Red Pepper and Turmeric-Flavored Virgin Olive Oil Oleogels Prepared with Whale Spermaceti Wax

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Abstract: This study aimed to prepare and evaluate ground red pepper and turmeric added virgin olive oil (VOO) oleogels with whale spermaceti wax (WSW) as organogelator. The concentration of WSW was 8 wt%, and each spice was added at 1 overall wt%. Prepared oleogels were analyzed for main physico-chemical, structural, thermal, rheological properties. Further, aromatics volatile compositions, sensory descriptive analysis and consumer tests were completed. Results indicated that the new oleogels were quite spreadable preparates with acceptable quality indices. The oleogels included β type polymorphs, and showed up to 38°C of peak melting temperatures. Rheological measurements proved true gel structure stable within applicable frequencies and above 38°C surrounding temperatures. The oleogels were thermoreversible, and their gel state was recoverable after high shear. Around 25 different aromatic volatile compounds were identified in the two oleogels, most shown to be originating from the VOO, and the spices added. The panel defined and scored the samples with 12 sensory descriptive (hardness, spreadability, liquefaction, sandiness, olive fruit, grassy, waxy, rancid, bitter, hay, cooling and mouth coating) terms. Sensory scores were mostly similar to each other and also within the ranges given in the literature for similar spreadable fat products. Consumer test identified the samples with liked scores (above 4 in 5-max point scale) for appearance, aroma, flavour and overall acceptability. In conclusion, ground spices enriched VOO oleogels with WSW were developed successively to offer consumers spreadable olive oil products to extend consumption patterns with special flavors and health benefits of the spices.

Key words: virgin olive oil, spices, whale spermaceti wax, oleogel, volatile, sensory

1 Introduction

Oleogels have introduced as alternative healthy structured fat sources for various food and non-food applications. Basically, an oleogel is a liquid vegetable oil trapped in a 3-D network created by the organic molecules called organogelators. Organogelators could immobilize the liquid oil through supramolecular assembles like crystal lattices, fibrils, liquid crystals, micelles, bilayers and agglomerates. These networks are usually self-formed via the interactions of sub-units and involve H-bonding, van der Waals attractions and π-π stacking between the phases. By this technology, oleogels with various hardness and rheological properties could be prepared from different liquid vegetable oils. The health advantage of oleogels results from the fact that during oleogelation, no saturation and/or isomerization of the fatty acids of original liquid vegetable oil occur. Further, the minor components of the starting liquid oil do not degrade and contained in the oleogel formed. Consequently, by the aim of food-grade, safe and sensorially acceptable organogelators, oleogels could be prepared with desired properties and utilized in food and non-food products.

Among others, waxes have been extensively used as food-grade organogelators. Especially sunflower wax, rice bran wax, carnauba wax, candelilla wax, berry wax, beeswax, shellac wax, and others have been investigated. Waxes pose advantages like being effective at low concentrations, creating durable and stable oleogels, yielding sensorially acceptable products and feasibility for cost and availability. Consequently, new plant and animal wax sources have been researched for oleogelation purposes.

Whale spermaceti wax (WSW) was investigated for oleogel formation in our previous study. Results indicated that WSW was very successful in gel formation, and its oleogel resembled beeswax oleogel in most properties. Consequently, WSW deserves more research in this area. The WSW used in this study was a pharmaceutical-grade product, and there was no food-grade product over the
available markets. Food-grade ingredients must not contain any poisonous or deleterious substances above any regulated threshold. Pharmaceutical-grade holds the same requirements in addition to other requirements for medical supplements and drugs\(^8\). Hence, pharmaceutical-grade WSW could be used as a food ingredient as long as its sensory properties were suitable for food products. The wax used was very pure, fine, and with no unpleasant taste and odor. Hence, we thought that it would be used in oleogel preparation. Consequently, WSW producers may extend their product portfolio to include food-grade waxes. Unfortunately, literature lacks about detailed chemical compositions of the WSW. It was stated that the WSW is extracted from the head sonar organ of the sperm whale (Physeter macrocephalus), and contains around 65-95% fatty esters, 5-30% triglycerides, 1-5% free alcohols, 0-3% acids. The wax ester fraction mostly includes C10:0, C12:0, C14:0, C16:0 and C18:1 fatty acids. Its melting range is given as 42-50°C, and it is used in cosmetics, pharmacy and candle manufacturing\(^9\).

Virgin olive oil (VOO) is a very healthy oil extensively consumed in the Mediterranean region and worldwide. It is common practice to add some ground spices (garlic, onion, thyme, pepper, oregano, rosemary, etc.) into olive oil and consume these spice-enriched oils in breakfast as dip, and as a sauce for other dishes\(^10\). There were some studies in literature about the preparation of spreadable olive oil oleogels with various plant waxes\(^11\). In this study, ground red pepper (paprika) and turmeric-enriched VOO were prepared as oleogels for the first time. Also, WSW was used as the organogelator to prepare and evaluate these new spice containing VOO oleogels. The aim was to prepare and characterize these new kinds of oleogels. In addition, volatile aromatics compositions of the prepared oleogels and the sensory descriptive analyses and consumer tests of the prepared oleogels, were provided. Since there was only one study about the oleogel of WSW\(^12\), and fairly limited studies with volatile aromatics composition and sensory analyses of the VOO oleogels, this study would greatly add up to the oleogel literature. Further, it may aim WSW producers to develop food-grade WSW for oleogel and other possible food applications.

2 Materials and Methods

2.1 Materials

The virgin olive oil (VOO) used in this study was purchased from a local factory (Çanakkale, Turkey) in October 2020 processing season as fresh produce. First-grade ground red pepper (paprika) and turmeric were bought from local stores. Pharmaceutical-grade whale spermaceti wax (WSW, Blanc de Baleine) was purchased from Doğan İlaç Hammaddeler Co. (İstanbul, Turkey). The ingredients used to prepare the oleogels in this study could be viewed in Fig. 1. All standards, chemicals and solvents used were of analytical grade and purchased from Sigma Chem. Co. (St. Louis, MO, USA) and Merck (Darmstadt, Germany).

2.2 Preparation of the oleogels

The concentration of WSW as an organogelator was decided after some pre-experiments. In our previous study, WSW was added at 10 wt% to prepare the oleogel\(^13\). Since that oleogel was observed as harder than commercial spreadable margarines, in this study, we prepared oleogels with 3, 5, and 8 wt% WSW, and then compared their sensory hardness and spreadability with commercial spreadable margarines, as pre-experiments. Then, 8 wt% organogelator addition level was selected to prepare the oleogel samples. To prepare 500 g oleogel, 460 g virgin olive oil (VOO) weighed and put into a glass beaker. Then, 40 g whale spermaceti wax (WSW) was weighed and put into the oil. The mixture was heated in a water bath at 80°C for around half an hour to melt and mix the wax fully. Finally, 5 g of spice (ground red pepper or turmeric) was put into the melted mixture and homogenized at 3000 rpm for 3 min to disperse the spice. Finally, the mixture left at ambient temperature (20±5°C) overnight for full gelation. The next day, the prepared oleogels were covered with aluminium foil, and placed into the refrigerator until analyses. The prepared oleogels could be observed in Fig. 1.

2.3 Physico-chemical analyses of the oleogels

The gelation time (GT) is required for a melted oleogel to solidify at a given temperature. Each oleogel sample was weighed (1 g) into capped tubes (16×100 mm) and melted in a water bath at 80°C for 30 min before taking to ambient temperature (20±5°C). The chronometer was started after taking the tubes to the ambient environment, and the time elapsed until the oleogels solidified were recorded as the GT. The occurrence of solidification (gelation) was decided by tilting tubes 180° and observing no flow\(^11\).

The oil binding capacity (OBC) was measured by putting 1 mL of melted oleogel into tared Eppendorf tubes, and then letting them gel overnight, and finally centrifugation (Sigma 2-16K, Osterode, Germany) at room temperature for 15 min (10,000 rpm), before drainage of the released liquid oil on the paper cloth. The tubes were weighed again, and the OBC was calculated gravimetrically\(^11\).

The solid fat index (SFI) of the oleogel samples was measured with a Minispec Bruker NMR Analyzer mq20 (Bruker Optics, Inc.) following ISO method\(^14\). The instrument’s calibration was completed with standards including 0, 31, and 73.5% solid fat. The melted oleogels were transferred into the NMR tubes (3.5 mL), and conditioned in a water bath at 0°C, and then in another water bath at 20°C for 1 h. Finally, the tubes were placed in the instrument, and SFI% was recorded.

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Spice-flavored Oleogels

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The instrumental color of the oleogel samples were measured with a Minolta CR-400 (Konica Minolta Sensing, Osaka, Japan) colourimeter according to CIE standards. The values L, a* and b* were recorded on multiple points of replicate samples.

The free fatty acidity (FFA) and peroxide values (PV) of the oleogels were determined following the methods of Ca 5a-40 and Cd 8-53, respectively. FFA was calculated over oleic acid as the major fatty acid in VOO, and PV was given as milliequivalent oxygen per kg sample.

2.4 X-ray diffraction patterns of the oleogels

PAAnalytical Empyrean model (The Netherlands) X-ray diffractometer and Cj 2-95 method were used to assess the polymorphic forms of the oleogel samples. The oleogel samples were kept at ambient temperature overnight and loaded at ambient temperature to the instrument’s sample holder with a spatula. A Cu source X-ray tube (λ = 1.54056 Å, 40 kV and 40 mA) was produced angular scans (2θ) from 2.0° to 50° by 2°/min scan rate to test the samples. The X’Pert HighScore Plus software (Malvern Panalytical Ltd., Royston, UK) of the instrument was used for data analysis.

2.5 Thermal analyses of the oleogels

The thermal properties of the oleogels were analyzed with a Perkin-Elmer 4000 Series Differential Scanning Calorimeter (DSC) (Groningen, The Netherlands). The instrument was previously calibrated with Indium and Zinc. Around 10 mg of each sample was placed into the aluminium pans and sealed. The temperature program was heating samples from 20°C to 100°C at 10°C/min rate; cooling samples from 100°C to −30°C at 10°C/min rate and keeping 3 min at that temperature for full crystallization, and finally heating samples again to 100°C at 10°C/min heating rate. This type of thermal cycling analysis is preferred to determine both crystallization and melting temperatures and enthalpies simultaneously. The Pyris 1 Manager Software of the instrument was used for the calculations.

2.6 Rheological analyses of the oleogels

Rheological analyses were completed with a DHR 2 rheometer (TA Instruments, USA) with a Peltier system (± 0.1°C) under the lower plate donated with cross-hatched parallel plate geometry (ϕ = 40 mm, gap 0.9 ± 0.1 mm). Excess quantities of each sample were scooped and deposited at the centre of the lower plate. After mounting, the sample was let to rest for three min to get its original consistency. Then, the excess sample was trimmed, and the final gap set before analysis. According to selected measurement parameters from the instrument software, the rheometer set itself automatically before starting the experiment. All rheological parameters were measured at 10°C to get a good comparison since all samples were fairly well solid at that temperature. The linear viscoelastic region (LVR) was determined with an amplitude sweep test (0.01-100% strain and 1 Hz frequency) at first for the validity of all further analyses. The LVR is defined as the region where a plateau for the storage (G’), and loss (G’’) moduli were observed.

The LVR region determined strain values and frequencies from 0.1 to 100 Hz were used for the frequency sweep tests at 10°C, and the G’ and G’” values of the samples were assessed. The test was performed using the same geometry, frequency, and sweep range of the LVR determination.

Fig. 1 The ingredients (virgin olive oil, whale spermaceti wax, red pepper, and turmeric), and the prepared oleogels (WWO-RP: whale spermaceti wax oleogel with red pepper, WWO-TM: whale spermaceti wax oleogel with turmeric).
recorded. To evaluate the structural recovery ability of the oleogels under specific stresses applied, a time sweep test was accomplished. Three gradient regions for strains were selected and applied to the samples at 10°C with 1 Hz frequency. The first region was applied for 180 s at each LVR region strains to simulate the resting condition. In the second region, strains well above each LVR region strains were applied for 180 s to simulate the structural breakdown conditions. Finally, strains much lower than those of the LVR strains were applied for 900 s to simulate the structural recovery region. This test yielded graphics about the structural recovery abilities of the samples exposed to stress and released from the stress.

To observe the effect of temperature on oleogel structure, a temperature ramp test was also applied. The test was done by heating the samples from 0°C to 80°C by 1°C/min heating rate at 1 Hz frequency within the LVR strains.

2.7 Volatile aromatics compound analysis of the oleogels

The volatile aromatics compounds present in the oleogel samples were analyzed following the modified technique of Yilmaz. Around 2.0 g of oleogel sample was weighed into vial (15 mL, Clear PTFE/silicone septa cap, Supelco) and closed with a silicone septum. The vial was placed into a water bath set at 60°C and kept for 15 min without fiber, and 30 min with fused silica SPME fiber CAR/PDMS (75 mm Fused Silica, Supelco Ltd., Bellefonte, PA, USA) inserted. The fiber was maintained in the head-space to adsorb volatiles present in the sample. The fiber was then inserted into the injection port of a Shimadzu GC-2010 Plus gas chromatograph for 5 min at 250°C for desorption of the volatile compounds. GC-MS analyses were performed with an MS-QP2010 plus mass spectrometer (Shimadzu Corporation, Kyoto, Japan). Helium was the carrier gas at a flow rate of 1.61 mL/min. An Rx-5Sil MS capillary column (30 m - 0.25 mm x 0.25 mm; catalog no: Restek 13623, Restek, Bellefonte, PA, USA) was used for compound separation. The injection port and the detector were held at 250°C. The column was held at 40°C for 2 min and then increased to 250°C at a heating rate of 4°C/min, and then kept for 5 min at that temperature. The temperatures of the ion source and the transfer line were 200 and 250°C, respectively. Electron impact mass spectra were recorded at ionization energy of 70 eV. GC-MS analyses were performed in scan mode in the 40-300 amu mass range. Volatile compounds were tentatively identified by Wiley, Nist, Tutor, and FFNSC mass spectra libraries. Retention times and % area values of the determined volatiles were provided.

2.8 Descriptive sensory analysis of the oleogels

Quantitative Descriptive Analysis (QDA) test was applied to describe sensory attributes of the oleogel samples. There were 7 female and 4 male panellists aged between 20 and 50 incorporating voluntarily to the evaluations. A consent form was signed indicating that the samples were safe, and provided to the panellists. The panel leader trained panelists at least 8 hours to select, define, and standardize the sensory terms used. The panel determined 14 descriptive sensory terms, and their definitions and references were given in Table 1. A 10 cm line-scale anchored between 0 at the left end for minimum intensity and 10 at the right end for maximum intensity was used to quantify the sensory attributes. In each session, the two samples coded with 3-digit numbers were served to the panel. Duplicate samples were served in different sessions randomly. All tests were carried out at room temperature under day-

| Table 1 | The panel defined sensory descriptive terms, their definitions and references used. |
|---------|----------------------------------------------------------------------------------|
| Definition | References |
| Hardness | Force required to push a knife into the sample | Min: Yoghurt, Max: Tallow |
| Spreadability | The level of deploying sample over a bread loaf | Min: Chewing gum, Max: Cream cheese |
| Liquefaction | Amount of liquid oil released after the sample was spread on bread surface | Min: Tallow, Max: Olive oil |
| Sandiness | The perceived gritty texture on tongue and palate | Min: Absent, Max: Semolina |
| Olive fruit | The flavor and aroma of fresh green olives | Min: Absent, Max: Green olive |
| Grassy | The smell of fresh cut grasses | Min: Absent, Max: Cut grass |
| Waxy | Aromas associated with waxes | Min: Absent, Max: Paraffin wax |
| Rancid | Aromas associated with oxidized oil | Min: Absent, Max: Used frying oil |
| Bitter | Main taste perceived on tongue from caffeine and pepper | Min: Absent, Max: Red pepper |
| Hay | The aromatics associated with sweet, dry grasses | Min: Absent, Max: Dry hay |
| Cooling | Cold feeling inside mouth | Min: Absent, Max: Menthol candy |
| Mouth coating | The perceived fatty coating inside mouth space | Min: Liquid oil, Max: Butter |
light, and the panellists were provided with a plastic knife, water, bread slices, apple slice and an expectoration cup.

2.9 Consumer tests of the oleogels

With 50 volunteer consumers, the appearance, spreadability, aroma, flavor, and acceptability of the oleogel samples were assessed. Oleogel samples were placed into transparent glass cups, coded with numbers, and served to consumers at room temperature together with slices of bread, a plastic knife, a piece of apple, water, and expectoration cups. The tests were done at different sittings on different days, and each consumer tested both samples twice randomly on different days. To score the consumer values, a 5-point hedonic scale (1 = dislike extremely to 5 = like extremely) was used.

2.10 Statistical analysis

The two kind oleogels were prepared at two different times as two replicates, and each replicate production sample were analyzed in triplicate. The collected data were given as mean values with standard deviations. The Analysis of Variance (ANOVA) and Tukey’s and Kruskal-Wallis tests were completed. The level of confidence was at least 95% for all. Statistical analyses were performed with Minitab v.16.1 software.

3 Results and Discussion

3.1 Physico-chemical properties

Some physico-chemical properties of the prepared oleogels were summarized in Table 2. Turmeric-flavoured oleogel (WWO-TM) had higher gelation times (15.0 min) than that of the (11.0 min) red pepper-flavoured oleogel (WWO-RP). Since the addition levels of the spices (1 wt% of overall weight) were the same, the difference must be due to the nature of the spice (its composition, size, morphology etc.). In our previous study, WSW oleogels prepared with sunflower oil at 5 wt% had around 12.3 min gelation time. In this study virgin olive oil (VOO) was used, and similar gelation times (GT) were measured. In wax oleogel literature GT values around 1.0-20.0 min were reported. Since GT strongly depends on organogelator concentration, direct comparison of the data is meaningless, but clearly, WSW yielded oleogels with added spices within reasonably shorter time scales. As a result, adding solid small particles (spices) was not effective on GT and gel formation ability. The oil binding capacity (OBC) values were also similar to the previous findings, and quite high. High OBC values (above 99%) indicate that the wax network formed in the oil was quite effective in immobilizing the liquid oil. Similar results were reported for previously studied wax oleogels.

Solid fat index (SFI) provides information about the ratio of total solid triglycerides in oils at a specified temperature, and if a fat (or margarine) solid at room temperature, it usually contains around 7-16% total solid triglycerides. The oleogels prepared in this study had approximately 3.5% SFI at 20°C, respectively (Table 2). Compared with margarines, the oleogels are fairly solid at 20°C, but include considerably lower levels of total solid fats. The solid contents of the oleogels were mainly results from the added wax and from the saturated triglycerides of the stock oil used. This is, in fact, the main advantage of the oleogels. Oleogels possess solid or semi-solid consistency at room temperature due to the entrapment of liquid oil within the networks of wax crystals. During oleogelation no saturation of fatty acids nor trans isomer formation occurs.

It was well known that the color of oleogels were developed by the oil kind, the organogelator and other ingredients added. In this study, the color and appearance of VOO used, the WSW and spices added could be observed in Fig. 1. Clearly color of the oleogels prepared developed according to the ratios of the ingredients (Table 2). The luminosity (L values) of samples were not significantly different and around 31.2-33.3 for WWO-RP and WWO-TM samples, respectively. Since VOO was a green colored oil, and the spices added were colored samples, the oleogels were less luminous than cream or white colored other solid fats, expectedly. The WWO-RP sample had 3.5 a* value, indicating presence of some red color, and a* color value was much lower (0.8) in the WWO-TM sample. Clearly, red pepper provided some pigment to enhance a* color value. Yellow components of the instrumental color were significantly higher in the WWO-TM sample (12.5) compared to WWO-RP sample (7.2), respectively. Since turmeric itself is

Table 2 The physico-chemical properties of the red pepper and turmeric-flavored virgin olive oil-whale spermaceti wax oleogels prepared.

|                         | WWO-RP          | WWO-TM          |
|-------------------------|-----------------|-----------------|
| Gelation Time (min)     | 11.0 ± 0.1b     | 15.0 ± 0.0a     |
| Oil Binding Capacity (%)| 100.0 ± 0.3b    | 99.9 ± 0.5b     |
| Solid Fat Index (20°C, %)| 3.5 ± 0.1a      | 3.5 ± 0.5b      |
| Color L value           | 31.2 ± 0.6a     | 33.3 ± 0.2a     |
| Color a* value          | 0.8 ± 0.0b      | 3.5 ± 0.1a      |
| Color b* value          | 7.2 ± 0.5b      | 12.5 ± 0.6a     |
| Peroxide Value (meq O₂/kg)| 16.1 ± 1.1a    | 21.1 ± 0.8a     |
| Free fatty Acidity (oleic %)| 1.6 ± 0.2a     | 1.3 ± 0.2b      |

WWO-RP: whale spermaceti wax oleogel with red pepper, WWO-TM: whale spermaceti wax oleogel with turmeric.

*Small letters within each row indicate significant differences among the oleogel samples for the mean ± SD values calculated from six determinations by one-way analysis of variance and Tukey’s test ($p \leq 0.05$).
a yellow-orange colored spice, this color difference is also expected. Overall, color of the oleogels were affected by the color of the added spices in addition to the color of natural VOO used to prepare them. Since olive oil consumers accustomed to accept VOO color, these spreadable VOO samples would be well perceived for their color properties.

Peroxide (PV) and free fatty acidity (FFA) values of all edible fats and oils were regulated worldwide, since higher PV and FFA values indicate degradation of sample, which end up with unhealthy compounds and sensory defects. The Turkish Codex for olive oil permits maximum values of 0.8% FFA and 20 meqO₂/kg oil PV for ‘extra virgin’ olive oil. Consequently, the PV of WWO-TM sample (21.1 meqO₂/kg oil) exceeds the limit value. This could be due to the added turmeric spice, since the other ingredients were the same in both samples. Turmeric may itself had some peroxides or caused oil to oxidize. Like all previously reported oleogel studies, we recommend vacuum or neutral gas atmosphere during oleogel preparation where heat applied (80°C) to melt the waxes at the presence of air. In actual industrial productions, these or other precautions could help to control oil oxidation. Also, some antioxidants could be used. The FFA values of the prepared oleogels (1.6 and 1.4%) also exceeded the codex limits for extra virgin olive oil. But the same codex indicate acceptance of FFA values until 2.0%, and PV values until 20 meqO₂/kg oil for ‘virgin olive oils’ (not extra virgin). Therefore, the prepared oleogels could still be accepted within the standards of virgin quality for olive oils. In fact, the oleogels were new spreadable products and would be acceptable with provided spreadability property, unique flavor and nutritional value by the olive oil consumers or by other consumers consuming spreadable breakfast margarines, as long as the main physico-chemical properties considered.

3.2 X-ray diffraction patterns

X-ray diffraction pattern analysis provides information about the polymorphic forms of solid fats. Polymorphic forms are the different solid phases of the same chemical composition that differ for crystalline structures but yield identical liquid phases upon melting. Basically, four main polymorphic crystals could be formed in edible fats. The glass transition state (γ polymorph) forms as a transition state once the melted fat source rapidly cooled. This form is transparent and unstable. It immediately converts to more stable α polymorph. This form yields fine, waxy, less stable, least dense, lowest melting point crystals with hexagonal chain packing. Intermediate density, medium melting point, orthorhombic chain packing crystals are called the β-type polymorph. This form yields very fine, creamy, soft texture, and immobilizes maximum amounts of liquid oil due to higher surface area. It was observed that under certain conditions, a fat with β' polymorph can transform to β polymorph, which shows the highest melting point, most dense and stable form, and yields coarse and sandy texture. Oil purity, triglyceride molecular forms, cooling rate, storage temperature, shear, and some other factors determine the polymorphic form of an individual fat source. Further, sensory quality and shelf-life of commercial solid fat products were greatly affected by the polymorphic types. Usually β' polymorphic form is preferred in margarine, spreads, and similar products. Sandy, coarse but very stable texture with harsh mouth feelings, is most evident with β polymorphic form, and it is preferred for sugar confectionery and bakery shortenings.

The X-ray diffraction patterns of the prepared oleogel samples were presented in Fig. 2. Pattern evaluations were completed according to official method Cj 2-95(16). The wide-angle (WAXS) region shows the diffracted angles between 6° and 18° and provides information about unit cell dimensions or polymorphism. Further, the size of the scattering object is around 2-10 Å in this region. The small-angle (SAXS) region is about 0.5-6°, and it provides information about lamellar spacing and crystal size. According to the method, a single peak at d = 4.15 Å indicate the α polymorphic form, which shows the highest melting point, most dense and stable form, and yields coarse and sandy texture. Oil purity, triglyceride molecular forms, cooling rate, storage temperature, shear, and some other factors determine the polymorphic form of an individual fat source. Further, sensory quality and shelf-life of commercial solid fat products were greatly affected by the polymorphic types. Usually β' polymorphic form is preferred in margarine, spreads, and similar products. Sandy, coarse but very stable texture with harsh mouth feelings, is most evident with β polymorphic form, and it is preferred for sugar confectionery and bakery shortenings(16, 21, 22).

Fig. 2 The X-ray diffraction patterns of the prepared oleogels (WWO-RP: whale spermaceti wax oleogel with red pepper, WWO-TM: whale spermaceti wax oleogel with turmeric).
polymorphic form, two peaks at positions \(d = 3.8\ \text{Å}\) and \(d = 4.2\ \text{Å}\) indicate the \(\beta\) polymorphic form, and a peak at position \(d = 4.6\ \text{Å}\) indicate the \(\beta\) polymorphic form. Both samples had peaks around 4.5-4.6 Å, together with a single 12.42 Å or 12.91 Å SAXS region peak (Fig. 2). After evaluating all peaks and patterns together, it could be said that the oleogels mostly contain \(\beta\) polymorphic form. Further, peaks at around 2.1-2.4 Å could be due to the spice particles present. Overall, these oleogels could be used as stable spreadable commodities or in food formulations.

3.3 Thermal properties

The DSC determined crystallization and melting onset, peak temperatures and enthalpies were summarized in Table 3. Both crystallization and melting onset and peak temperatures of the WWO-TM sample were significantly higher than those of the WWO-RP sample. Since the same VOO and the same amount of WSW were used to prepare the oleogels, these differences must be caused by the spices added. Both samples melt around body temperature, which could be accepted as a good property for the palate. In our previous study, the melting peak temperature of the 5 wt% WSW oleogels of sunflower oil was 33.4°C [3]. Samples in this study include 8 wt% of WSW, and prepared with VOO including the spices. Hence, the melting profiles seem quite similar and acceptable for spreadable type products [20]. Eventually, these products were fairly solid at room temperature and could be used as spreadable alternatives.

3.4 Rheological properties

At the start of rheological measurements, the amplitude sweep test ranges, it could be accepted as a true gel state. It was stated that storage modulus describes the solid-like or elastic properties of a sample, while loss modulus describes the liquid-like or viscous portions of a sample. Consequently, if a sample provides \(G' > G''\) conditions within the frequency and/or amplitude sweep test ranges, it could be accepted as a true gel. This situation was also explained with loss factor (\(\tan \delta\)) values, and loss factor lower than 0.1 indicates the gel-state [17]. The storage modulus values of the WWP-RP and WWO-TM samples were 100.000-200.000 Pa and 50.000-70.000 Pa, respectively (Fig. 3). Obviously, WWO-RP had higher storage modulus values and hence was more strong gel. Likewise, the loss modulus ranges of the samples were 80.000-100.000 Pa and 32.000-45.000 Pa for the WWO-RP and WWO-TM samples. Further, for both samples within the applied frequency range, no crossover point \((G' = G'')\) was reached, indicating that the samples kept their gelled state through the applied frequency range. Consequently, both samples could have enough long-term stability. This finding is very good to claim suitability of the prepared oleogel samples for distribution as stable gels in the food-supply chain.

To evaluate time-dependent viscoelastic behavior of the prepared oleogels under dynamic-mechanical conditions, a time sweep test was also conducted (Fig. 4). As explained

![Image](https://via.placeholder.com/150)

**Table 3** The thermal properties of the red pepper and turmeric-flavored virgin olive oil-whale spermaceti wax oleogels prepared.

|                      | Crystallization |          | Melting          |
|----------------------|-----------------|----------|------------------|
|                      | Onsetc (°C)     | Peak (Tc, °C) | ΔHc (J/g)      |
| WWO-RP               | 28.6 ± 0.4*     | 26.3 ± 0.2°   | -11.8 ± 0.2°    |
| WWO-TM               | 33.3 ± 0.0°     | 28.4 ± 0.2°   | -10.8 ± 0.2°    |
|                      | Onsetm (°C)     | Peak (Tm, °C) | ΔHc (J/g)       |
| WWO-RP               | 24.6 ± 0.1°     | 35.2 ± 0.1°   | 12.4 ± 0.0°     |
| WWO-TM               | 25.8 ± 0.1°     | 37.2 ± 0.1°   | 12.4 ± 0.2°     |

WWO-RP: whale spermaceti wax oleogel with red pepper, WWO-TM: whale spermaceti wax oleogel with turmeric. *Small letters within each column indicate significant differences among the oleogel samples for the mean ± SD values calculated from six determinations by one-way analysis of variance and Tukey’s test \((p \leq 0.05)\).

![Fig. 3](https://via.placeholder.com/150)

**Fig. 3** The frequency sweep test graphic of the prepared oleogels (WWO-RP: whale spermaceti wax oleogel with red pepper, WWO-TM: whale spermaceti wax oleogel with turmeric).
in the method section, three-time domains were applied to the samples to assess their structural recovery abilities when they were exposed and then released to higher strains than their LVR strains. As could be observed from Fig. 4, at the first region, the samples were in resting state (under LVR strains), and their $G'$ values were higher than their $G''$ values, expectedly. In the second-time domain, the samples were exposed to strains higher than their LVR strains to destruct their structures. In fact, it was achieved since in both samples, the loss modulus ($G''$) values became higher than the storage modulus ($G'$) values. Hence the samples lost their gelled state and became liquid under the applied high shear. On the third-domain, the applied high shear was removed and strains lower than their LVR strains were applied to simulate the structural recovery region. At this region, the storage modulus values enhanced above to their loss modulus values, similar to the initial resting region. Consequently, the samples were re-gelled, after removal of high shear, indicating full structural recovery. This behavior was typical in almost all previously studied wax oleogels\cite{2, 12, 21}. It is well known that food processing operations like mixing, whipping, foaming, transportation etc. could dissipate high energy upon oleogel samples to yield high shear to cause their structures be lost. After ceasing the energy input, the wax oleogels, including the samples in this study could recover their structure to be solid and spreadable again. This could be credited an advantage of the oleogels.

The effects of heating oleogel samples from 0°C to 50°C was tested with a temperature ramp test under continuous frequency and amplitude, and the graphics were presented in Fig. 5. The changes in the storage modulus ($G'$), loss modulus ($G''$), and tanδ values through the heating ramp could be observed in the graphics. Clearly, as temperature increased, both storage ($G'$) and loss ($G''$) modulus values were decreased in both samples, respectively. The thermal DSC data (Table 3) indicated the peak melting temperatures of 35.2 and 37.2°C for the WWO-RP and WWO-TM samples. The temperature ramp data agree with thermal DSC data that the samples were stable until around 35-38°C. For both samples, the storage modulus values were higher than the loss modulus values until the cross-over points ($G' = G''$), where the samples totally melt and freely flows as liquid. In fact, until around 30°C, both samples had fairly linear $G'$ and $G''$ values, indicating that they were quite solid, after that point, they started to soften, and completely melted at around 35-38°C. This data prove that the prepared oleogels could stay as gels during the summer season at room temperature unless not exposed to direct sunlight. If they kept in the refrigerator, they became fairly well solid and still spreadable as well.

### 3.5 Volatile aromatics profile

The volatile aromatics compositions of the WWO-TM and WWO-RP oleogels prepared in this study were listed in Table 4. There were a total of 25 different aromatic compounds identified in both samples. The WWO-TM sample was higher in the volatiles and included 22 different compounds, while WWO-RP sample had only 13 different com-
Table 4: The volatile aromatics composition of turmeric and red pepper-flavored virgin olive oil-whale spermaceti wax oleogels.

| RT* (min) | Volatile Compound | Aroma Definition** | Mean Peak Area | Peak Value (%) | Mean Peak Area | Peak Value (%) |
|-----------|-------------------|--------------------|----------------|---------------|----------------|---------------|
| 1.378     | Ethanol           | Strong alcoholic, ethereal, medical | 126018 ± 1150 | 1.4           | nd.            | nd.            |
| 1.537     | 2-Propanone, 1-hydroxy | Pungent, sweet caramellic, ethereal | nd.           | nd.           | 62770 ± 2300  | 1.1            |
| 1.451     | 2-Propanone       | Solvent, ethereal, apple, pear       | 62180 ± 3005  | 0.7           | nd.            | nd.            |
| 1.833     | Acetic acid       | Sharp, pungent, sour, vinegar        | 396177 ± 23782| 4.4           | 676953 ± 705  | 11.9           |
| 1.921     | Ethyl Acetate     | Ethereal, fruity, sweet, weedy, green | 53943 ± 5811  | 0.6           | 87847 ± 2755  | 1.5            |
| 2.357     | 2-Methylbutanal   | Musty, cocoa, coffee, nutty         | nd.           | nd.           | 11798 ± 855   | 0.2            |
| 2.561     | 1-Pentene-3-ol    | Ethereal, horseradish, green, vegetable | nd.           | nd.           | 63560 ± 4655  | 1.1            |
| 2.595     | 2-Pentanone       | Sweet, fruity and banana-like with a fermented nuance | 46959 ± 605  | 0.5           | nd.            | nd.            |
| 3.695     | 2-Pentanal, (E)- | Pungent, green, fruity, apple, tomato | 32632 ± 1350  | 0.4           | nd.            | nd.            |
| 4.677     | Hexanal           | Fresh green, fatty, aldehydic, grass, leafy | 599703 ± 51800| 6.6           | 523903 ± 555  | 9.2            |
| 6.172     | 2-Hexenal, (E)   | Green, banana, aldehydic, fatty, cheesy | 1559863 ± 845850| 17.2          | 845449 ± 7350  | 14.8           |
| 6.291     | 3-Hexen-1-ol, (Z) | Green, grassy, melon rind-like with a pungent freshness | 72164 ± 1150  | 0.8           | nd.            | nd.            |
| 6.636     | 2-Hexen-1-ol, (E) | Fresh, fatty, green, fruity, vegetative, with leafy and herbal nuances | 199662 ± 14965 | 2.2           | 109288 ± 8650  | 1.9            |
| 7.352     | Styrene           | Sweet, balsam, floral, plastic      | 43726 ± 1005  | 0.5           | nd.            | nd.            |
| 9.781     | trans-2-Heptenal  | Intense green, fatty, oily, with fruity overtones | 38140 ± 3572  | 0.4           | nd.            | nd.            |
| 11.020    | beta-Myrcene      | Peppery, terpene, spicy, balsam     | 88936 ± 17005 | 1.0           | 51202 ± 487   | 0.9            |
| 12.513    | dL-Limonene       | Citrus, herbal, terpene, camphor    | 2874936 ± 751555 | 31.7         | 1590232 ± 28300 | 27.8          |
| 13.227    | beta-Ocimene      | Green, tropical, woody with floral and vegetable nuances | 71439 ± 3800  | 0.8           | 29332 ± 3745  | 0.5            |
| 15.941    | Nonanal           | Waxy, aldehydic, rose, fresh, orris, orange peel, fatty, peely | 105370 ± 13745 | 1.2           | 43799 ± 4003  | 0.8            |
| 27.393    | alpha-trans-Bergamotene | Woody, warm tea | 28532 ± 595  | 0.3           | nd.            | nd.            |
| 27.626    | beta-Cedrene      | Cedarwood, woody                   | 55898 ± 1200  | 0.6           | nd.            | nd.            |
| 28.941    | ar-Curcumene      | Herbal                            | 83538 ± 1045  | 0.9           | nd.            | nd.            |
| 29.701    | Farnesene <(E,E)-, alpha- | Citrus, herbal, lavender, bergamot, myrrh, neroli, green | 137402 ± 3468 | 1.5           | 48489 ± 5462  | 0.9            |
| 30.314    | beta-Sesquiphellandrene | Herbal, fruity, woody | 75562 ± 7045  | 0.8           | nd.            | nd.            |
| 34.520    | ar-Turmerone      | Herbal, turmeric                   | 57907 ± 585   | 0.6           | nd.            | nd.            |

*RT: retention time, **Aromatic definitions of the volatile compounds were found from the web page: https://www.thegoodscentscopy.com/index.html#

Both oleogels were prepared from VOO, WSW and the spices at the given weight proportions; hence, the volatiles identified must be originating from the ingredients used to prepare them, respectively. VOO is a virgin and aromatics rich oil, and more than 100 volatiles are listed for VOO in the book of Boskou(10). After screening VOO volatiles listed in the book and Table 4 together, the aromatics coming from the oil were identified. Consequently, in oleogel samples, the aromatics ethanol, acetic acid, ethyl acetate, 2-methylbutanal, 2-pentenal, hexanal, 2-hexenal, 3-hexen-1-ol, 2-hexen-1-ol, trans-2-heptanal, and nonanal were coming from the VOO, respectively. Most of these aro-
matics were defined with green, fruity, musty, fatty, leafy, tomato, and sour aroma definition terms (Table 4). In fact, these descriptions were also used for virgin olive oils. Since the prepared oleogels resemble the aroma profile of VOO, it would be good for VOO consumers to perceive the loved virgin olive oil aroma from the prepared oleogels. Further, some aromatics must be originating from the added spices, turmeric and red pepper (paprika). The volatile compositions of different Curcuma species (turmeric) were studied and the major components were listed as turmerone, curlone, ar-turmerone, sesquiphellandrene, zingiberene, germacrone, terpinolene, ar-curcume, and phellandrene. In WWO-TM sample, the co-existing aromatics of beta-myrcene, ar-curcume, beta-sesquiphellandrene, and ar-turmerone were identified. These volatiles provide terpene, herbal, woody and balsam aroma notes. Since the spices were added only at 1 wt% of overall weight of the oleogel, and VOO is very aromatic rich oil, less kinds and quantities of spice aromatics were measured in the oleogel samples. Similarly, aromatics of red pepper (paprika) including oleogel (WWO-RP) were compared with the volatile compositions of various red pepper samples measured and published. The authors identified around 136 different volatiles in different types of red pepper samples, and listed the most important compounds as beta-myrcene, p-cymene, alpha-thujene, alpha-limonene, (E)-2-hexenal, heptanal, (Z)-2-heptenal, octanal, (E)-2-nonenal, nonanal, hexanal, 2-methylpropanal, 2- and 3-methyl butanal, ethanol, 3-methylbutanol, (E)-2-octen-1-ol, (Z)-2-penten-1-ol, 2-ethyl-1-hexanol, 3-hydroxy-2-butanone, 6-methyl-3,5-heptadien-2-one, isobutyl butyrate, and acetic acid. The similar compounds determined in the WWO-RP sample were ethanol, acetic acid, 2-methylbutanal, hexanal, trans-2-heptanal, beta-myrcene, limonene, and nonanal. These volatile compounds were usually associated with ethereal, pungent, musty, green, peppery, herbal, and peely aroma descriptions.

The WSW used as the organogelator was a faint, odorless, white flake (Fig. 1). There is no data about the volatile constituents of the WSW to compare with the samples. In the oleogel samples, nonanal was present with its waxy aroma description. Also, styrene was quantified in one sample, and defined as plastic. These two compounds may be originating from the WSW, in addition to the common fatty described volatiles like hexanal, 2-hexenal, 2-hexen-1-ol, trans-2-heptanal, and nonanal. Of course, these fatty compounds were also present in the olive oil used. Since waxy aroma is not liked much in oleogels, its absence in WSW could be accepted as an advantage over the other waxes.

The peak% values of the identified volatile compounds were also provided in Table 4. The highest concentrations of limonene (31.7 and 27.8%), 2-hexenal (17.2 and 14.8%), hexanal (6.6 and 9.2%), and acetic acid (4.4 and 11.9%) were measured in the WWO-TM and WWO-RP samples, respectively. These peak% values represent the concentrations of the compounds in the samples, but in aroma science, the concentration of an individual compound is not directly related to its perceived aroma intensity by humans. To describe perception intensity of an aromatic volatile compound, the odor/aroma/threshold values must be known for each combination. Odor threshold was defined as the minimum concentration at which human subjects perceive the aroma. It was also well known that some volatiles produce very sensible aromas at very low concentrations, and some volatiles could be perceived only at very high concentrations. Thereby, there is no direct relation of its existing concentration with the aroma potency for any aromatic volatile. Characteristics aroma of a food sample is rather a cumulative effect of all aromatic volatiles present. Further, the matrix in which the volatile compound exists greatly affects the release and perception of the aroma. Odor threshold values of some pure compounds in water or oil media have been published, but most of them are still unknown. Therefore, aromatic volatile analysis of oleogel samples could only provide information about the existence and amount of the aromatic volatile compounds, but not their human perceptions. Therefore, sensory analyses must be completed to understand and manage the product formulation and processing to end up with successful products.

3.6 Sensory descriptions

A trained sensory panel has defined the oleogel samples with 12 sensory descriptor terms, and the results were given in Table 5. The sensory description terms, definitions, and references were presented in Table 1, and the analysis procedures were explained in the method section. Sensory `hardness’ was defined as the force required pushing a knife into the sample, and yoghurt and tallow were used as the references for minimum and maximum intensities. Both oleogel samples had around 6.0 score, indicating that the samples were harder than yoghurt and softer than tallow. In fact, they were just similar to spreadable kitchen margarines. This sensory data was also following the rheological data provided in Fig. 3. Both samples had full ”spreadability”, as tested by deploying sample over a bread loaf. For the purpose of developing spreadable VOO oleogels, this finding is perfect. The ’liquefaction’ was defined as the amount of liquid oil release during the spread of oleogel on the bread surface. It measures sample melting by kinetic energy input. Both samples had a quite low melting. In fact, we know from the oil binding capacity tests that for any reason, if the oleogels melt, and then if the reason of melting (kinetic energy or heating) ceased, the melted gels re-solidify. This is the well-known and mostly credited phenomenon of the oleogels. Depending on the purpose of solid fat applications, different levels
Spice-flavored Oleogels

Table 5 The sensory quantitative descriptive analysis (QDA) results of the red pepper and turmeric-flavored virgin olive oil-whale spermacteri wax oleogels prepared.

|                     | WWO-RP        | WWO-TM        |
|---------------------|---------------|---------------|
| Hardness            | 6.1 ± 0.5*    | 6.0 ± 0.5*    |
| Spreadability       | 10.0 ± 0.0    | 10.0 ± 0.0    |
| Liquefaction        | 2.3 ± 0.1     | 2.0 ± 0.1     |
| Sandiness           | 1.5 ± 0.5     | 1.0 ± 0.5     |
| Olive fruit         | 5.3 ± 0.2     | 4.8 ± 0.3     |
| Grassy              | 4.2 ± 0.3     | 4.6 ± 0.3     |
| Waxy                | 0.5 ± 0.0     | 0.5 ± 0.0     |
| Rancid              | 0.5 ± 0.0     | 0.5 ± 0.0     |
| Bitter              | 5.0 ± 0.0     | 0.0 ± 0.0     |
| Hay                 | 2.8 ± 0.5     | 2.6 ± 0.2     |
| Cooling             | 1.5 ± 0.5     | 2.5 ± 0.5     |
| Mouth coating       | 5.4 ± 0.5     | 5.8 ± 0.5     |

WWO-RP: whale spermacteri wax oleogel with red pepper, WWO-TM: whale spermacteri wax oleogel with turmeric.

* Small letters within each row indicate significant differences among the oleogel samples for the mean ± SD values calculated from six determinations by one-way analysis of variance and Kruskal-Wallis test ($p < 0.05$).

Aroma notes must be coming from VOO and the spices as well. There were very small scores of ‘waxy’ measured in both samples (0.5), compared to 10 score of paraffin wax used as the reference. This finding was also well correlated with the aromatic volatile data (Table 4), indicating few compounds described with waxy aroma descriptions. In fact, this could be accepted one advantage of WSW, compared to other plant and animal waxes, which yielded higher waxy scores$^{11-13}$. The ‘rancid’ scores of the samples (0.5) were very low, and no extensive oxidation occurred during oleogel preparation. This could be attributed to natural stability of VOO and to the added spices, which provide many anti-oxidant compounds. The ‘bitter’ attribute was determined (5.0) in the WWO-RP sample, but not in the WWO-TM sample, expectedly. Since WWO-RP sample had only 1 wt% of red pepper added, the bitter score reduced to half value of the initial red pepper. Of course WWO-TM sample did not have any bitterness. ‘Hay’ was defined as he aromatics associated with dry grasses, and scores of 2.8 and 2.6 were measured in the samples. The aromatics defined with hay or similar terms were usually associated with the added spices. For any solid fat including cocoa butter, a ‘cooling’ effect was observed once the sample melts in mouth space. The cooling score of WWO-TM sample was significantly higher than that of the WWO-RP sample, and this must be caused from some compounds released from turmeric into the oleogel, since all ingredient were the same. Lastly, panel defined ‘mouth coating’ as the perceived fatty coating over palate with the maximum value referenced by butter. Obviously, the oleogels had almost half value mouth coating compared with butter. Overall, the panel sensory descriptions of the oleogel samples mostly in agreement with the physical and/or instrumental measurement data, and could provide information for interested readers. Since market success of any food product is heavily dependent on consumer perceptions, we conducted a limited number consumer test for the prepared oleogel samples, in addition to the sensory descriptive analyses and aromatic profile data.

3.7 Consumer tests

With 50 volunteer consumers, and 5-point hedonic scale (1 = dislike extremely to 5 = like extremely) the oleogel samples were evaluated (Table 6). For appearance, spreadability, and acceptability, WWO-RP sample had significantly higher scores than WWO-TM sample. Although the differences were statistically significant, they were not as large as one point. Both samples had very high appearance (4.4 and 4.3) scores, indicating that the consumer liked the scene of the oleogels. Sample spreadsabilities were a little lower (3.9 and 3.7), but still above the neutrality (neither like-nor dislike, 3.0) point. Hence, improvement of spreadability to fulfill consumer demand was apparently identified. Aroma (4.1 and 4.2) and flavor (4.3 and 4.2) scores were in the liked-region by the consumers for the WWO-RP and WWO-TM samples. Total acceptability of WWO-RP
sample (4.2) was higher than that of the WWO-TM sample (4.1). Overall, the spice-flavored oleogels were liked and accepted by the consumers. Further studies with a higher number of participating consumers and some product informations provided before the test to the participants might be conducted to gain more information about the consumer attitudes towards the new oleogels. Then, necessary corrective actions (adjustment of ingredient ratios and/or kinds, processing parameters etc.) could be taken.

4 Conclusions

In this study, virgin olive oil (VOO)-whale spermaceti wax (WSW) oleogels were prepared for the first time in the literature with added spices (turmeric and red pepper). Common physico-chemical characterization analyses, thermal analysis, rheology, aromatics volatiles compositions, sensory descriptive analysis and consumer tests were provided. Since these types of comprehensive data for oleogels are still accumulating in literature, the data and discussion provided with this study would be important. Results pointed out that WSW was quite good in forming stable and rheologically proper oleogels. The liked flavor and health benefits of the VOO and the spices could be delivered to consumers, who may like to consume liquid olive oil in spreadable form. Further, the added spices could provide bio-active substances with functional food properties and antioxidant molecules, in turn, may extent the shelf-life of the prepared oleogels during storage against oxidation. Also, it must be kept in mind that during the oleogelation process, the oil and added wax heated up to 80°C to melt and completely mix the wax, and meantime the spices were added and vigorously homogenized. Hence, at that temperature the microbial load of the spices must greatly be reduced to provide another advantage of the spices.

Table 6 The consumer test results of the red pepper and turmeric-flavored virgin olive oil-whale spermaceti wax oleogels prepared.

|                      | WWO-RP | WWO-TM |
|----------------------|--------|--------|
| Appearance           | 4.4 ± 0.6* | 4.3 ± 0.9* |
| Spreadability        | 3.9 ± 0.9a | 3.7 ± 0.9b |
| Aroma                | 4.1 ± 0.8a | 4.0 ± 0.8a |
| Flavor               | 4.3 ± 0.7a | 4.2 ± 0.8a |
| Acceptability        | 4.2 ± 0.8a | 4.1 ± 0.7b |

WWO-RP: whale spermaceti wax oleogel with red pepper, WWO-TM: whale spermaceti wax oleogel with turmeric.

*Small letters within each row indicate significant differences among the oleogel samples for the mean ± SD values calculated from six determinations by one-way analysis of variance and Kruskal-Wallis test (p ≤ 0.05).

Technology. Similar products with other spices (garlic, onion, thyme, rosemary etc.) and different wax concentrations could be prepared and extensively evaluated to provide new olive oil products for the extending markets. In conclusion, healthy, tasty, spreadable VOO oleogels could be prepared successfully. Attention of WSW producers to provide this wax in food-grade or GRAS situation was also anticipated.

Conflict of Interest

The authors have declared no conflict of interest.

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