Research Article

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Synthesis and characterization of antioxidant-enriched *Moringa* oil-based edible oleogel

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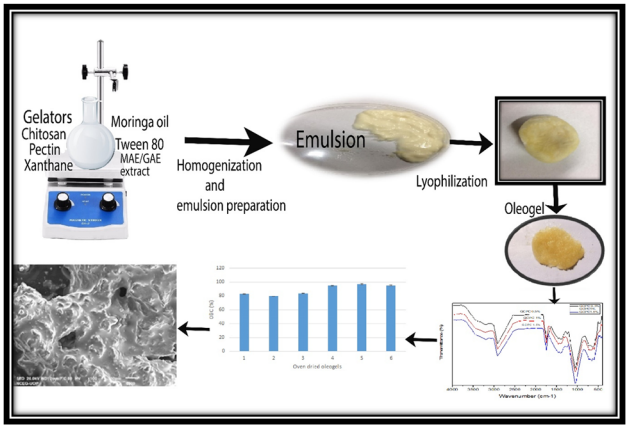
Abstract: This study aims to formulate and optimize *Moringa oleifera* (*Moringa*) oil oleogels using pectin (PC) and chitosan (CS) as gelling agents. These include monogelator oleogels, utilizing either PC or CS as a single gelling agent, and binary gelator oleogels, incorporating a combination of both PC and CS. Among the binary gelator oleogel compositions, the most stable oleogel OPCCS2 was further studied with the addition of antioxidants. The important antioxidant compounds of gallic acid equivalents (GAEs)/*Moringa* antioxidant extracts (MAEs) were quantified by the use of various assays. The oil-binding capacity (OBC) of the most stable oleogel MCPC1.5% was 99.94 ± 0.05. The lower peroxide value of antioxidant-rich oleogels at 1.5% concentration of GAEs (4.34 ± 0.025) and MAEs (4.32 ± 0.03) suggested its richness of phenols to retard the lipid peroxidation of oil. The opaque appearances of the formulations were studied via polarizing light microscopy. The molecular interaction study through FTIR analysis revealed the hydrogen bond interactions between the carboxyl groups of fatty acids and hydroxyl groups of polysaccharide chains. The differential scanning calorimeter analysis further confirmed the presence of strong interactions between polysaccharide chains and the oil phase. These findings indicate that the optimized oleogel formulations have the potential for imminent advances by exhibiting improved texture, biocompatibility, enhanced OBC, and stability.



Graphical abstract

Keywords: antioxidants, emulsion, grapefruit peel, Moringa seed oil, oleogel, solid fats

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Nomenclature

OPC27	*Moringa* oil–pectin oleogel
OCS30	*Moringa* oil–chitosan oleogel
OPCCS2	*Moringa* oil–chitosan–pectin oleogel
MCPC1.5%	*Moringa* oil–chitosan–pectin oleogel with 1.5% *Moringa* antioxidant extract addition
GCPC1.5%	*Moringa* oil–chitosan–pectin oleogel with 1.5% grapefruit antioxidant extract addition
GAE	Grapefruit peel antioxidant extract
MAE	*Moringa* seed antioxidant extract

1 Introduction

Solid fats are indispensable ingredients widely used in bakery products and significantly influence food qualities such as aroma, flavor, texture, and shelf life [1]. However, these solid fats contain saturated and trans fats; their excessive intake has been discouraged due to the development of cardiovascular diseases [2]. The prevention of cardiovascular diseases prostrates in eliminating trans fats from processed food. In this regard, the oleogelation approach presents a promising alternative oil structuring technique to deliver healthy lipid phases in the macroscopic semi-solid network [3].

Oleogelation has been a hot topic in recent research to stabilize healthy solid fats. Conventionally, oleogelation is accomplished by the gelation of liquid oil with different hydrophobic structuring agents such as glycerol, resins, and waxes [1]. It has been established as an efficient physical technique by using food-grade hydrophilic polymers to convert the liquid oil phase into a solid-like gel without influencing the chemical properties of the oil. Polymers such as polysaccharides, cellulose, and its derivatives have been applied to prepare healthy solid fats using healthy and balanced fatty acid profiles of vegetable oils [4,5]. However, there is limited literature on developing emulsion template oleogels using citrus pectin (PC) and chitosan (CS). Based on this, it would be interesting to produce ideal oleogels with CS and PC polymers using emulsion templates. The use of unique antioxidant extracts of grapefruit peels (gallic acid equivalents [GAEs]) and *M. oleifera* (*Moringa* antioxidant extracts [MAEs]) seed residues, which otherwise remain unutilized, in formulating antioxidant-enriched oleogels has not been encountered yet.

CS is a biodegradable, eco-friendly, and amphiphilic polymer that readily dissolves in acidic media due to amino group protonation at the C position [6]. PC is a plant-based heteropolysaccharide, bio-economic hydrocolloid, and a commercial organic surfactant commonly used to make stable jams and jellies without affecting the flavor of the base material. It is present in citrus fruit peels such as lemons and oranges [7]. It possesses an excellent emulsifying and gel-forming capacity and acts as a surface-active agent [8]. Previous research studies [8–10] demonstrated its gelation ability in the water phase and stabilization of the oil–water interface, thus highlighting its potential use in oleogelation.

Stabilizers are components that are added during the formation of oleogel to guarantee homogeneous mixing of the polymer and oil. In this work, xanthan gum (XG) and tween-80 were employed to stabilize the PC and CS emulsions. Coupling the stabilizing agent with amphiphilic polysaccharide is convenient for stabilizing oil in water emulsion to develop oleogel. XG, a heteropolysaccharide, is produced by fermentation microorganisms *Xanthomonas campestris.* It exists in branched form with a cellulose backbone and exhibits a negative charge due to the presence of a carboxylic group in its structure. It can link various positively charged molecules [11].

Moringa seed oil, a healthy edible oil, is a rich source of various antioxidants such as tocopherols, β-sitosterols, catechin, moringine, quercetin, zeatin, and ferulic acid. Its fatty acid profile is identical to olive oil [12]. The healthy monounsaturated fatty acids to saturated fatty acid MUFA/SFA fatty acid profile, along with a lower amount of polyunsaturated fatty acids (PUFA) of this characteristic oil, is associated with lowering the risk of cardiovascular diseases and sterol fractions, and are known to involve in the cholesterol metabolism [13,14]. The oil derived from the seeds of *M. oleifera* can serve as a good liquid phase for emulsion template oleogel formulation. However, the presence of unsaturated fatty acids in large amounts in oils causes rapid oxidation [15].

Synthetic antioxidants have been encouraged to impart long-term stability to foods. Commercially available artificial antioxidants include butylated hydroxyanisole, butylated hydroxytoluene, and tertiary butylhydroquinone [16]. However, health concerns have been raised over time due to the high uptake of synthetic antioxidants [17]. In recent years, interest has been focused on natural antioxidants to provide stability and increase consumer propensity. Dietary polyphenols are natural antioxidants present in plants and protect against damage caused by free radicals [18].

The citrus peels contain larger amounts of flavonoids than those of the other components of the fruit. *Citrus* flavonoids are reported to possess valuable biological activities such as antiviral, anti-inflammatory, and anticancer activities, helping reduce the fragility of blood capillaries [11]. Epidemiological studies prove that the fiber derived from grapefruit peels lowers the risk of gastrointestinal disorders and promotes physiological functions by reducing total serum cholesterol [19]. The phytoconstituents present in seeds of *M. oleifera* exhibit anti-ulcer, anti-inflammatory, and antimicrobial properties [20]. These have good antioxidant properties due to flavonoids like catechin, quercetin, and phenolic acids such as cinnamic and ferulic acids [21]. However, the high amount of active phenolic hydroxyl present in polyphenols of plants with high antioxidant activities can reduce their solubility in oil. For a practical purpose, these polyphenols are employed in biopolymers where these polyphenols interact with polysaccharides to develop colloidal complexes [22]. It contributes a feasible approach to include polyphenolic extracts into emulsion template oleogels.

In this research, the emulsion-based oleogels were prepared and divided into three portions: mono-gelator

oleogels and binary-gelator oleogels without the addition of antioxidants, binary-gelator oleogels containing antioxidant extracts. According to the emulsion template method, *Moringa* oil was chosen as the oil phase, CS and PC as oil structuring agents, and XG and tween-80 as emulsion stabilizers to prepare the edible *Moringa* oil-based oleogels. Then, the antioxidant extracts of grapefruit peels (GAEs) and *M. oleifera* seeds (MAEs) were applied further to increase the stability of oil in the oleogels. The water phase of resultant emulsions was removed through a drying oven and freeze dryer to observe the most effective drying method. The dried products were further sheared to obtain the final *Moringa* oil oleogels. The optimization of oleogels was done at minimal concentrations of gelling agents to increase the oil-binding capacity (OBC) and obtain the desired gel strength. The final oleogels were evaluated by microstructure, OBC, oxidative stability, molecular interactions, and thermal behavior.

2 Materials and methods

2.1 Collection of materials

The *M. oleifera* seeds were purchased from local seed markets of Tehsil Gojra and Faisalabad (Pakistan). The seed's wings and coats were removed from the mature seed pods. Good-quality dried seeds were selected and powdered to uniform particle size using an electric grinder.

The grape fruits were purchased from local fruit shops in District Sheikhupura and Faisalabad. The cleaned peels were then dried in shade for 7 days. Good quality dried peels were selected and powdered to uniform particle size using an electric grinder. *M. oleifera* taxonomic classification is as follows:

Kingdom	Plantae
Division	Magnoliophyta
Class	Magnoliopsida
Order	Capparales
Family	Moringaceae
Genus	*Moringa*
Species	*oleifera*

2.2 Chemicals

Acetic acid (1%), potassium phosphate buffer (0.2 M, pH 6.6), potassium ferricyanide (1%), trichloro acetic acid (10%), ferric chloride ($FeCl_3 \cdot 6H_2O$) (0.1%), Na_2CO_3 (20%), $AlCl_3$ (10%), gallic acid, $NaNO_2$, (5%), 1 M NaOH, Folin–Ciocalteu reagent, 2,2-diphenyl-1-picrylhydrazyl (DPPH) reagent, ABTS (2,2′-azino-di-[3-ethyl benzthiazoline sulfate (6)]) reagent, distilled water, *n*-hexane solvent, acetone, methanol, ethanol, CS powder, PC powder, gum Arabic, sodium sulfate, XG, Tween 80, and *M. oleifera* seed oil.

2.3 Extraction of oil

Dried seeds were powered to uniform particle size. The sample-to-solvent ratio was kept constant for all the methods. About 50 g of the seed powder was pre-weighed for oil extraction that was carried out in Soxhlet apparatus (Model EMEA3) by 250–300 ml *n*-hexane (96% purity) as a solvent at 60°C. The extraction process was repeated for 4–6 runs to obtain a reasonable quantity of oil for 2–6 h. After extraction, the oil was recovered by evaporating the solvent under a vacuum in a rotary evaporator (Model 503) at 65°C [23]. The seed residues obtained after the defatting process were used to extract the antioxidants. The same protocol was repeated for grapefruit peel defatting.

2.4 Extraction of antioxidants

For antioxidant extraction, 30 g of seed residue (in each batch) was soaked in 60 ml of 80% methanol at room temperature for 24 h. After 30 min successful sonication, the samples were allowed to homogenize on an orbital shaker (120 rpm) for 7–8 h. The extracts were separated from the residues through the Whatman No. 1 filter paper. The residues were rinsed twice with fresh solvent. The filtrates were heated in a water bath at 45°C to evaporate the solvent for 2–4 h. The concentrated extracts were allowed to stand in the air to evaporate any residual solvent for a maximum 24 h. The concentrated extracts were weighed to calculate the yield and then at 4°C until further use [24] (Figure 1). The same protocol was followed to obtain the antioxidant extracts of grapefruit peels (Figure 2).

2.5 Dispersion and emulsion preparation

The emulsions were prepared according to the method described in the literature with some modifications [25,26]. The oil in water (O/W) emulsions of *Moringa* seed oil (3 ml) and distilled water (2 ml) were prepared by stirring at room temperature. Aqueous dispersions of PC (0.15 g) and XG (0.1 g) were made in distilled water (Tables 1 and 2). The CS polymer stock solution was prepared by adding the exact amount of CS

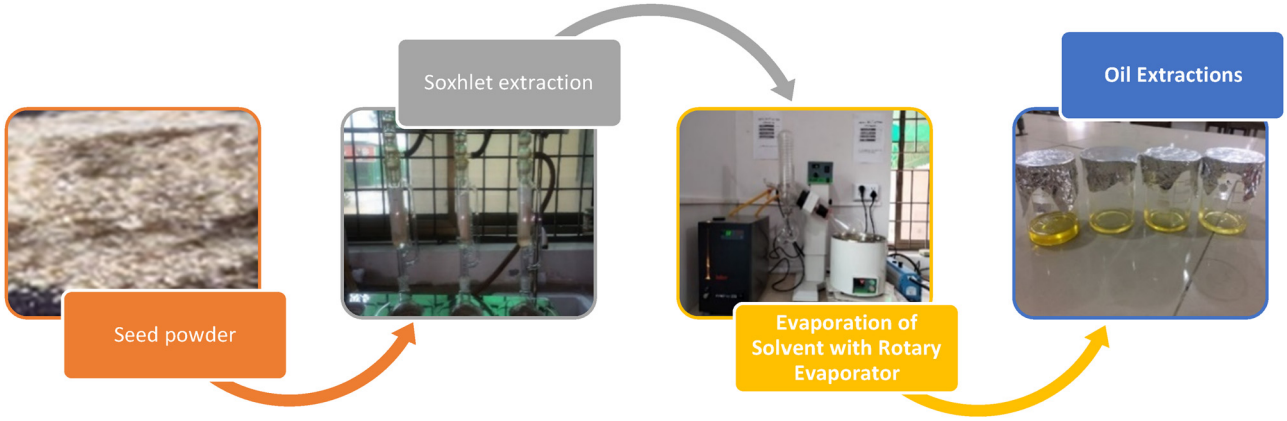

Figure 1: Extraction of oil from *M. oleifera* seeds.

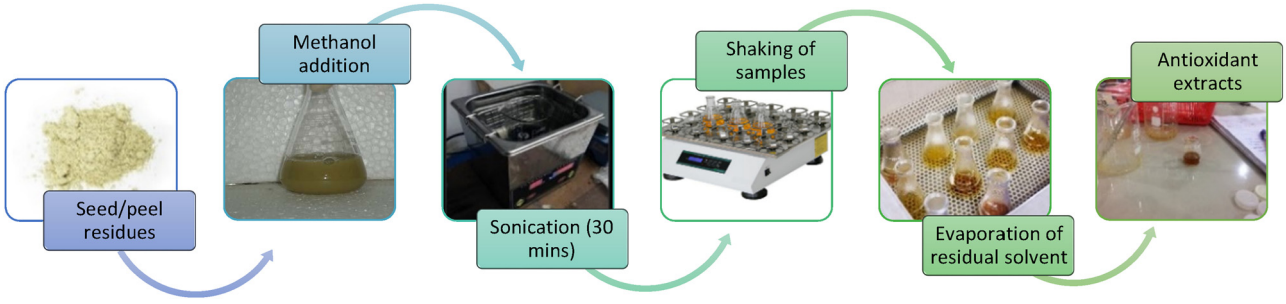

Figure 2: Extraction of antioxidants from grapefruit peel/*Moringza* seed residues.

(0.15 g) powder into 1% acetic acid solution under mixing to disperse the particles uniformly. These dispersions were stirred separately for 1 h at room temperature. These aqueous solutions of PC, CS, and XG were added separately into final O/W emulsions at 800 rpm for 3 h using a mechanical stirrer. For antioxidant oleogels, the extracts of GAEs/MAEs (0.5, 1, and 1.5%) were substantially dispersed in the O/W emulsions (Table 3). The final emulsions were refrigerated at 4°C for 24 h to ensure complete hydration.

2.6 Emulsion-template oleogel preparation

The emulsions were dried using a lyophilizer (freeze drier) at 4°C for 24 h and a hot air oven at 50°C for 24–48 h to remove the water phase. The drying process was monitored until reaching a constant weight. The dried samples were ground with a domestic grinder for 5 s to obtain oleogels [4] (Figure 3).

2.7 Determination of OBC

The OBC of oleogels was measured using the centrifuge method. The weight of an empty (1.5 ml) Eppendorf tube was measured. Approximately 1 g of oleogel sample was refrigerated for 1 h. Then, the weight of the filled Eppendorf tube was measured and was centrifuged at 8,000 rpm for 5 min. The supernatant as released liquid oil was drained

Table 1: Formulation trials screened to find stable oleogel emulsions with PC as the oleogelator

Sample code	MO (ml)	DW (ml)	CS (g)	PC (g)	AG (g)	XG (g)	Tween 80 (µl)	Stability status
$OPCAG_1$	5	3.5	—	0.05	0.05	—	—	Broken
$OPCAG_2$	5	3.5	—	0.05	0.1	—	100	Broken
$OPCAG_3$	5	3.5	—	0.1	0.1	—	100	Broken
$OPCXG_1$	5	3.5	—	0.05	—	0.1	100	Stable (4 days)
$OPCXG_2$	5	3.5	—	0.1	—	0.1	100	Stable (one week)

Table 2: Composition of the synthesized oleogels without the addition of antioxidant

Sample code	MO (ml)	PC (g)	CS (g)	XG (g)	Tween 80 (µl)	DW (ml)
OPC27	3	0.15	—	0.1	100	2
OCS30	3	—	0.45	0.1	100	2
$OPCCS_1$	3	0.23	0.075	0.1	100	2
$OPCCS_2$	3	0.075	0.23	0.1	100	2
$OPCCS_3$	3	0.15	0.15	0.1	100	2

completely by turning over the tubes on the filter paper for 30 min. The weight of the Eppendorf tube after the complete removal of oil was measured. The oil loss values were calculated using equation (1) [27]:

$$\text{OBC}(\%) = 100 - \text{Released Oil}(100\%), \quad \text{Released oil}(\%) = (b - a) - (c - a)/(b - a) \times 100. \quad (1)$$

2.8 Determination of antioxidant activity

The radical scavenging activity of *Moringa* seeds and grapefruit peel methanolic extracts against the DPPH radical was determined according to Yaqoob et al. [28]. About 50 µl of the extract for each six samples of *Moringa* and ten samples of grapefruit was taken separately in 4.5 ml of ethanol. About 0.5 ml of freshly prepared solution of DPPH was added to the reaction mixture, and the absorbance value was measured at 517 nm after 10 and 30 min. Radical scavenging activity was calculated using the following equation:

$$\%\text{inhibition} \times A\text{blank} - A\text{sample}/A\text{blank} \times 100. \quad (2)$$

Table 3: Composition of synthesized oleogels with antioxidant extracts

Sample Code	MO (g)	MAEs (%)	GFAEs (%)	PC (g)	CS (g)	XG (g)	DW (ml)
MCPC 0.5%	3	0.5%	—	0.075	0.23	0.1	2
MCPC 1%	3	1%	—	0.075	0.23	0.1	2
MCPC 1.5%	3	1.5%	—	0.075	0.23	0.1	2
GCPC 0.5%	3	—	0.5%	0.075	0.23	0.1	2
GCPC 1%	3	—	1%	0.075	0.23	0.1	2
GCPC 1.5%	3	—	1.5%	0.075	0.23	0.1	2

Table 4: Antioxidant activities of grapefruit peels (collected from Sheikhupura and Faisalabad) and *M. oleifera* seed (collected from Gojra and Faisalabad) residue extracts (values are mean ± standard deviation, $n = 3$)

Antioxidant extracts	DPPH scavenging activity (%)	FRAP	TFC (mg CE/g-dw)	TPC (mg GAE/g-dw)
GAE_{Skp1}	70.57 ± 1.97^{abc}	1.06 ± 0.03^{c}	1.61 ± 0.03^{i}	14.13 ± 0.02^{i}
GAE_{Skp2}	86.55 ± 1.51^{fg}	1.36 ± 0.02^{f}	1.33 ± 0.04^{ef}	13.96 ± 0.03^{h}
GAE_{Skp3}	88.30 ± 0.61^{g}	1.24 ± 0.02^{def}	1.54 ± 0.01^{hi}	13.90 ± 0.02^{gh}
GAE_{Skp4}	85.43 ± 0.92^{fg}	1.04 ± 0.03^{bc}	1.42 ± 0.03^{fg}	13.79 ± 0.02
GAE_{Skp5}	78.39 ± 1.76^{de}	1.30 ± 0.04^{ef}	1.45 ± 0.02^{gh}	13.73 ± 0.03^{ef}
GAE_{Fsd1}	86.75 ± 1.88^{fg}	0.92 ± 0.03^{ab}	1.48 ± 0.02^{gh}	13.68 ± 0.03^{def}
GAE_{Fsd2}	68.61 ± 2.00^{ab}	1.64 ± 0.02^{gh}	1.41 ± 0.02^{fg}	13.61 ± 0.03^{cde}
GAE_{Fsd3}	82.41 ± 1.45^{efg}	0.89 ± 0.01^{a}	1.43 ± 0.03^{g}	13.55 ± 0.04^{bcd}
GAE_{Fsd4}	82.43 ± 1.45^{efg}	1.02 ± 0.01^{bc}	1.22 ± 0.02^{bcd}	13.48 ± 0.02^{bc}
GAE_{Fsd5}	66.48 ± 2.02^{a}	1.13 ± 0.09^{cd}	1.32 ± 0.01^{e}	13.44 ± 0.03^{b}
MAE_{Goj1}	77.91 ± 1.82^{de}	1.65 ± 0.05^{gh}	1.16 ± 0.05^{bc}	7.74 ± 0.02^{a}
MAE_{Goj2}	74.45 ± 1.06^{bcd}	1.56 ± 0.07^{g}	1.06 ± 0.05^{a}	7.72 ± 0.01^{a}
MAE_{Goj3}	84.89 ± 3.75^{fg}	1.99 ± 0.05^{i}	1.24 ± 0.02^{cde}	7.67 ± 0.01^{a}
MAE_{Fsd4}	75.06 ± 1.70^{cd}	1.21 ± 0.02^{de}	1.14 ± 0.04^{ab}	7.65 ± 0.02^{a}
MAE_{Fsd5}	83.25 ± 2.75^{efg}	1.66 ± 0.05^{gh}	1.30 ± 0.03^{de}	7.64 ± 0.01^{a}
MAE_{Fsd6}	82.16 ± 2.62^{ef}	1.73 ± 0.03^{h}	1.31 ± 0.01^{de}	7.62 ± 0.02^{a}

All experiments were performed in triplicate, and the results are reported as mean ± SD. ANOVA was performed along with Tukey's test to compute the significant difference between different groups. Different lowercase alphabets in a column present significant statistical difference between samples.

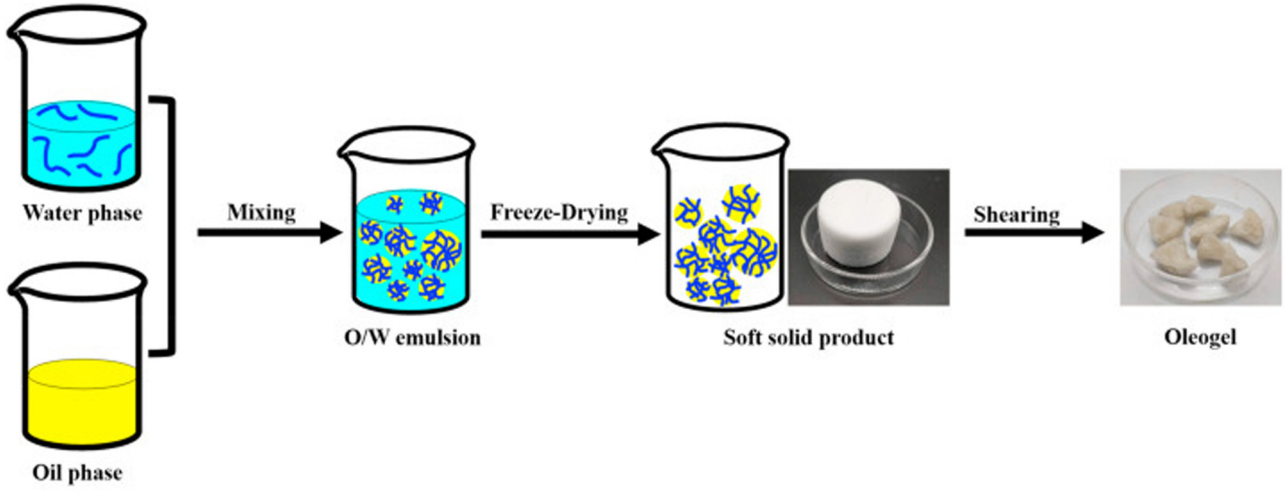

Figure 3: Schematic for the synthesis of emulsion template oleogel.

2.9 Determination of total phenolic content (TPC)

The TPC of grapefruit peel/*Moringa* seed extracts was determined following the procedure of Yaqoob et al. [28]. The TPC of the extracts was determined using the Folin–Ciocalteu reagent. About 50 mg from each *Moringa* and grapefruit extract was taken separately, containing 0.5 ml of freshly prepared diluted Folin–Ciocalteu reagent. The reaction mixture was diluted to 7.5 ml with deionized water. These mixtures were kept at room temperature for 10 min. Then, 1.5 ml of 20% sodium carbonate (w/v) was added to the mixture. Afterward, the mixture was heated at 40°C for 20 min and cooled in the ice bath. The absorbance was measured at 755 nm on a UV spectrophotometer. The amount of total phenolics was calculated by using a calibration curve for gallic acid (R^2 = 0.9988). The results are expressed as gallic acid (mg/10 g) of extract.

2.10 Determination of total flavonoid content (TFC)

TFC was determined following the procedure by Sultana et al. [29]. 1 ml of aqueous containing 0.1 g/ml of the grapefruit peel/*Moringa* seed extract was taken in 5 ml of distilled water, followed by the addition of 0.3 ml of 5% $NaNO_2$. After 5 min, 0.6 ml of 10% $AlCl_3$ was added to the reaction mixture. After another 5 min, 2 ml of 1 M NaOH was added, and the volume was made up with distilled water. The solution was homogenized, and finally, absorbance was measured at 510 nm. TFC was expressed as catechol equivalents per grapefruit peel/*Moringa* seed extract.

2.11 Determination of reducing power

The reducing power of the grapefruit peel/*Moringa* seed extracts was determined by Sultana et al. [29]. About 10 mg of seed extracts was added to 5 ml of sodium phosphate buffer (0.2 M, pH 6.6) and 5 ml of potassium ferricyanide (1%). The mixture was shaken for 20 min at 50°C. Then, 5 ml of 10% trichloroacetic acid was added, and finally, 1.0 ml of ferric chloride (0.1%) was added to the reaction mixture. The absorbance was measured at 700 nm.

2.12 Characterization of oleogels

The microstructure of emulsion template oleogels was visualized with a scanning electron microscope (SEM). Scanning electron microscopy (JSM IT-100 Jeol, Japan) was employed to obtain the micrographs of the samples. The textural properties were studied using a compound light microscope. A small drop of heated sample was placed onto a microscope glass slide and conditioned at 4°C overnight. The samples were analyzed at 25°C using a 20× magnification [30].

2.13 FTIR analysis

The IR spectra of oleogels were obtained using a Spectrum 65 FTIR spectrophotometer equipped with an attenuated total reflection sampling accessory. All the measurements were recorded between the wavelengths of 4,000 and 600 cm^{-1} [27].

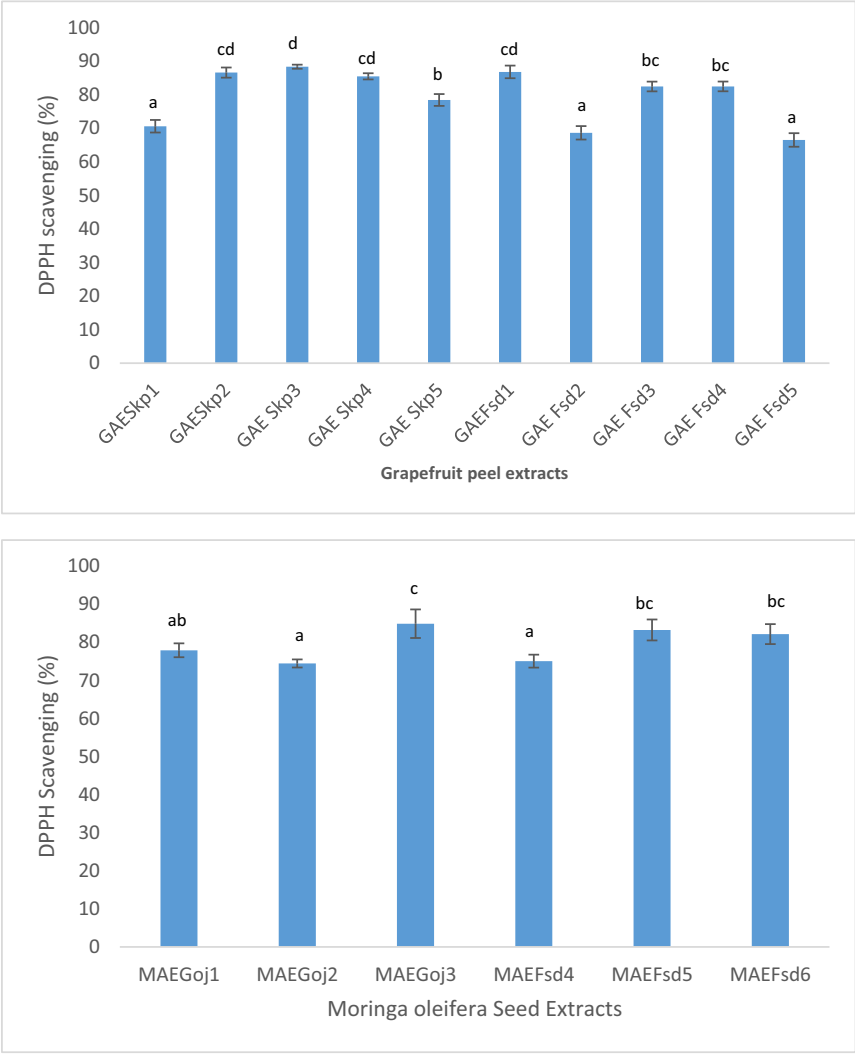

Figure 4: DPPH scavenging activities (%) of methanolic grapefruit peel extracts collected from two different regions and methanolic extracts of *M. oleifera* seed using DPPH.

2.14 Thermal analysis

Thermal measurements of 8–9 mg of the oleogel sample were carried out using a differential scanning calorimeter (DSC; DSC 250, TA Instruments, USA). The oleogel samples in sealed aluminum pans were heated from 30 to 150°C at a heating rate of 5°C/min under N_2 atmosphere.

2.15 Determination of oxidative stability

The primary lipid oxidation of oleogel samples was determined by peroxide value (PV) measurements. Oxidative stability during the storage period of oleogels at 3–4°C was determined following the AOCS method [31]. The PVs (meq O_2 kg^{-1}) of the oleogel samples were measured by the acetic acid–chloroform method and potassium iodide solution.

2.16 Statistical analysis

All measurements were taken in triplicates of the oleogel samples. The results are represented as mean values with standard deviations. The data were analyzed by ANOVA, the significance level (α) was 0.05, and the multiple comparisons of the means were accomplished by Tukey's test using SPSS statistics.

3 Results and discussion

3.1 DPPH antioxidant activity and ferric reducing power (FRAP) assay

The extracts prepared with 80% aqueous methanol were the most effective scavengers of DPPH. The methanolic

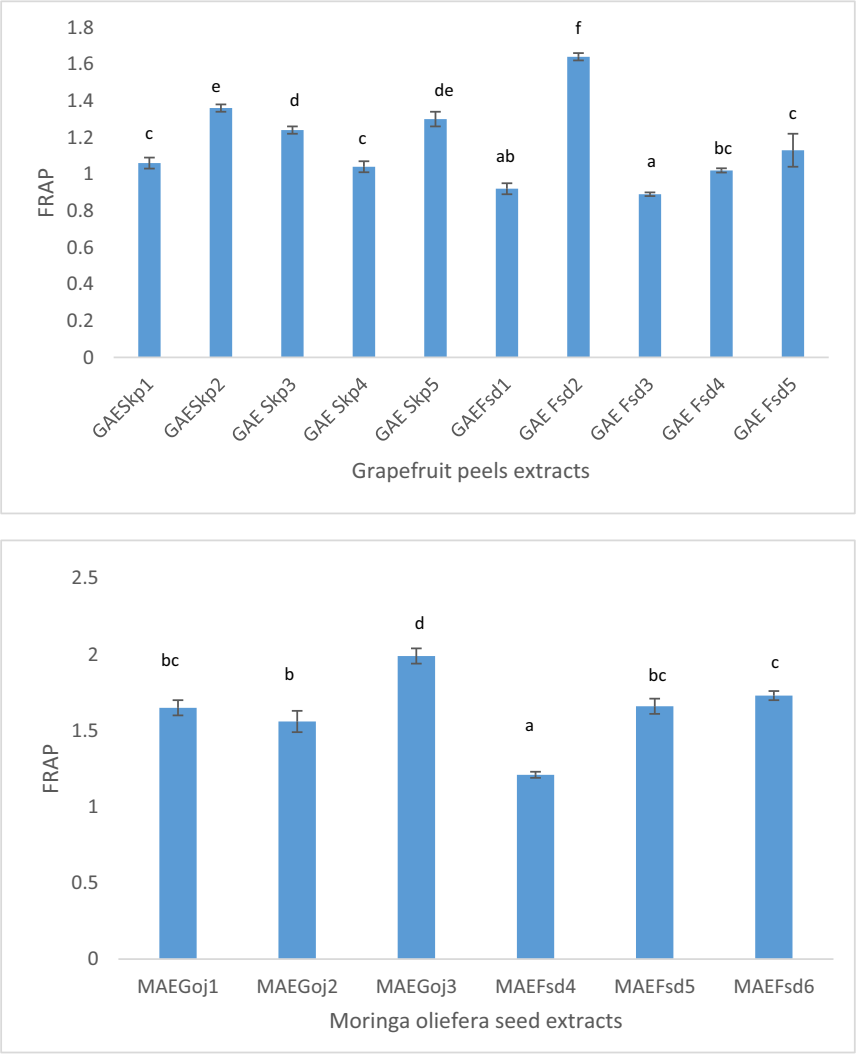

Figure 5: Ferric reducing power of grapefruit peel extracts and of *M. oleifera* seed extracts (data represent the mean ± SD (n = 3).

extracts of grapefruit peels and *M. oleifera* seed extracts were analyzed to determine their radical scavenging power. In the current analysis, ten extracts that were taken from the peel powder of grapefruit showed the highest DPPH radical scavenging activity. The GAE_{skp1}, GAE_{skp2}, GAE_{skp4}, GAE_{skp5}, GAE_{fsd6}, GAE_{fsd7}, and GAE_{fsd8} samples showed maximum antioxidant activity with percentages of 80, 82, 90, 88, 84, 80 and 86%, respectively, as shown in Figure 4a.

Similarly, *M. oleifera* seed extracts were also analyzed for their radical scavenging power. Out of six seed extracts, MAE3, MAE5, and MAE6 exhibited 84.52, 83.25, and 82.16% radical inhibition, respectively. The remaining samples also showed good scavenging activity (Figure 4b). However, the results revealed that all the samples from grapefruit peels and *M. oleifera* seed extracts have high and significant antioxidant capacities. Thus, plant peels and seeds are a good source of antioxidants. In previous studies, it was reported that grapefruit, lemon, and orange peels significantly showed the highest radical scavenging powers of 76.4, 73.2, and 70.5%, respectively (Table 4).

3.2 FRAP

The results of the reducing power assay confirmed that three extracts, GAEskp2, GAEskp4, and GAEfsd7, showed maximum reducing powers of 1.388, 1.337, and 1.677, respectively, as shown in Figure 5(a). GAEskp1, GAEskp3, GAEfsd9, and GAEfsd10 have also shown high reducing powers but less than those of GAEskp2, GAEskp4, and GAEfsd7. These results were found to be in close agreement with those of Babbar et al. [32], in which

Kinnow peel extracts showed a 1.60 value of reducing power.

The FRAP values of MAE3, MAE5, and MAE6 extracts were higher among the total six samples of seed extracts of *M. oleifera*, as shown in Figure 5(b). The extracts with higher reducing power were further chosen to be used in the synthesis of antioxidant oleogels because these extracts have a good ability to prevent the oxidation process.

3.3 Total flavonoid and phenolic content

The TFCs in *Citrus paradisi* peel extract range from 1.32 to 1.61 mg/g. Grapefruit peels taken from the Sheikhupura region have shown the highest TFC values. According to previously reported research, the methanolic extract of *Citrus grandis* exhibited 1.04 mg QE/g TFC [33]. Flavonoids and polyphenols are good hydrogen donors, and their antioxidant ability varies from one compound to another. The rate of flavonoid and phenolic content is affected by numerous factors like the presence of certain chemical groups (organic acids, aromatic amines, ascorbic acid, and sugars) that may also react with the Folin–Ciocalteu reagent. The difference in values of TFC as compared to previously reported values may be described on the basis of the nature of soil and agro-climatic conditions, different chemical compositions, and the efficiency of solvent to extract bioactive compounds. The methanolic extracts of *M. oleifera* seeds were analyzed for TFC determination. Catechol equivalent (mg CAE/g of aqueous extract) was used as a standard to access the flavonoid content (Figure S1). The amount of flavonoid recorded was 1.61 ± 0.01 mg CAE/g of aqueous extract from the total antioxidant content of *Moringa* seeds. The values ranged from 1.24 ± 0.02 to 1.61 ± 0.01 mg CAE/g of aqueous extract, as shown in Table 1. The amount of TFC was found to be lower than the study reported by Mohammed and Manan [34]. As is known, flavonoids are polyphenolic compounds whose structures consist of aromatic rings with multiple hydroxyl groups. They are mostly polar in nature; therefore, they show more solubility in polar solvents like ether. The lower amount of TFC in the case of methanolic extracts of *Moringa* seeds is in close agreement with a previous study [35].

The TPCs in *Citrus paradisi* peel extracts ranged from 13.44 (±0.03) to 14.03 (±0.02) mg/g. The grapefruit peel taken from the Sheikhupura region shows the highest TPC values compared to other extracts. These values indicated that 1 g of the peel extract contains a phenol amount approximately equivalent to 13.44–14.03 mg of pure gallic acid (Figure S1). A previous study reported that Kinnow peel has 17.5 mg GAE/g-dw of phenols [32]. The TPC of the *Moringa* extracts recorded was 7.74 ± 0.02 mg GAE/ml of aqueous extract of the total antioxidant content in *M. oleifera* seeds. The TPC was much higher than TFC, which indicated the richness of phenolic over flavonoids in the methanolic extracts of *M. oleifera* seed [34]. They reported a TPC of 10.179 ± 2.984 mg of GAE/g of dry matter of total antioxidant content of *M. oleifera* seed extract. The concentration of total phenolics can be affected or vary by cultivar, region, storage time, and methods of extraction and drying.

3.4 Determination of OBC

The OBCs of prepared oleogels ranged from 79.36 ± 0.1 to 99.94 ± 0.05. The drying process was observed by recording the fraction weight loss of emulsions periodically until it reached a constant value (Figure S3). The time required for the complete drying of emulsions in a hot air oven at 50°C ranged from 28 to 48 and 24 h at 4°C for lyophilization (Figure S4). The OBC of oleogels was not significantly influenced by the drying methods of emulsions (oven and freeze-drying). However, the freeze-drying (lyophilization) method was preferred over oven drying (Figure S5). The addition of MAEs and GAEs at 0.5, 1, and 1.5% also showed a reduction in oil loss from oleogels. This behavior can be considered due to the formation of stable concentrated emulsions because of the developing thick double layer on the oil droplets and enhancement of continuous phase viscosity in the presence of XG as a stabilizer, resulting in closely packed oleogel network and enhancement in oil entrapment [4]. The increase in the concentrations of MAEs/GAEs above 2% made the hydrophilic groups insufficient to interact with XG to form a stable double-layer interface at O/W emulsion. Therefore, 1.5 wt% was the suitable concentration for MAEs and GAEs to prepare stable oleogel. In fact, it is known that oleogelator compounds play an effective role in oil inhibition and make them stable (Figure 6). Hence, stable oleogel retaining the liquid oil was obtained with MAEs/GAEs, CS, and PC as structuring materials, which met the standard requirements of oleogels.

3.5 Microstructure observation

The microstructure of oleogel samples under a compound light microscope is shown in Figure S6. The polymeric entanglement in the $OPCCS_2$ oleogel had branched chain-

like networks, while in the case of antioxidant (MCPC 1.5%) oleogels, the polymeric chains appeared in bulk; the network became denser, revealing the oil phase as dark sheets. It can relate to the presence of a larger amount of tocopherol and polyphenolic content in MCPC1.5% oleogels. Also, the appearance of compact structures occurs due to the lower extent of coalescence resulting from the immobilization by the polysaccharide chain entanglements.

The morphological observation of binary component oleogels with or without antioxidant extract (MCPC) incorporation was acquired by cryo-SEM. It clearly revealed the compact aggregates, rough surfaces, and irregular morphology of the polymeric network of oleogels (Figure S7). A more inhomogeneous and rougher surface compared to non-emulsified CS and PC was mainly due to the hydrophobic patches of oleic acid. In particular, it was possible to observe a few structural changes due to the collapse of the structure during the dehydration process. Because the drying method prevents the collapsing of the colloidal network during water removal. Furthermore, the interface structured by surface-active and non-surface-active polysaccharides around the oil droplets contributed to preventing the isolation of oil droplets. A large number of oil droplets appear to be dispersed in the continuous phase of polysaccharides through close packing of oil droplets without internal contact.

3.6 FTIR analysis

The information about the chemical bond interactions in the oleogel samples was obtained by FTIR spectroscopy, which covered the wave range of 4,000–500 cm^{-1}. A significant similarity was observed in the IR spectrum of polysaccharides. In view of the molecular nature of various components present in the oleogel samples, the IR spectra of oven-dried and lyophilized oleogels were expected to be almost identical, with peaks at nearly the same wavelengths as indicated in the FTIR spectra. There were some spectral differences between oleogels (without antioxidants) and antioxidants containing oleogels. The oven-dried oleogels of the OPC27 oleogel showed a broad peak in the region 3,283–3,468 $cm^{-1,}$ and the OCS30 oleogel showed peak broadening behavior at around 3,271–3,395 cm^{-1} (Figures 7a and S8). The OPCCS series of oleogel samples also showed broad peaks at around 3,217–3,468 cm^{-1} (Figures 7b and S9).

A very small peak at 3,471 cm^{-1} in the spectra of pure *Moringa* oil was observed (Figure S10).

From the spectra of polysaccharides (Figure S11), broad peaks in the 3,226–3,422 cm^{-1} wave range were also observed. This peak-broadening behavior corresponds to –OH stretching vibrations that are related to intermolecular or intramolecular hydrogen bonding within the chains of polysaccharides. In antioxidant-enriched oleogels, these –OH stretching peaks (Figure 7c and d) ranged from 3,217 to 3,485 cm^{-1}. These peaks suggested the presence of intermolecular hydrogen bonding interactions in the oleogel samples due to the presence of hydroxyl groups of hydrophilic polysaccharide chains, as evident from the deformations in the 3,200–3,400 cm^{-1} range bands. In the formation of polymer oleogels, hydrogen bonding is considered the main interaction force. The crystal network of the ethyl-cellulose (EC) oleogel was also formed due to hydrogen bonding with the polymer chains [36].

The other peaks observed in the spectra of oleogels can also be explained by comparing them with pure *Moringa* oil (Figure S12) and polysaccharides. For instance, the peaks observed at 2,924 and 1,746 cm^{-1} revealed the

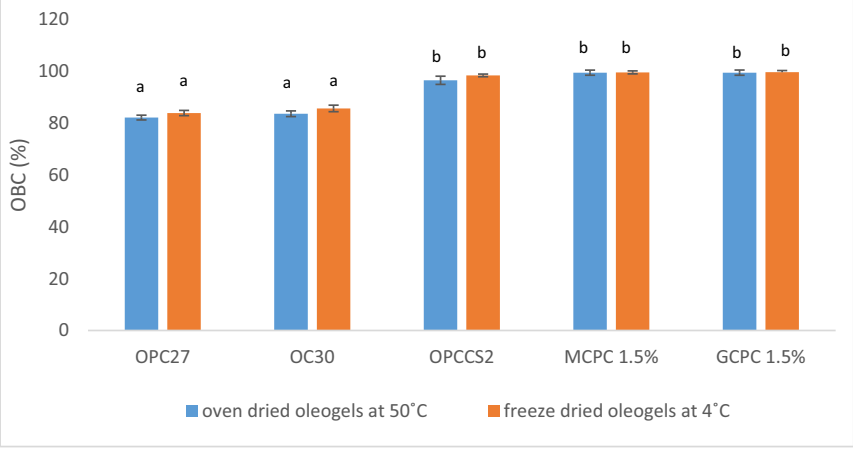

Figure 6: OBC (%) of lyophilized antioxidant oleogels.

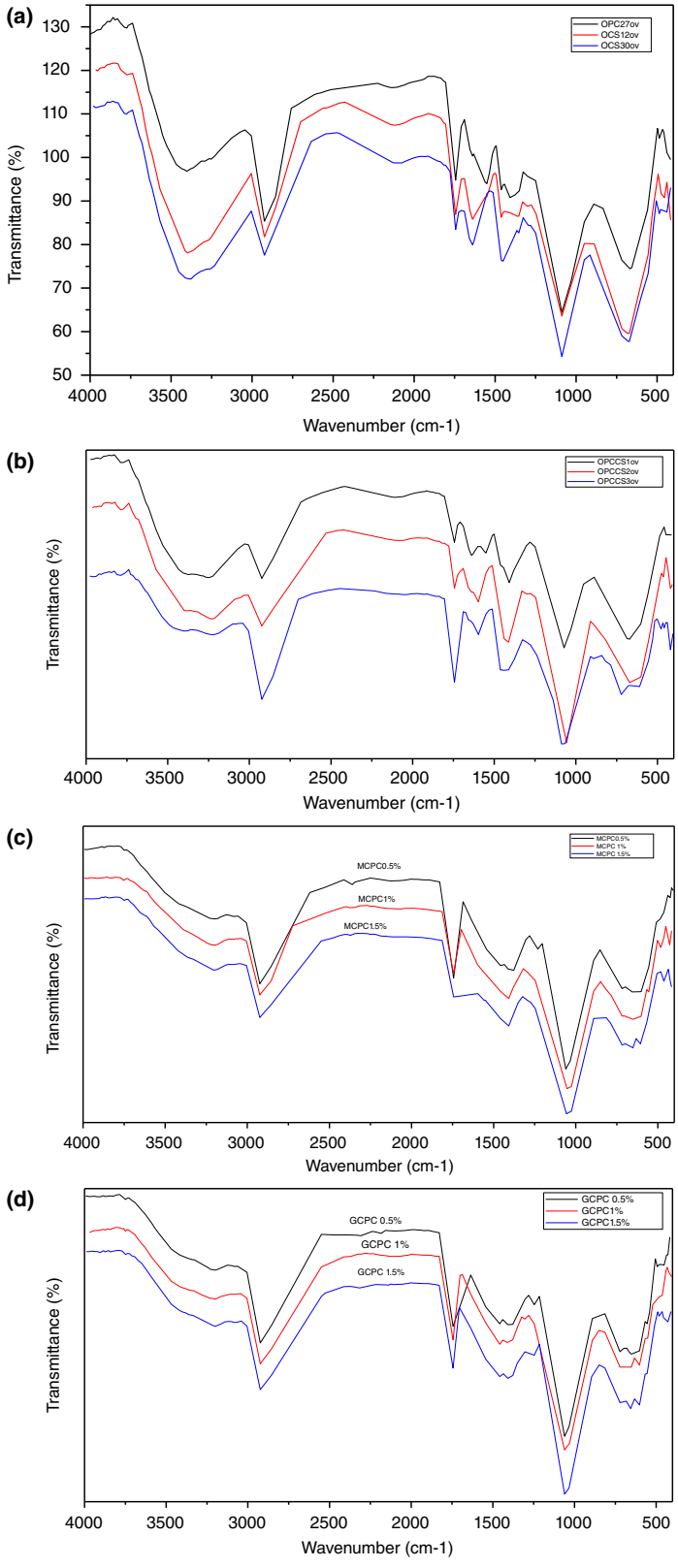

Figure 7: (a) FTIR spectra of oven-dried single oleogelator oleogel OPC27, OCS12, and OCS30. (b) FTIR spectra of oven-dried oleogelator blend oleogels OPCCS1, OPCCS2, and OPCCS3. (c) FTIR spectra of *M. oleifera* seed antioxidant oleogels MCPC0.5%, MCPC 1%, and MCPC 1.5%, MCPC 1%, and MCPC 1.5%. (d) FTIR spectra of grapefruit peel antioxidant oleogels GCPC0.5%, GCPC 1%, and GCPC 1.5%.

abundance of triglycerides in the samples. The peaks observed at 2,924 cm^{-1} from pure oil and polysaccharides powder indicated C–H stretching of $–CH_2$. Similarly, the oven-dried (2,912 cm^{-1}), lyophilized (2,926 cm^{-1}), and antioxidant-enriched lyophilized oleogels also showed C–H stretching vibrations of CH_2 at 2,926 cm^{-1}. The sharp intensity absorption peaks at 1,743 cm^{-1} were observed in *Moringa* oil, polysaccharides, and all oleogel samples. It confirmed the presence of C═O stretching vibrations of esters. The presence of fatty acids was confirmed by peaks at 1,631 cm^{-1} that relate to C═C stretching of disubstituted *cis*-olefins, and the intensity of this peak was used to determine the total unsaturation. The overlapped peaks identified at 1,462 and 1,354 cm^{-1} represented the C–H stretching of CH_3 and CH_2 groups, respectively, from *Moringa* oil and polysaccharides. At lower frequencies, the band near 726 cm^{-1} is assigned to the CH_2 group rocking vibrations and deformation of $–CH_2$ in *cis*-substituted olefins that build up the long-chain mono and PUFA of the *Moringa* seed oil. The appearance of peaks at 1,087 and 671 cm^{-1} corresponds to the C–O stretching of C–O–C and C–X bending of halo compounds.

The results revealed that oleogelation does not affect the chemical properties of the oil, and different molecular arrangements occur due to the self-assemblies of polymeric oleogelators and the oil. The appearance of spectra can be affected by oleogelation techniques and the oil. It can be noticed that there was no variability in the appearance of peaks or shifts for the oleogels in comparison to the polysaccharides and the oil, which indicates the presence of physical interactions and no changes in the chemical structures, are the reasons for oleogel formation.

3.7 DSC analysis

The DSC thermographs of *Moringa*-oil-structured single-component and multicomponent oleogels in the range of 30–100°C are shown in Figure S13. The heat flow (W g^{-1}) versus temperature for the heating cycle was analyzed to determine the melting behavior of different oleogel samples. All oleogel samples exhibited single exothermic peaks. The mono-component oleogels, namely OPC27 and OCS30, had peak melting temperatures at 47 and 53°C, respectively. The overall melting behavior of XG-stabilized PC oleogel was apparently different from CS oleogel, which was probably due to the heterogeneous nature of structuring agents employed, as melting peak behavior depends mainly on their concentration and type rather than the liquid oil phase used. Therefore, the differences in thermal properties may be attributed to the properties of oleogelators. A previous study proved this thermal curve behavior of oleogel samples by using three different vegetable oils. These melting temperatures of oleogels were lower than the pure polymers.

The melting profiles of oleogels prepared with different blends (CS 75: PC 25) displayed a eutectic system. They had DSC melting peaks in between the melting peaks of mono-component oleogels. The combined features of CS and PC are presented. The $OPCCS_2$ oleogel showed a melting exothermic peak at 43°C, whereas the melting behavior of MCPC1.5% was quite interesting. The results showed a decrease in T_{onst} when the antioxidant extract was added to the OPCCS2-based oleogel. It had a significantly lower temperature at 42°C. These lower melting temperatures suggest the strong interactions between the structuring polymers in this proportion; the visible difference is observed in gel networks and the formation of mechanically stable gels.

This eutectic behavior can also positively correlate with the increase in oil retention of the corresponding oleogels. It is, however, interesting to note that the melting temperature of PC oleogel was lower among all the oleogel samples. Thus, the thermal behavior of all oleogel samples was acceptable for developing desirable oleogels.

3.8 Oxidative stability of the oleogel samples

The PV of the samples was found to be in line with the standard value (<10 meq O_2/kg) for fresh vegetable oil. The oxidation rate of *M. oleifera* oil samples was found to increase with an increase in temperature. The PV values of oil samples, named OA, OB, OC, OD, and OE, were recorded as 3.36 ± 0.02, 3.92 ± 0.42, 4.66 ± 0.23, 4.66 ± 0.32, and 2.95 ± 0.18, respectively. It was suggested that the heating process caused the rapid deterioration of fatty acids, as can be observed by comparing the values with reference oil PVs. At maximum heating temperature (80°C) during this study, the value of peroxide started to decline or became constant (Figure 8(a)). This result was closely consistent with the report of Adejuma et al. [37], who also noticed a decrease in the PV of *M. oleifera* oil with the increase in temperature.

For oleogel samples, the oxidation rate was found to be lower than the original oil. All oleogel samples were heated at 80°C, and the changes in their PV were observed each month. As expected, the oleogel samples had effectively inhibited the oxidation of oil. The PV of oven-dried and freeze-dried oleogel samples were calculated separately,

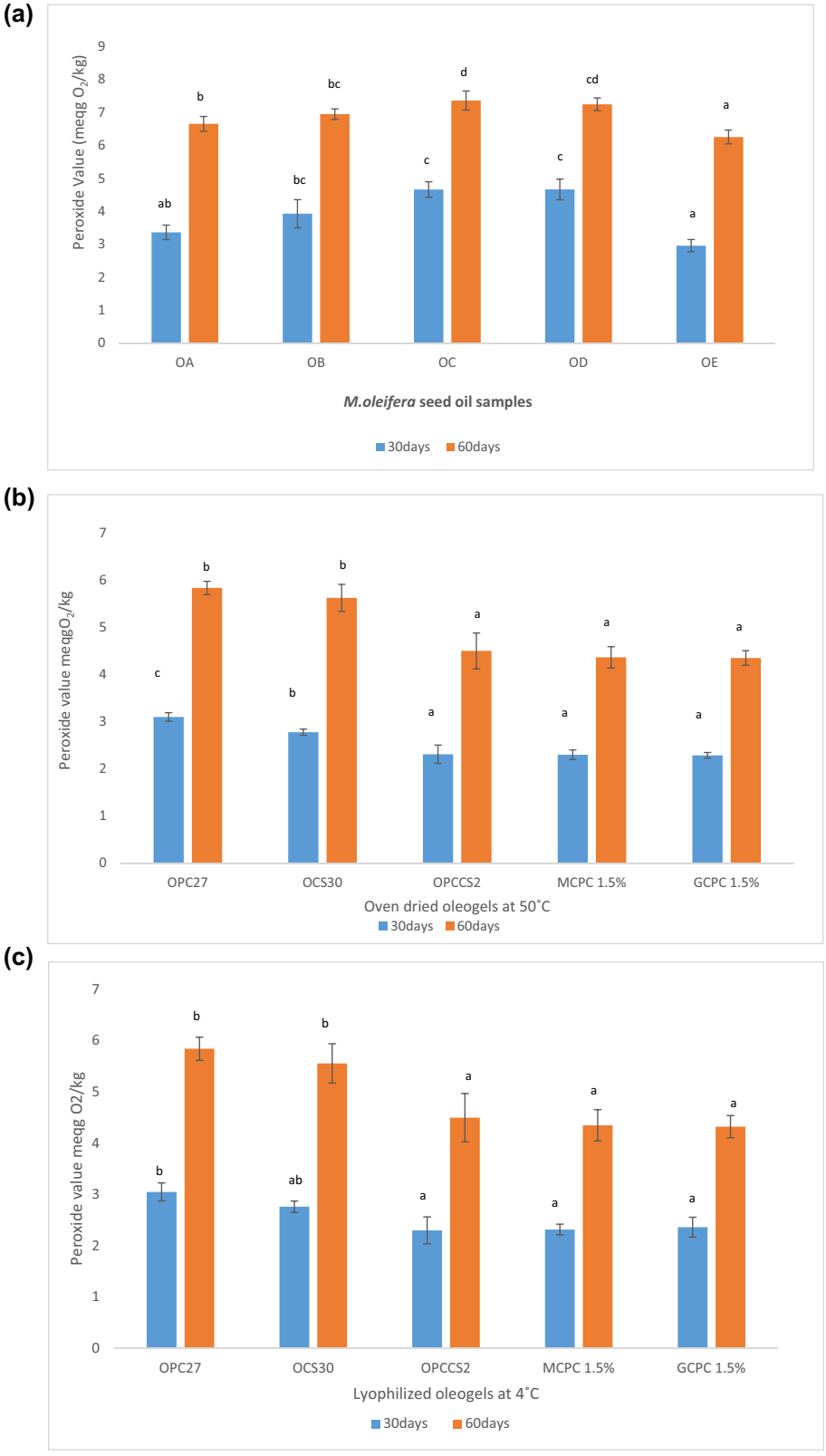

Figure 8: (a) PVs of *M. oleifera* seed oil samples heated at 50, 60, 70, and 80°C; each value is an average of three determinations, mean ± SD. (b) PVs of the oleogel samples; each value is an average of three determinations, mean ± SD. (c) PVs of lyophilized oleogel samples; each value is an average of three determinations, mean ± SD.

and a slight increase in PV of oven-dried samples was noticed. This may be associated with the previous heating process of emulsions during the water phase removal, which clarified that the PVs of oleogels stored at higher temperatures are always higher than those at lower temperatures; this result was consistent with the previous report [27]. The oven-dried OPC27 oleogel had a higher PV of 3.09 ± 0.09 – 5.85 ± 0.06, and the freeze-dried OPC27 oleogel had a PV of 3.04 ± 0.17 – 5.84 ± 0.11 during the 2-month storage, as shown in Figure 11(b) and (c). The oven-dried OCS30 oleogel had a higher PV of 2.71 ± 0.11 – 5.59 ± 0.38, and the freeze-dried OCS30 oleogel had a PV of 2.77 ± 0.06 – 5.62 ± 0.28. The oven-dried OPCCS2 oleogel had a higher PV of 2.28 ± 0.10 to 4.47 ± 0.10, and the freeze-dried OPCCS2 oleogel had a PV of 2.23 ± 0.06 – 4.36 ± 0.22 during 30 and 60 days of storage, respectively.

The addition of antioxidants (GAEs/MAEs) also played an important role in improving the oxidative stability of the oleogel samples. At higher concentrations (1.5%) of GAEs/MAEs, oleogels exhibited lower PV compared to the other oleogel (without extract) samples (Figures S11–S13). It ranged from 2.28 ± 0.10 to 4.35 ± 0.30 and 2.26 ± 0.19 to 4.34 ± 0.22 for GAE/MAE oleogels during 30 and 60 days, respectively.

The oleogelation of *M. oleifera* oil with polysaccharides and MAEs/GAEs caused inhibition of the oil phase due to its immobilization in the gel network. The stability of oleogels against the oxidation process may be attributed to the polysaccharide gel network, which entrapped the liquid oil, thereby retarding the oxidation process. The other reason for lower oxidative damage to oleogels was the addition of antioxidant extract (GAEs/MAEs), which played an essential role in improving the antioxidant activity.

4 Conclusions

These contemporary investigations substantiate the optimal formulation of *Moringa* oil-structured emulsion template oleogels. The biopolymers CS, PC, and XG were used at minimal concentrations to formulate mono-component and multicomponent semi-solid structures binding in the healthy liquid oil. Incorporating natural antioxidants from grapefruit peel and *Moringa* seed residues into these lipid systems presented a possible way to prevent oxidative damage. They exhibited better shelf-life during storage due to the antioxidant contents derived from the plant sources. Both mono- and multicomponent-formulated oleogels showed no noteworthy chemical change as confirmed by FTIR results and DSC results exhibited improved melting points of the multicomponent oleogel. The oleogels have tightly packed structures of oil droplets in the matrix of polysaccharides, adequate to prevent the coalescence of oil during water evaporation. The multicomponent oleogels prepared with varying concentrations of polysaccharides had good gel stability and OBCs. This provides insight into new dimensions of commercial food research owing to the better OBC, stability, and compatibility with the respective polymers. Further research should focus on the addition of other health-beneficial plant antioxidants, new gelling agents, and lipids in the formation of novel oleogels. Future directions include investigations regarding the long-term stability and storage of the edible oleogels within a wide range of conditions such as temperature, light, and change in active functionalities/parameters with time and the effects of these parameters on the quality and integrity of oleogels.

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References

[1] Pehlivanoğlu H, Demirci M, Toker OS, Konar N, Karasu S, Sagdic O. Oleogels, a promising structured oil for decreasing saturated fatty

acid concentrations: Production and food-based applications. Crit Rev Food Sci Nutr. 2018;58(8):1330–41.
[2] Demirkesen I, Mert B. Recent developments of oleogel utilizations in bakery products. Crit Rev Food Sci Nutr. 2020;60(14):2460–79.
[3] Ngan Kee N, Foster E, Marquina C, Tan A, Pang S, O'Brien TJ, et al. Production, health implications and applications of oleogels as fat replacer in food system: A review. J Am Oil Chemists Soc. 2023;100(9):681–97.
[4] Tavernier I, Patel AR, Van der Meeren P, Dewettinck K. Emulsion-templated liquid oil structuring with soy protein and soy protein: κ-carrageenan complexes. Food Hydrocoll. 2017;65:107–20.
[5] Patel AR. Oil structuring: concepts, overview and future perspectives. United Kingdom: Royal Society of Chemistry; 2017.
[6] Farasati Far B, Naimi-Jamal MR, Jahanbakhshi M, Rostamani H, Karimi M, Keihankhadiv S. Synthesis and characterization of chitosan/collagen/polycaprolactone hydrogel films with enhanced biocompatibility and hydrophilicity for artificial tendon applications. Int J Biol Macromol. 2023;253:127448.
[7] Ciriminna R, Fidalgo A, Delisi R, Ilharco LM, Pagliaro M. Pectin production and global market. Agro Food Ind Hi-Tech. 2016;27(5):17–20.
[8] Verkempinck SHE, Kyomugasho C, Salvia-Trujillo L, Denis S, Bourgeois M, Van Loey AM, et al. Emulsion stabilizing properties of citrus pectin and its interactions with conventional emulsifiers in oil-in-water emulsions. Food Hydrocoll. 2018;85:144–57.
[9] Schmidt U, Schütz L, Schuchmann H. Interfacial and emulsifying properties of citrus pectin: Interaction of pH, ionic strength and degree of esterification. Food Hydrocoll. 2017;62:288–98.
[10] Chan SY, Choo WS, Young DJ, Loh XJ. Pectin as a rheology modifier: Origin, structure, commercial production and rheology. Carbohydr Polym. 2017;161:118–39.
[11] Ma L, Chai C, Wu W, Qi P, Liu X, Hao J. Hydrogels as the plant culture substrates: A review. Carbohydr Polym. 2023;305:120544.
[12] Ahmadou F, Bourais I, Aqil Y, Shariati MA, Hlebová M, Martsynkovsky S, et al. Physiochemical characteristics and fatty acids composition of *Moringa* oleifera oil of far north cameroon. J Microbiol Biotechnol food Sci. 2023;13:2.
[13] Schwingshackl L, Hoffmann G. Monounsaturated fatty acids, olive oil and health status: a systematic review and meta-analysis of cohort studies. Lipids Health Dis. 2014;13(1):1–15.
[14] Ras RT, Geleijnse JM, Trautwein EA. LDL-cholesterol-lowering effect of plant sterols and stanols across different dose ranges: a meta-analysis of randomised controlled studies. Br J Nutr. 2014;112(2):214–9.
[15] Patel AR, Rajarethinem PS, Grędowska A, Turhan O, Lesaffer A, De Vos WH, et al. Edible applications of shellac oleogels: spreads, chocolate paste and cakes. Food & Funct. 2014;5(4):645–52.
[16] Mika M, Antończyk A, Wikiera A. Influence of synthetic antioxidants used in food technology on the bioavailability and metabolism of lipids - in vitro studies. Pol J Food Nutr Sci. 2023;73(1):95–107.
[17] Engin AB, Bukan N, Kurukahvecioglu O, Memis L, Engin A. Effect of butylated hydroxytoluene (E321) pretreatment versus l-arginine on liver injury after sub-lethal dose of endotoxin administration. Environ Toxicol Pharmacol. 2011;32(3):457–64.
[18] Borsoi FT, Neri-Numa IA, de Oliveira WQ, de Araújo FF, Pastore GM. Dietary polyphenols and their relationship to the modulation of non-communicable chronic diseases and epigenetic mechanisms: A mini-review. Food Chem: Mol Sci. 2023;6(30):100–55.
[19] Haokip SW, Sheikh KA, Das S, Devi OB, Singh YD, Wangchu L, et al. Unraveling physicochemical profiles and bioactivities of citrus peel essential oils: a comprehensive review. Eur Food Res Technol. 2023;249:2821–34.
[20] Stohs SJ, Hartman MJ. Review of the safety and efficacy of *Moringa* oleifera. Phytother Res. 2015;29(6):796–804.
[21] Singh RG, Negi PS, Radha C. Phenolic composition, antioxidant and antimicrobial activities of free and bound phenolic extracts of *Moringa* oleifera seed flour. J Funct foods. 2013;5(4):1883–91.
[22] Li R, Peng S, Zhang R, Dai T, Fu G, Wan Y, et al. Formation and characterization of oil-in-water emulsions stabilized by polyphenol-polysaccharide complexes: Tannic acid and β-glucan. Food Res Int. 2019;123:266–75.
[23] Özcan MM, Ghafoor K, Al Juhaimi F, Ahmed I, Babiker E. Effect of cold-press and soxhlet extraction on fatty acids, tocopherols and sterol contents of the *Moringa* seed oils. South Afr J Botany. 2019;124:333–7.
[24] Fatima T, Mian AH, Khan Z, Khan AM, Anwar F, Tariq A, et al. Citrus sinensis a potential solution against superbugs. Appl Nanosci. 2020;10(12):5077–83.
[25] Abdolmaleki K, Alizadeh L, Nayebzadeh K, Hosseini SM, Shahin R. Oleogel production based on binary and ternary mixtures of sodium caseinate, xanthan gum, and guar gum: Optimization of hydrocolloids concentration and drying method. J Texture Stud. 2020;51(2):290–9.
[26] Luo S-Z, Hu XF, Jia YJ, Pan LH, Zheng Z, Zhao YY, et al. Camellia oil-based oleogels structuring with tea polyphenol-palmitate particles and citrus pectin by emulsion-templated method: Preparation, characterization and potential application. Food Hydrocoll. 2019;95:76–87.
[27] Yılmaz E, Öğütcü M. Properties and stability of hazelnut oil organogels with beeswax and monoglyceride. J Am Oil Chemist Soc. 2014;91(6):1007–17.
[28] Yaqoob N, Zahira A, Kamal S, Almas M, Rehman S. Development of multifunctional bioactive food packaging based on silver nanoparticles/grape fruit peel extract reinforced PVA composites. Mater Today Commun. 2023;37:107529.
[29] Sultana B, Anwar F, Ashraf M. Effect of extraction solvent/technique on the antioxidant activity of selected medicinal plant extracts. Molecules (Basel, Switz). 2009;14:2167–80.
[30] Bin Sintang MD, Rimaux T, Van de Walle D, Dewettinck K, Patel AR. Oil structuring properties of monoglycerides and phytosterols mixtures. Eur J Lipid Sci Technol. 2017;119(3):1500517.
[31] Millao S, Iturra N, Contardo I, Morales E, Quilaqueo M, Rubilar M. Structuring of oils with high PUFA content: Evaluation of the formulation conditions on the oxidative stability and structural properties of ethylcellulose oleogels. Food Chem. 2023;405:134772.
[32] Babbar N, Oberoi HS, Uppal DS, Patil RT. Total phenolic content and antioxidant capacity of extracts obtained from six important fruit residues. Food Res Int. 2011;44(1):391–6.
[33] Khettal B, Kadri N, Tighilet K, Adjebli A, Dahmoune F, Maiza-Benabdeslam F. Phenolic compounds from Citrus leaves: Antioxidant activity and enzymatic browning inhibition. J Complement Integr Med. 2017;14(1):20160030.
[34] Mohammed S, Manan F. Analysis of total phenolics, tannins and flavonoids from *Moringa* oleifera seed extract. J Chem Pharm Res. 2015;7(1):132–5.
[35] Lalas S, Tsaknis J. Extraction and identification of natural antioxidant from the seeds of the *Moringa* oleifera tree variety of Malawi. J Am Oil Chem Soc. 2002;79(7):677–83.
[36] Laredo T, Barbut S, Marangoni AG. Molecular interactions of polymer oleogelation. Soft Matter. 2011;7(6):2734–43.
[37] Adejumo B, Alakowe A, Obi D. Effect of heat treatment on the characteristics and oil yield of *Moringa* oleifera seeds. Int J Eng Sci. 2013;2(1):232–9.