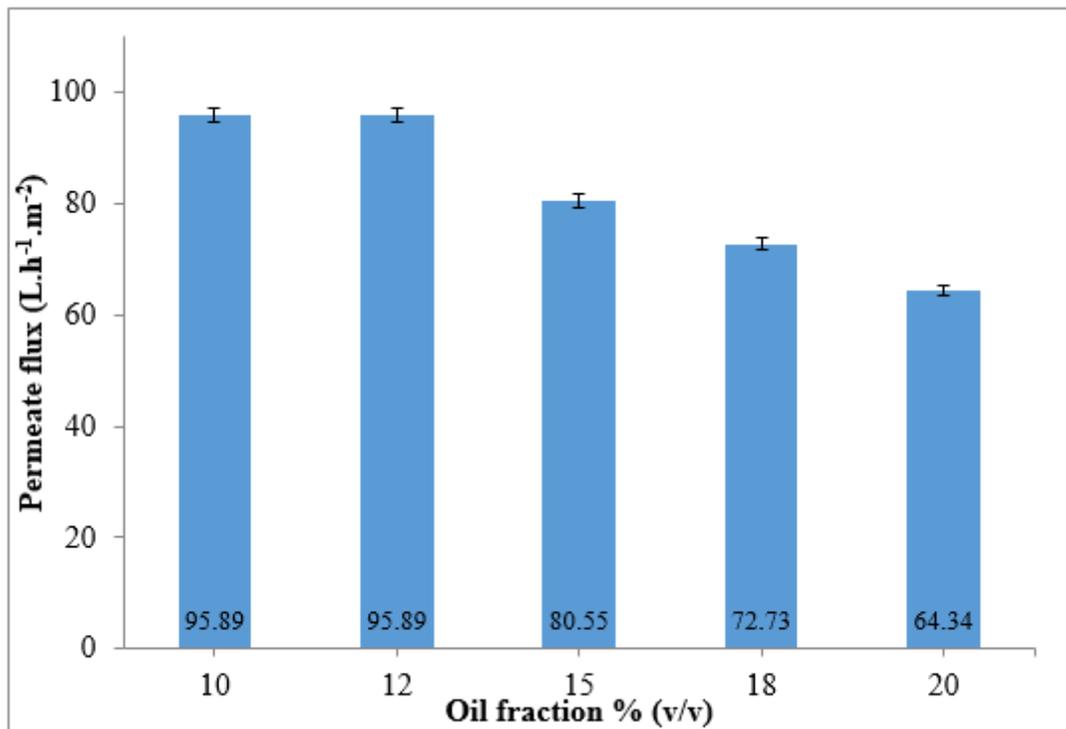


Figure 6

Effect of oil fraction (%v/v) on permeate flux in homogenization of emulsions containing 0.1% w/v lecithin at 9 bar

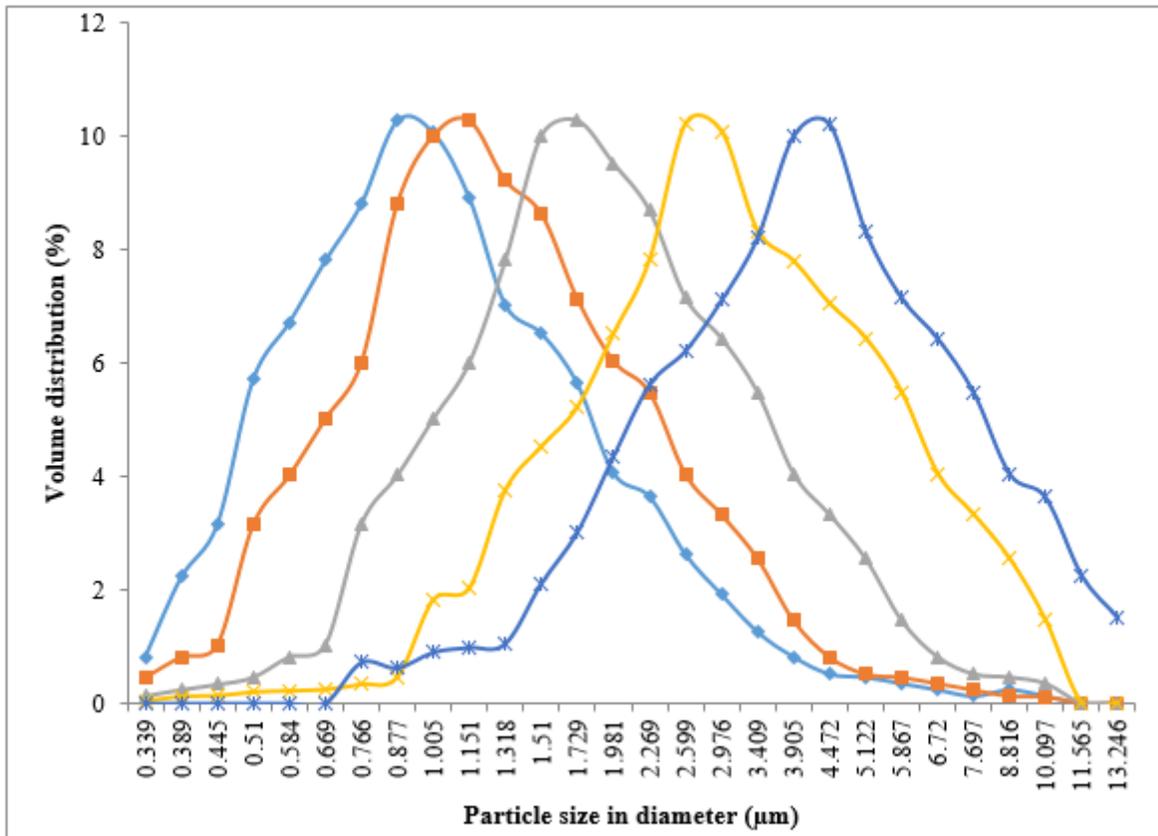


Figure 7

Effect of lecithin ratio (%w/v) on particle size distribution in homogenization of emulsions containing 10% v/v WGO at 9 bar

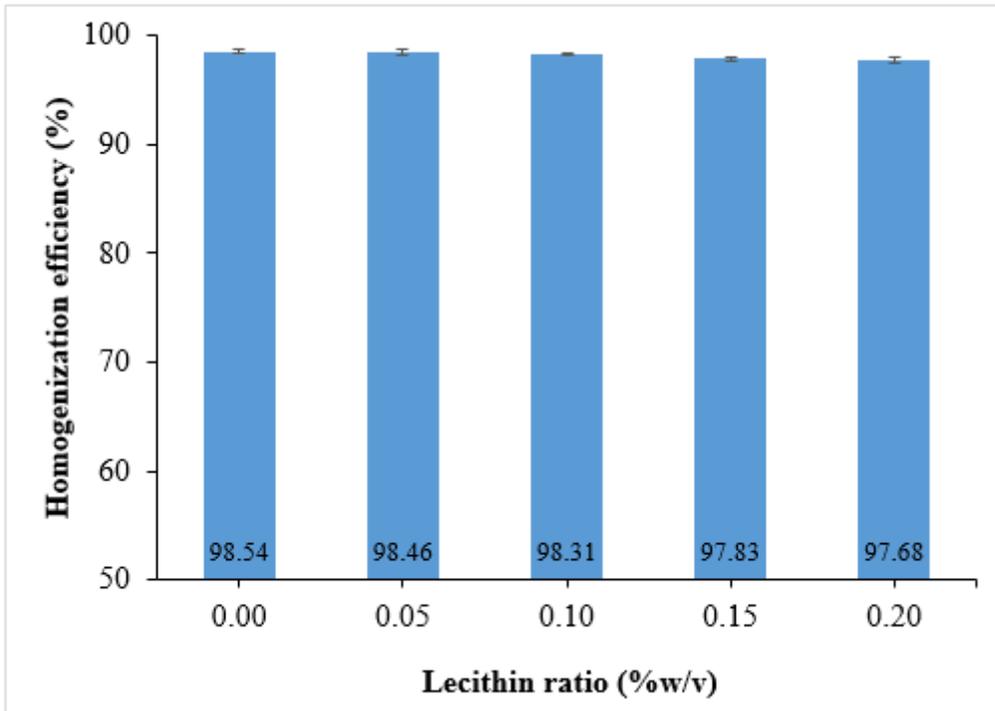


Figure 8

Effect of lecithin ratio (%w/v) on efficiency in homogenization of emulsions containing 10% v/v WGO at 9 bar

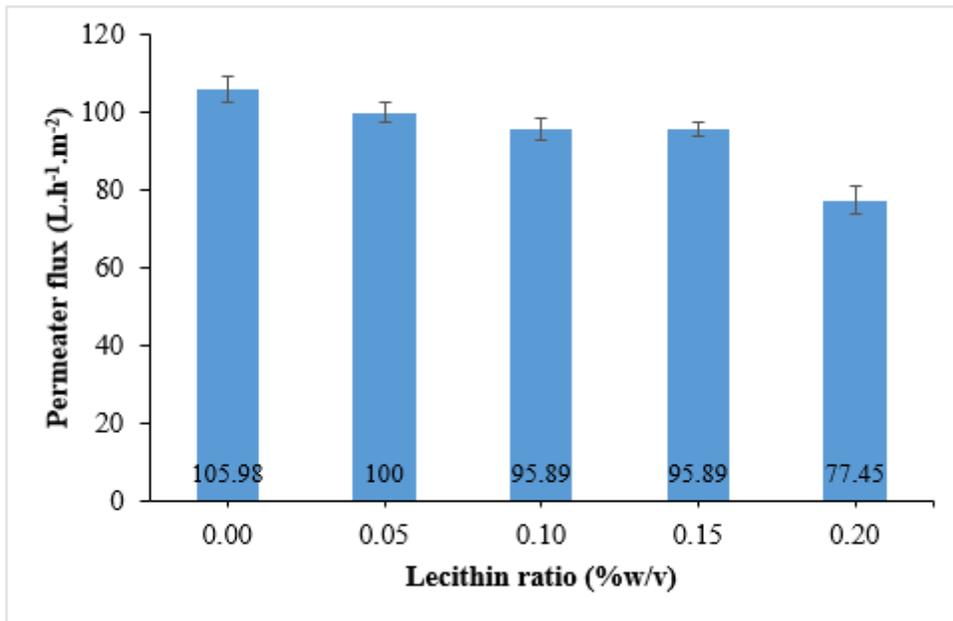


Figure 9

Effect of lecithin ratio (%w/v) on permeate flux in homogenization of emulsions containing 10% v/v WGO at 9 bar