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Effect of sesamol on the physical and chemical stability of plant-based flaxseed oil-in-water emulsions stabilized by proteins or phospholipids

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Abstract

Plant-based polyphenols are increasingly being explored as functional ingredients in emulsified food systems. In this study, the effects of sesamol on the physical and chemical stability of flaxseed oil-in-water emulsions stabilized by either phospholipids (sunflower) or proteins (whey or pea) were investigated. In the absence of sesamol, the protein-based emulsions displayed better physical stability than the phospholipid-based ones, which was related to their smaller particle diameter and higher particle charge. For the phospholipid-based emulsions, sesamol addition did not improve their physical stability, but it did inhibit lipid oxidation. In particular, it decreased the formation of secondary oxidation products, with a 65% reduction in TBARs formation compared to the control after 8 days storage. For the protein-based emulsions, sesamol addition reduced particle aggregation and inhibited lipid oxidation, reducing the secondary oxidation products by around 85% after 19 days storage. The inhibitory efficiency of sesamol in the pea protein-based emulsions was comparable to that in the whey protein-based ones. The effects of sesamol on the physical and chemical stability of the emulsions were related to its partitioning between the oil, water, and interfacial layers. This study suggests that adding sesamol to plant-based emulsions may improve their physical and chemical stability, thereby extending their shelf life.

Keywords: sesamol, sunflower phospholipids, whey protein isolate, pea protein, oil-in-water emulsions, stability

1. Introduction

Food manufacturers are developing an increasing number of commercial products from plant-derived ingredients due to environmental, health, and ethical reasons. Flaxseed oil contains high levels of plant-based omega-3 fatty acids (α-linolenic acid, ALA), whose consumption has been linked to a reduction in obesity-related chronic diseases [1]. Oil-in-water emulsions are particularly suitable vehicles for incorporating ALA into a diverse range of food products because of their good water-dispersibility, high bioavailability, and design flexibility [2]. However, emulsified polyunsaturated lipids are prone to rapid oxidation because of their high specific surface areas [3]. The rate of lipid oxidation can, however, be reduced by careful selection of the emulsifier used to coat the lipid droplets, as well as by the addition of appropriate antioxidants [4, 5].

Plant-based phospholipids are natural surface-active substances that have been shown to be suitable for forming and stabilizing emulsions by inhibiting droplet coalescence through a combination of steric and electrostatic effects [6]. The ability of these plant-based phospholipids to form and stabilize emulsions depends on their chemical structure, *e.g.,* head and tail groups type [2, 7-9]. Sunflower phospholipids have been reported to have comparable emulsifying properties as soybean phospholipids [10].

However, sunflower phospholipids have an advantage for some applications because they are produced from a non-genetically modified source and are not a common source of allergens [8, 11].

Many food proteins are also an important source of plant-based emulsifiers because of their natural amphiphilic properties. Pea protein has been shown to exhibit similar emulsifying properties to whey protein isolate (WPI), is not considered to be a major food allergen, and can be used in plant-based foods [12]. Amphiphilic proteins adsorb to oil droplet surfaces during homogenization, and then stabilize them by forming relatively thick and charged coatings that produce strong steric and electrostatic repulsive forces [13]. Moreover, some proteins can improve the chemical stability of emulsions by inhibiting lipid oxidation, which has been attributed to their free radical scavenging and transition metal chelating properties [5, 14]. The chemical stability of emulsions can also be enhanced by adding plant-based antioxidants, such as some polyphenols [15]. Polyphenols naturally contain structural motifs that exhibit antioxidant activity, such as catechol moieties and hydroxyl groups. Polyphenols can be made to reside at the oil-water interface (where lipid oxidation usually occurs) *via* physical interactions with adsorbed emulsifiers[16, 17]. For instance, it has been reported that the hydroxyl groups on polyphenols form hydrogen bonds with the oxygen groups

on phospholipid headgroups, thereby causing them to become located at the oil-water interface [18]. Once present at an interface, polyphenols can inhibit lipid oxidation reactions that occur there [19]. Many polyphenols can also bind to proteins through physical or chemical interactions, such as hydrogen bonding, hydrophobic attraction, and covalent bonding [20]. In addition, the presence of the polyphenols may promote conformational changes in the protein molecules, which can improve their emulsification and antioxidant properties [21].

Sesamol is a plant-based polyphenol that has been reported to exhibit biological activities that could improve human health, such as inhibition of lipid oxidation and DNA cleavage [22]. The antioxidant activity of sesamol is mainly attributed to the presence of a phenolic group attached to a benzodioxole group [23]. Studies have shown that sesamol exhibits comparable or better antioxidant activity than a synthetic (tert-butylhydroquinone, TBHQ) and another natural (rosemary extract) antioxidant [24, 25]. Other studies have shown that the anti-inflammatory activity, bioavailability, and stability of sesamol were improved by encapsulating it within phosphatidylcholine mixed micelles or colloidal delivery systems [23, 26]. In our previous research, it was found that sesamol could significantly improve the physical and chemical stability of emulsions formulated using a plant-based surfactant (quillaja saponin), which was

attributed to its ability to strengthen the interfacial layer [27]. In the current study, our aim was to assess whether sesamol could also improve the stability of plant-based emulsions stabilized by other emulsifiers: proteins and phospholipids.

The effects of sesamol on the physical and chemical stability of flaxseed oil-in-water emulsions formulated using four kinds of natural emulsifier were examined: two sunflower phospholipids (Sunlipon 90 and Sunlipon 65), a plant-based protein (pea protein), and a widely used animal-based protein (whey protein). Initially, the ability of these emulsifiers to form and stabilize the emulsions were compared. Then, the potential mechanism of interaction between the sesamol and the emulsifiers was determined. Finally, links between interfacial and physicochemical properties were established to provide guidelines for the application of sesamol in plant-based emulsified foods.

2 Materials and methods

2.1 Materials

Flaxseed oil was purchased from Hongjingyuan Oil Co., Ltd (Xilingol, China). Sunlipon 90 (S90) and Sunlipon 65 (S65) extracted from sunflower oil were kindly provided by Perimondo (New York, NY, USA). Whey protein isolate (WPI, protein content of 92%) and pea protein (PP, protein content of 55%) were obtained from

Agropur Coopérative (Saint-Hubert, Longueuil, Canada) and Ingredion, Inc. (Bridgewater, NJ), respectively. Sesamol (98%) and all chemicals, including 2,2´-azobis-2-methylpropanimidamide dihydrochloride (AAPH), 2-thiobarbituric acid (TBA), trichloroacetic acid (TCA), 1,1,3,3-tetramethoxypropane (TMP), ethylenediaminetetraacetic acid (EDTA), sodium azide, and cumene hydroperoxide, were of analytical grade and purchased from Sigma-Aldrich (Saint Louis, MO, USA). A peroxyl radical sensitive dye (BODIPY® 665/676) was obtained from Invitrogen Incorporated (Carlsbad, CA). Water purified by a Milli-Q system was utilized for all the experiments. Information about the oil-water partition coefficient (log*P*) and topological polar surface area (TPSA) of the sesamol were calculated using ChemBioFinder.

2.2 Preparation of emulsions

Oil-in-water emulsions were prepared according to a previous report [28]. Briefly, aqueous phases were prepared by dissolving the emulsifiers (S65, S90, PP, or WPI) at a concentration of 2% (w/w) in 10 mM phosphate buffer solution (PBS, pH 7.0) under continuous stirring at room temperature (25°C). Oil phases were prepared by mixing sesamol (400 μM) and flaxseed oil together. The sesamol was first dissolved in chloroform, then flaxseed oil was added and the mixture was stirred to ensure

dissolution. Coarse emulsions with an oil fraction of 10% (w/w) were fabricated by blending the aqueous phase and oil phase together using a high-speed mixing device (IKA, T25, Germany) at 10,000 rpm for 2 min. These emulsions were then passed three times through a microfluidizer (M-110EH30, Microfluidics, Newton, MA) at an operating pressure of 12,000 psi.

2.3 Accelerated storage experiment

Freshly prepared emulsions were rapidly transferred into screw-capped glass vials. Storage stability studies were then performed by incubating the emulsions at 55℃ in the dark. During storage, samples were periodically collected and used for further analysis.

2.4 Particle size and ζ-potential measurement

Particle size ($d_{3,2}$) and electrical charge (ζ-potential) of emulsions were measured by static light scattering (SLS, Mastersizer 2000, Malvern Instruments, Westborough, MA) and phase-analysis light scattering (ZetasizerNanoZS, Malvern Instruments, Worcestershire, U.K.), respectively. Refractive indexes of the flaxseed oil and aqueous phase used in the calculations were 1.490 and 1.330, respectively [29].

2.5 Morphology observation

Emulsion morphology was determined by confocal laser scanning microscopy

(CLSM, Nikon D-Eclipse C1 80i, Nikon, Melville, NY). A mixture of emulsion and phosphate buffer solution (v/v, 1:1) was dyed with Nile red dissolved in ethanol solution (1 mg/mL). Confocal microscopy mages were then captured and processed using the instrument software program (NISElements, Nikon, Melville, NY).

2.6 Laser scanning profiling

The physical stability of emulsions during storage at 55℃ was monitored using a Turbiscan Lab Expert stability analyzer (Formulaction, France) according to a method reported previously [30]. The turbiscan stability index (TSI) of the emulsions was calculated at each storage time from the raw data using the computer program connected to the instrument (Turbiscan Easy Soft).

2.7 Primary and secondary oxidation products analysis

The peroxide value (PV) of each emulsion was measured to quantify the amount of primary oxidation products present during storage using a solvent extraction/colorimetric method described in detail previously [31, 32]. The hydroperoxide concentrations were calculated using a cumene hydroperoxide standard curve. All values are expressed as mmol cumene hydroperoxide equivalents per kg of oil.

The secondary oxidation products were characterized by determining the concentration of TBARS using a colorimetric method described previously [28]. The

concentrations of TBARS are expressed as 1,1,3,3-tetraethoxypropane (TEP) equivalents per kg oil (mmol).

2.8 Analysis of sesamol concentration in aqueous phase of emulsions

Changes in the concentration of sesamol in the aqueous phase of the emulsions over time were determined according to a method described previously with some slight modifications [28, 33]. Briefly, 1 mL of emulsion was centrifuged at 50,377 g for 1 h at 4℃. The aqueous phase was then collected carefully using a needle and syringe. The sesamol concentration in the aqueous phase was then detected by liquid chromatography. The normalized concentration in the aqueous phase was then calculated using equation (1):

$$\text{Normalized aqueous phase concentration} = \frac{C(t)}{C_0} \quad (1)$$

Here, C_0 is the initial concentration of sesamol added to the emulsions, and *C(t)* is the sesamol concentration present in the aqueous phase at a particular storage time (*t*).

2.9 Interfacial tension measurement

The dynamic interfacial tension at an oil-water interface, which was formed by a pendant water drop submerged into a cuvette filled with flaxseed oil, was recorded using a drop profile tensiometer (Tracker, Teclis Technologies, France) based on a method described previously [34]. The same emulsifier (2%, w/w in the water phase) and

sesamol (400 μM in the oil phase) concentrations as were used in the emulsions were utilized to measure the interfacial tension. The measurement chamber was directly connected to a syringe with a plunger through a screw thread and the water drop volume was controlled and monitored using a video camera. All of the experiments were performed at 25°C. The dynamic interfacial tension was monitored for 3600 s, while the area of the drop was kept at an appropriate size. The values of the interfacial tension (γ) were calculated by analyzing the shape of the pendent drop according to the Gauss-Laplace equation.

2.10 Free radical permeation assays

The measurement of peroxyl radical permeation from the aqueous to the oil phase in the emulsions was carried out using a method described previously [35]. A stock dye solution (5 mg/mL) was formed by dissolving peroxyl radical sensitive dye (BODIPY® 665/676) in chloroform (10-fold dilution) to obtain a working dye solution. After adding the working dye solution to flaxseed oil, a final dye with a concentration of 25 μg/g in oil was obtained. An O/W emulsion was prepared using the approach described in Section 2.2.

An AAPH system was used to generate peroxyl free radicals in the aqueous phase [36]. Emulsions and AAPH were mixed together and then placed in a 96-well plate along

with the controls (emulsions mixed with an equal volume of phosphate buffer). Changes in the fluorescence intensity of the BODIPY were then measured using a plate-reader (Spectramax M2, Molecular Devices, Carlsbad, CA). The excitation and emission wavelengths for the fluorescence measurements were 620 nm and 675 nm, respectively. Equation (2) was used to calculate the relative fluorescence intensity as a percentage:

$$\text{Relative fluorescence intensity} = \frac{I_{t\ AAPH}/I_{0\ AAPH}}{I_{t\ control}/I_{0\ control}} \times 100 \quad (2)$$

Here, $I_{t\ \text{AAPH}}$ and $I_{t\ \text{control}}$ are the fluorescence intensities of the emulsion after treatment with AAPH or buffer solution (control) for *t* minutes, respectively. Similarly, $I_{0\ \text{AAPH}}$ and $I_{0\ \text{control}}$ are the fluorescence intensities of the emulsion immediately after the addition of AAPH or buffer solution (t = 0 min).

2.11 Statistical analysis

All data are reported as mean ± standard deviations. Significant differences ($P < 0.05$) between the results were calculated by analysis of variance (ANOVA) and Tukey's test using SPSS 21.0 software (SPSS, Inc., Chicago, IL, USA).

3. Results and discussion

3.1 Particle size and ζ-potential

The effects of sesamol on mean particle diameter (d(3,2)) and electrical charge (ζ-potential) of the flaxseed oil-in-water emulsions are presented in **Fig. 1**. The mean

diameters of the droplets prepared using sunflower phospholipids (~0.34 μm) were appreciably larger than those prepared using proteins (~0.15μm) on the initial day (0 d) and throughout storage. Interestingly, the incorporation of sesamol into the S90-emulsions significantly decreased the initial mean droplet diameter (~35%). This result suggests that the presence of sesamol may have promoted droplet formation inside the homogenizer or inhibited droplet aggregation after homogenization [37]. The presence of sesamol did not appear to have a significant inhibitory effect on particle growth during storage, which suggests that it may have facilitated the formation of small droplets during homogenization, rather than inhibiting aggregation. This may have been due to its ability to reduce the interfacial tension (see later). Interestingly, there was an appreciable decrease in the mean droplet diameter for the phospholipid-emulsions after prolonged storage (Fig. 1A). This effect can be attributed to coalescence and oiling-off of the oil droplets (see later). Specifically, coalescence leads to an increase in oil droplet size, which promotes further coalescence (since the coalescence rate tends to increase with increasing droplet size), eventually leading to the formation of a separate oil layer on top of the emulsions. As a result, there may have been a greater fraction of smaller droplets remaining in the emulsions after the larger droplets coalesced and formed a separate oil layer. Thus, the particle size measured by light scattering only reflected that

of the smaller oil droplets remaining in the emulsions (which were more resistant to coalescence), rather than the true overall droplet size.

As shown in **Fig. 1B**, all the oil droplets had a negative charge, but the magnitude of the initial ζ-potential depended on emulsifier type: WPI > PP > S65 > S90. The S65 ingredient has been reported to contain a higher level of anionic phospholipids than the S90 ingredient [7], such as phosphatidic acid (PA, 1%), phosphatidylinositol (PI, 1%), phosphatidylglycerol (PG, 1%), and acyl-Phosphatidylethanolamine (acyl-PE, 2%). This difference in ingredient composition would therefore account for the fact that S65-coated oil droplets had a higher negative charge [2, 7, 38]. Both the protein-stabilized emulsions maintained a strong negative ζ-potential throughout storage.

The incorporation of sesamol had no obvious effects on the ζ-potentials of all the emulsions, which suggests that it did not alter the electrical characteristics of the droplet surfaces. The droplets in all the emulsions became more negatively charged after 28 days of storage. This suggests that there was some alteration in the interfacial composition during storage, which may have been due to the generation of amphiphilic anionic lipid hydrolysis or oxidation products, such as free fatty acids or organic acids [28].

3.2 Emulsion morphology

Fig. 2 shows changes in the overall appearance and microstructure of emulsions fabricated using different emulsifiers during storage. Initially, all the emulsions had a homogeneous creamy appearance suggesting that the oil droplets were evenly distributed. During storage, however, phase separation occurred in some of the emulsions. A layer of separated oil was visible on top of the S90-emulsions after 7 d storage, which was attributed to rapid droplet coalescence and creaming. The S90-emulsions initially contained relatively large oil droplets (0.35 μm) with relatively weak charges (**Figs. 1A** and **B**) and so they are likely to be particularly prone to gravitational separation and aggregation. The presence of sesamol in the S90-emulsions did not prevent the formation of an oil layer on their surface after 7d storage, suggesting that polyphenol addition did not improve their aggregation stability.

A thin cream layer was observed on top of the S65-emulsions after 14 d storage, but they were much more stable to oiling-off than the S90-emulsions (**Fig. 2A**). According to Stokes law (Equation 3), the creaming rate of the droplets in the S90 and S65 emulsions were 0.53 and 0.41 mm/day respectively, which was consistent with the visual observations of the samples. The initial particle diameters of the S65-emulsions were similar to those of the SS65-emulsions (about 0.31 μm), which suggested that the

addition of sesamol did not impact oil droplet formation for this phospholipid. However, the presence of sesamol within the oil phase retarded droplet creaming in the S65-emulsions, which suggests that it improved their aggregation stability during storage.

Equation (3) was used to calculate the creaming rate of the oil droplets in the emulsions:

$$V_{Stokes} = -\frac{2gr^2(\rho_2 - \rho_1)}{9\eta_1}, \quad (3)$$

Here, g is the gravitational constant, r is the droplet radius, ρ is the density, η is the shear viscosity, and the subscripts 1 and 2 refer to the continuous and dispersed phases, respectively. This equation shows that the creaming rate should increase as the initial size of the oil droplets increases, or if their size increases during storage due to aggregation.

For the protein emulsifiers, a thin oil layer was only observed on top of the emulsions after 28 d storage. The presence of sesamol appeared to improve the physical stability of the WPI- and PP-emulsions, as demonstrated by the fact that they maintained their uniform creamy appearance throughout storage (**Fig. 2A**). These results suggest that sesamol addition may be particularly effective at improving the physical stability of protein-coated oil droplets.

The confocal microscopy analysis of the emulsion microstructures (**Fig. 2B**)

supported the particle size and creaming stability results. Initially, the emulsions produced using the phospholipids contained larger oil droplets than those produced by the proteins. After 28 d storage, there was evidence of some flocculation and coalescence in the emulsions, with the extent of droplet aggregation depending on the type of emulsifier used. The presence of sesamol did not have a significant effect on the measured particle size during storage (Fig. 1A), which suggests that it did not impact droplet aggregation in the protein-based emulsions (Fig.2 B). This effect might be explained by the different sample pretreatments for the two analytical techniques[2]. For the light scattering measurement, the dilution and stirring process might have broken down some weak flocs in the emulsions. In contrast, much less sample preparation is required for confocal microscopy analysis, leading to a more accurate reflection of the actual particle characteristics in the original emulsions [28].

3.3 Laser scanning profiling

The kinetics of emulsion breakdown were quantified by measuring changes in the TSI parameter during storage [39]: a lower TSI value means an emulsion possesses higher stability [40, 41]. This parameter is calculated from changes in the transmission or backscattering of light from an emulsion when a laser is scanned in the vertical direction. The evolution of the TSI parameter during storage for the different emulsions is

compared in **Fig. 3**.

In the absence of sesamol, the emulsion stability results were as follows: WPI>PP>S65>S90. This result suggests that the protein-emulsions were more resistant to changes over time than the phospholipid-ones. With the exception of the S90-emulsions, the addition of sesamol decreased the TSI values of S65 and PP-emulsions, which is indicative of an improvement in emulsion stability. The WPI-emulsions had the lowest TSI values, whereas there was little difference between the WPI and SWPI samples. The good resistance of these systems to separation can be attributed to the small initial droplet size (0.13 to 0.15 μm) of the emulsions, as well as their resistance to aggregation during storage. This result is consistent with the ability of the sesamol to reduce coalescence, creaming, and oiling off reported earlier (**Fig. 2**). The S90-stabilized emulsions exhibited poor physical stability, even in the presence of sesamol, which can be attributed to its relatively large initial droplet size and low droplet charge, as discussed previously.

3.4 Primary and secondary oxidation products analysis

The impact of emulsifier type and sesamol addition on the chemical stability of the flaxseed oil emulsions was then measured. **Fig. 4A** shows changes in lipid oxidation primary products (hydroperoxides) and secondary products (TBARS) during storage at

55℃. The concentration of hydroperoxides increased sharply for the S65- and S90-emulsions after 6 and 2 days of storage, respectively. Conversely, the protein-emulsions maintained a relatively low level of hydroperoxides throughout storage, suggesting that they had better oxidative stability. In the case of TBARS, the lag phase was around 2 days for both phospholipid-emulsions, but around 10 and 16 days for the PP- and WPI-emulsions, respectively (**Fig. 4B**). Again, these results show that the proteins were more effective antioxidants than the phospholipids in these emulsions.

The addition of sesamol improved the oxidative stability of all the emulsions, as demonstrated by its ability to inhibit the formation of hydroperoxides and TBARS. The oxidative stability followed a similar trend in the presence of sesamol as in its absence: sesamol+WPI (SWPI) > sesamol+PP (SPP)> sesamol+S65 (SS65)> sesamol+S90 (SS90). Interestingly, sesamol had a lower ability to inhibit the formation of secondary oxidation products in the SS90-emulsions (at 6d of storage, ~16%) than in the other emulsions. For instance, TBARS formation decreased by about 65% after 8 days storage and by about 85% after 19 days storage for the SS65- and protein-emulsions, respectively. The addition of sesamol improved the oxidative stability of the S90-emulsions, which is probably due to the known antioxidant activity of this polyphenol. The inhibitory efficiency of the sesamol was comparable in the PP- and WPI-emulsions,

which suggests that the plant-based protein may be able to replace the animal-based one in this application.

The inhibitory effect of sesamol on lipid oxidation can be attributed to its chemical reactivity. The benzodioxole group of this polyphenol is known to undergo demethylation with hydroxyl radicals (·OH) to produce another antioxidant molecule, *i.e.,* 1,2-dihydroxy benzene (**Fig. 5A**) [42]. Furthermore, the phenolic group of sesamol can undergo a redox transition, involving a transfer of one electron or hydrogen atom to a free radical, to scavenge radicals and consequently produce less reactive sesamolyl radicals, *i.e.,* $SO^{\cdot}$ **Fig. 5B**) [43]. These radicals can then decay and produce dimers along with other less-reactive products, which slows down the oxidation reaction [44].

The antioxidation ability of sesamol in emulsions was also found to depend on emulsifier type, which may be due to a number of reasons. Firstly, there may be differences in the antioxidant properties of the emulsifiers. It has been reported that phospholipids with lower phosphatidylcholine contents have higher free radical scavenging abilities [2]. Moreover, proteins contain a number of exposed amino acids that have antioxidant properties [4] and so their ability to inhibit oxidation depends on their amino acid sequence and three-dimensional structure. Secondly, the interfacial complexes formed due to the interaction of the sesamol with the emulsifiers may also

have affected the oxidative stability of the emulsions. As mentioned earlier, hydroxyl groups on sesamol molecules can form hydrogen bonds with phospholipid headgroups, thereby bringing the polyphenols close to the oil droplet surfaces where lipid oxidation normally occurs. Previously, it has been reported that sesamol can inhibit lipid peroxidation during the initiation and propagation stages [45]. Sesamol can also form non-covalent complexes with proteins, which can cause the protein molecules to unfold and expose sulfhydryl groups, thereby increasing their antioxidant activity [46].

3.5 Analysis of sesamol concentration in aqueous phase of emulsions

The location of antioxidants in emulsions is known to play a pivotal role in determining their antioxidant activities [28]. For this reason, changes in the concentration of sesamol in the aqueous phase of the emulsions was measured throughout storage (**Fig. 6**). The concentration of the sesamol in the aqueous phase would be expected to depend on a number of factors: chemical degradation during oxidation; movement into the oil phase; and, binding to free or adsorbed emulsifier molecules.

For the S65- and S90-emulsions, the percentage of sesamol in the aqueous phase decreased from around 65% to 45% and from around 56% to 40% during 28 days storage, respectively. Sesamol is predominantly lipophilic but only moderately so (log *P*=1.29), which means that it has some solubility in water (~38.8 mg mL^{-1}) [47].

Considering the relatively small dimensions and low TPSA (≈38.69<<140 Å^2) of this molecule, it is plausible that it can rapidly move from the water phase, through the phospholipid layers, and into the oil droplets [48]. The observed reduction in the sesamol concentration during storage in the phospholipid-emulsions may therefore have been because it was partly consumed during the lipid oxidation reaction and/or it migrated into the oil droplets [49]. The presence of some of the polyphenol molecules within the oil phase or at the droplet surfaces may then have led to antioxidant properties.

The partitioning behavior of the sesamol was quite different for the protein-emulsions. In particular, for the PP-emulsions, the partitioning behavior of sesamol was quite unexpected (**Fig. 6**). Initially, the level of sesamol in the aqueous phase was around 44%, suggesting that it was partitioning fairly evenly between the oil droplets and aqueous phase. During storage, the percentage of sesamol in the aqueous phase increased during the first 15 days, but then decreased during longer storage. This suggests that some of the sesamol may have become bound to non-adsorbed pea proteins during storage and then chemically degraded, but further analysis is required to support this hypothesis.

For the WPI-emulsions, the percentage of sesamol in the aqueous phase progressively decreased from around 100% to 20% during 28 days storage. This

suggests that almost all of the sesamol was initially present in the aqueous phase, but then it either degraded or moved into the oil droplets during storage. It is possible that the sesamol initially remained bound to non-adsorbed proteins in the aqueous phase, which prevented it from moving into the oil phase. During storage, however, some of the sesamol may have preferentially partitioning into the oil phase. Alternatively, some of the sesamol may have chemically degraded. It has been reported that the degradation rate of sesamol in the presence of free radicals is much faster when they are surrounded by water than by oil. For instance, sesamol reacted with •OH and •OOH radicals about 1.6 and 7339 times faster in aqueous solutions than in nonpolar media [50]. Consequently, there may have been a rapid degradation of the sesamol in the WPI-emulsions because it mainly started in the aqueous phase.

3.6 Interfacial tension analysis

The interfacial tension of the flaxseed oil-water interface was measured at pH 7.0 as a function of time (**Fig. 7**). The interfacial tension decreased rapidly during the first 1000 s, and then dropped more slowly at longer times until a relatively constant value was attained. The interfacial adsorption rate decreased in the following order: WPI>PP>S90>S65. Thus, the proteins adsorbed more rapidly to the oil-water interface than the phospholipids. This is probably because the phospholipids self-assemble in

aqueous solutions to form large molecular assemblies (such as liposomes or bilayers) that move relatively slowly [6].

The addition of sesamol reduced the oil-water interfacial tension by an amount that depended on emulsifier type: SS65>SS90>SPP>SWPI. These results suggest that the polyphenol reduced the interfacial tension by adsorbing to the oil-water interface and/or changing the structural organization of the adsorbed emulsifier molecules. It has been reported that there is a relationship between interfacial tension and emulsion formation [51]. A low interfacial tension facilitates the formation of small droplets during homogenization. The fact that smaller oil droplets were produced in the emulsions prepared from the proteins than in those prepared from the phospholipids may therefore have been due to their faster adsorption and lower interfacial tension. On the other hand, a low interfacial tension does not guarantee good emulsion stability, which is demonstrated by the fact that the S90 gave a low interfacial tension but produced emulsions containing relatively large droplets.

3.7 Permeation of free radicals

Lipid oxidation can be promoted by the permeation of free radicals generated in the water or interfacial phases into the oil droplets [35]. For this reason, fluorescence spectroscopy was used to provide some insights into the ability of the interfacial layers

to retard free radical permeation into the oil droplets. In general, the fluorescence intensity of a radical-sensitive non-polar dye (BOPIDY) encapsulated in the oil phase decreased as a function of time after AAPH treatment (**Fig. 8**). This decrease is indicative of the movement of free radicals generated in the water phase into the oil phase where the dye was located. The rate of free radical permeation clearly depended on the nature of the emulsifier system used.

Initially, we consider the behavior of the emulsions in the absence of sesamol. For the S65-emulsions, the fluorescence intensity of the radical-sensitive dye remained close to 95% during the first 400 min but then decreased. Qualitatively, a similar behavior was observed in the WPI- and PP-emulsions but the decrease occurred after about 200 min, which suggested that the free radicals could penetrate into the protein-coated droplets more easily. In contrast, for the S90-emulsions, the relative fluorescence intensity of the radical-sensitive dye decreased immediately, indicating that this type of phospholipid did not provide a good barrier to radical permeation. Interestingly, there was actually a slight increase in the relative fluorescence intensity during the early stages of storage in many of the samples (**Fig. 8**). The origin of this effect is not currently known but it may be due to interactions of the free radicals with the emulsifier molecules in the interfacial layer.

The addition of sesamol increased the time that the fluorescence intensity remained high in all the samples, but especially in the SS65-, SWPI-, and SPP-emulsions, which remained close to 100% for 600 min. In contrast, the relative fluorescence intensity for the SS90-emulsions decreased to approximately 80% after 500 min and then declined rapidly. This result suggests that the interfacial barrier formed by the S90-sesamol complexes was relatively poor, which may account for the poor stability of the SS90-emulsions. Interestingly, the S65-sesamol complexes were more resistant to permeation by the free radicals than the protein-sesamol complexes, even though they had a lower stability to oxidation. The origin of this effect is currently unknown, but it may be because the free radicals generated in the AAPH assay or different from those normally generated during emulsion storage.

Sesamol addition was more effective at inhibiting free radical permeation in the emulsions stabilized by the proteins than in those stabilized by the phospholipids (**Fig. 8**). This may have been because the polyphenols could alter the structural organization of the protein molecules in ways that enhanced their antioxidant activity. Sesamol may have promoted the partial unfolding of the protein molecules, which altered their surface activity, interfacial packing, and interfacial interactions, thereby improving emulsion formation and stability [52]. A tighter and thicker interfacial layer around the

oil droplets would be more effective at inhibiting the permeation of peroxyl radicals. In addition, protein unfolding may have exposed more antioxidative amino acid residues at the protein surfaces, such as tryptophan, histidine, lysine, cysteine, and tyrosine [53], which interacted with the free radicals and prevented them reaching the droplet interior.

4. Conclusion

This study investigated the effects of sesamol addition on the physical and chemical stability of flaxseed oil-in-water emulsions formulated using natural phospholipids or proteins. The physical stability of the emulsions depended strongly on emulsifier type, with the stability decreasing in the order: whey protein > pea protein > sunflower phospholipids. Sesamol addition improved the physical stability of most of the emulsions, as well as increasing their resistance to lipid oxidation, with the degree of the effect depending on emulsifier type. These effects can be attributed to the ability of the antioxidant sesamol molecules to adsorb to the droplet surfaces and interact with the emulsifiers, thereby modulating the interfacial properties of the system. The nature of the interfacial layer was also shown to impact the ability of free radicals generated in the water phase to permeate into the oil droplets. Overall, our results showed that sesamol can be used to improve the physicochemical stability of plant-based food emulsions, which may lead to improvements in product quality and reductions in waste,

thereby improving the healthiness and sustainability of the food system.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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Figure Legends

Fig. 1. Effects of sesamol on mean particle diameters d (3,2) (A) and ζ-potential (B) of phospholipid/protein-based flaxseed oil-in-water emulsions during the storage (0-28 d). Data points represent means ± standard deviations.

Fig. 2. Effects of sesamol on the visual appearance (A) and microstructure (B) of emulsions formed by different emulsifiers during 0-28 d of storage (55℃, in dark). The letters of a, b, c, d, e, f, g, and h represent S65, SS65, S90, SS90, PP, SPP, WPI, and SWPI successively. The scale bar in (B) is 5 μm.

Fig. 3. Variation of TSI value as a function of storage time for phospholipid/protein-based flaxseed oil-in-water emulsions.

Fig. 4. Lipid hydroperoxide (A) and thiobarbituric acid-reactive substances (B) formation in phospholipid/protein-based flaxseed oil-in-water emulsions at 55 ℃ for 28 d. Data points and error bars represent means standard deviations (n = 3).

Fig. 5. (A): The benzodioxole group may undergo demethylation with hydroxyl radical to produce 1,2-dihydroxy benzene (Adapted from Yoshito Kumagai, 1991). (B): The mechanism of sesamol scavenging free radicals (Adapted from Jayaraj, P et, al. 2020).

Fig. 6. The partitioning changes of sesamol into the aqueous phase of flaxseed O/W emulsions at 55 ℃, 4 weeks of storage. Data points and error bars represent means (n=3) standard deviations with two samples were prepared and repeated at least twice.

Fig. 7. Dynamic interfacial tension as a function of time for samples containing sesamol and different types of emulsifiers (S65, S90, PP, WPI) at flaxseed oil/water interface.

Fig. 8. Permeation of peroxyl radicals from aqueous phase to oil phase of emulsions along with result of nonlinear regression. Each data point represents an average of standard deviation in 3 independent measurements.

Figures

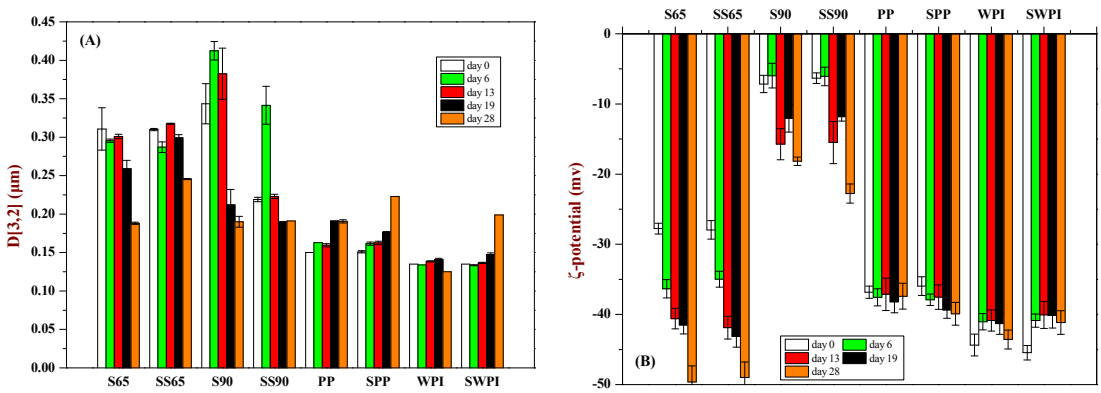
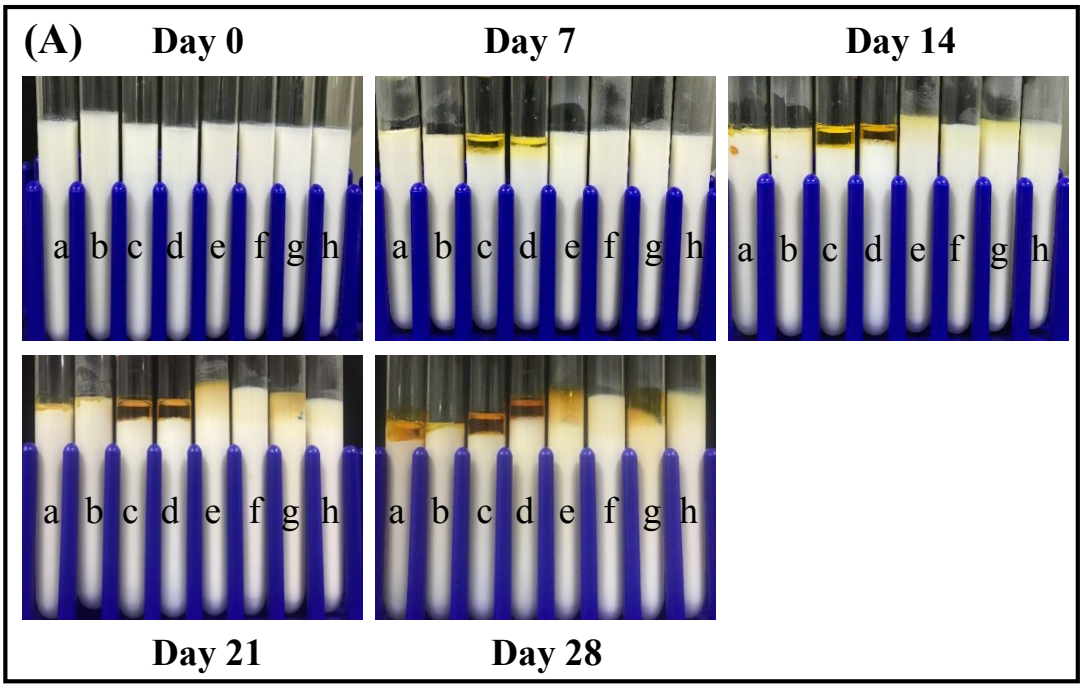


Fig. 1



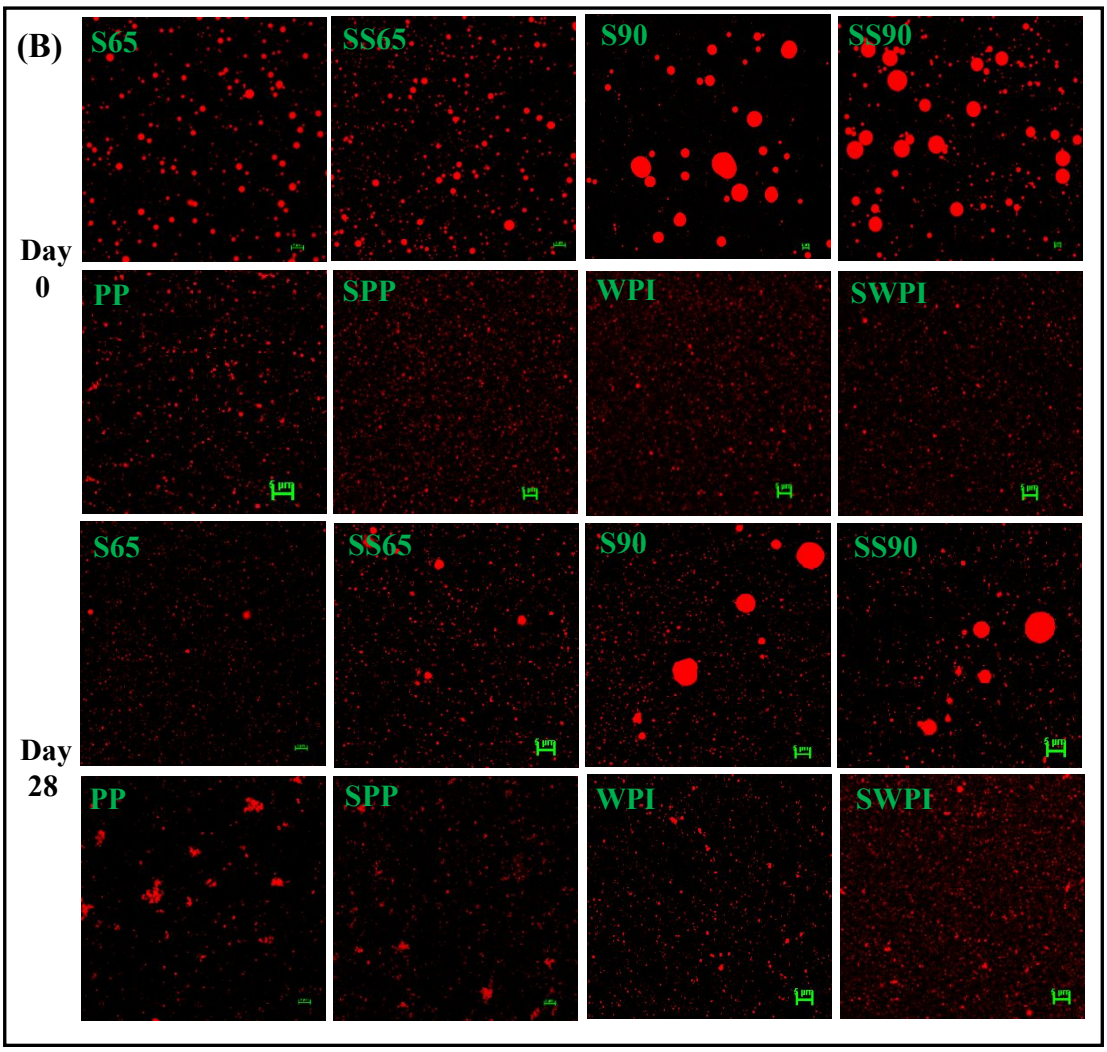

(B)
S65
SS65
S90
SS90
Day 0
PP
SPP
WPI
SWPI
S65
SS65
S90
SS90
Day 28
PP
SPP
WPI
SWPI

Fig. 2

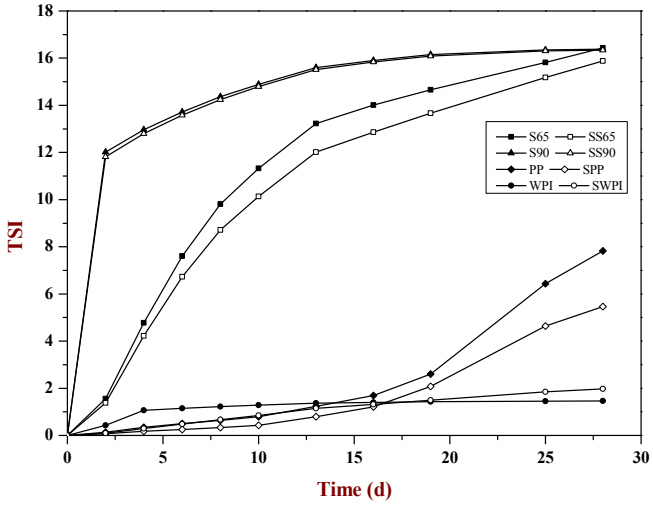

TSI
Time (d)
S65
SS65
S90
SS90
PP
SPP
WPI
SWPI

Fig. 3

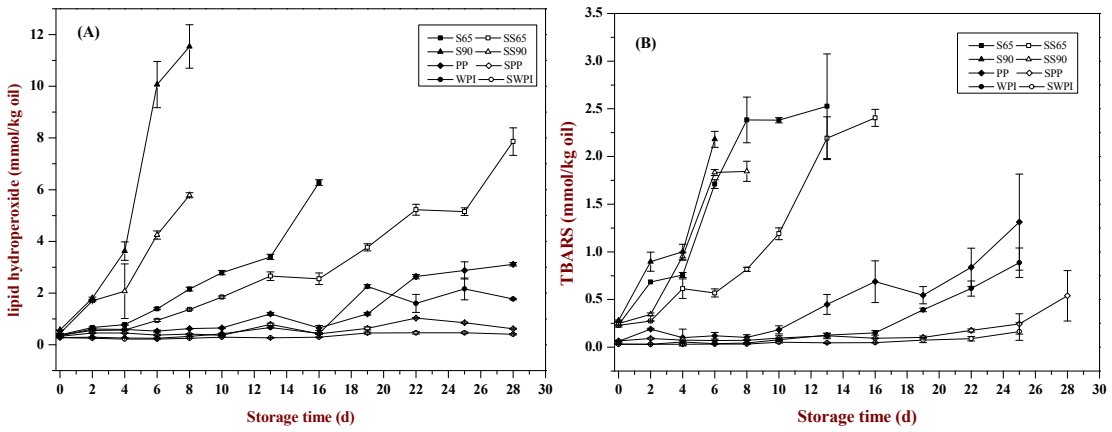


Fig. 4

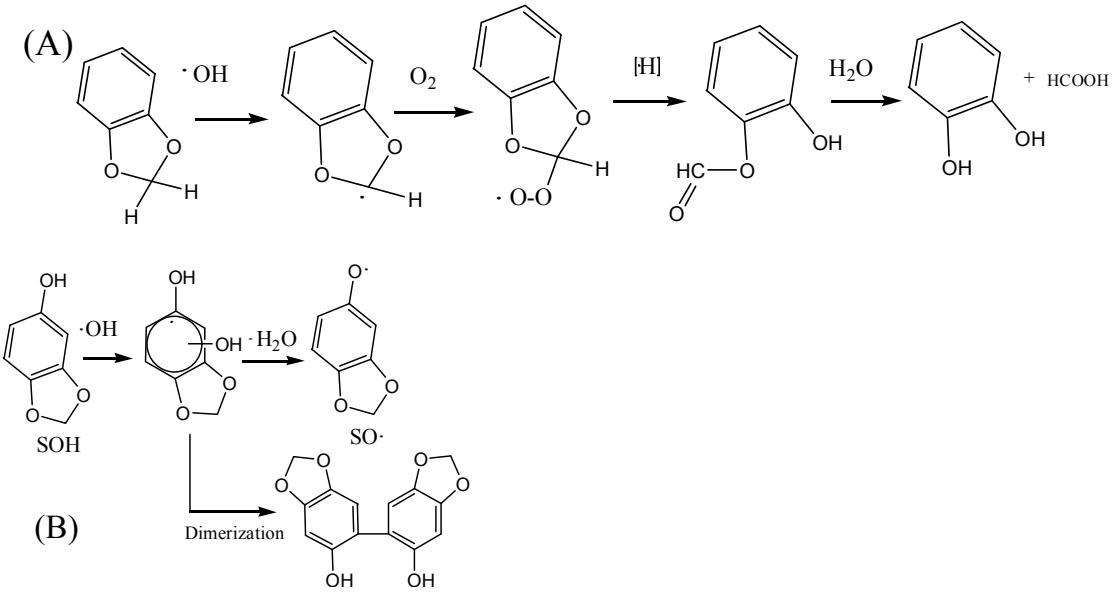


Fig. 5

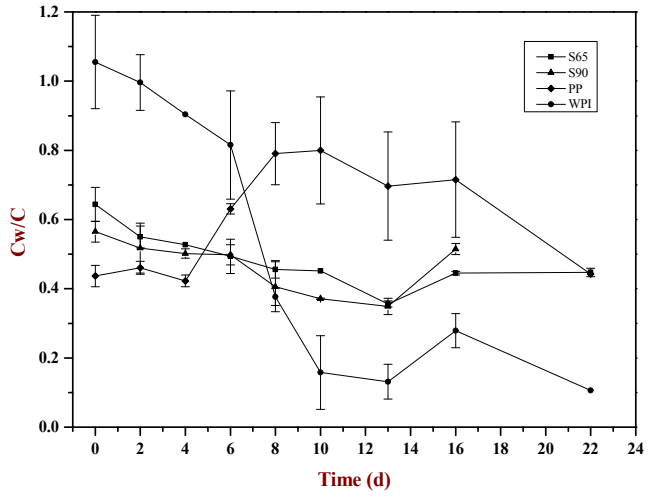


Fig. 6

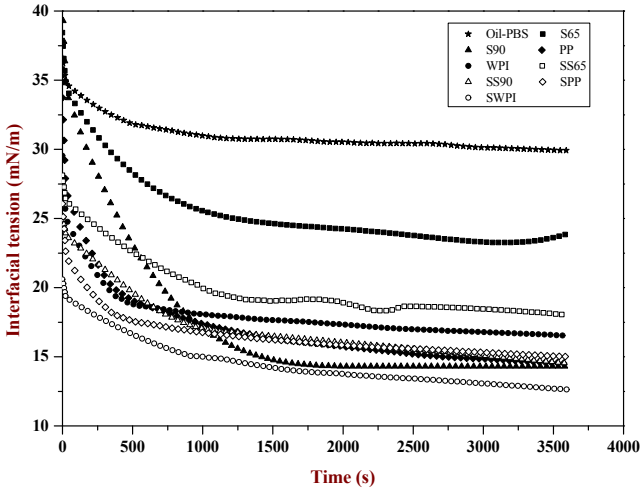

Oil-PBS
S65
S90
PP
WPI
SS65
SS90
SPP
SWPI
Interfacial tension (mN/m)
Time (s)
40
35
30
25
20
15
10
0
500
1000
1500
2000
2500
3000
3500
4000

Fig. 7

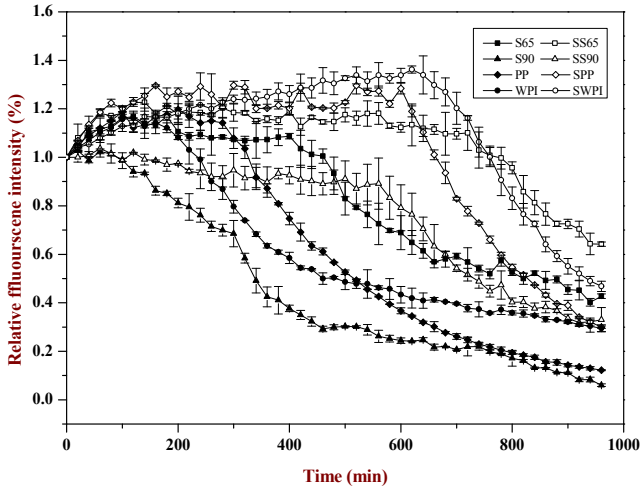

S65
SS65
S90
SS90
PP
SPP
WPI
SWPI
Relative ffluourscene intensity (%)
Time (min)
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
200
400
600
800
1000

Fig. 8

Highlights

- Sesamol had a weak inhibitory effect on lipid oxidation of Sunlipon 90-emulsion.
- Sesamol had comparable inhibition of lipid oxidation efficiency for PP and WPI-emulsions.
- Sesamol could increase the interface barrier performance and reduce the permeation rate of peroxy radicals.
- The hydrogen bonds enabled inhibition of lipid peroxidation during the initiation and propagation stage.

Graphical Abstract