



Optimizing the Utilization of Tangerine Peels Using Supercritical CO₂ Extraction and Spray-drying to Prepare Novel Functional Instant Tangerine Beverage



Takwa M. Elgohary^{a,b*}, Hammam E. M. Bahlol^a, Manal M. Ramadan^b, Mohamed K. Morsy^a, Rasha S. Mohamed^c

^aDepartment of Food Technology, Faculty of Agriculture, Benha University, 13736 Qalubia, Egypt

^bChemistry of Flavour and Aroma Department, National Research Centre, Cairo, Egypt

^cNutrition and Food Sciences Department, National Research Centre, Cairo, Egypt

Abstract

Thus, returning to the food processing chain as an additive and providing innovative and sustainable methods for using food waste, citrus peel, a waste product of the citrus processing sector, shows potential economic benefits. This study aims to maximizing the utilization of tangerine peels (TP) to offer a new formulation of a functional instant beverage made of different tangerine peels extracts (oil, ethanol and aqueous extracts). Supercritical carbon dioxide (SC-CO₂), a green extraction method, was used to extract the TP oil. Ethanol and aqueous extracts were prepared using 70% ethanol and hot water respectively. Antioxidant activity (by DPPH, ABTS, reducing power methods), total Phenolic and flavonoid of all extracts were determined colorimetrically. Volatile and non-volatile compounds were determined using GC-MS and HPLC. The new tangerine beverage was made using an equal amount of ethanol and aqueous extracts (1g%) and 0.1g% oil. The instant drink was made into a powder using the microencapsulation process by the spray drying method; a zeta-sizer and an electron microscope were used to assess the powder's encapsulation efficiency. Tests were conducted to evaluate colour, flavour, taste, and general acceptability. According to the results, all extracts (oil, ethanol, and aqueous) have strong antioxidant properties and include a sizable number of bioactive substances. Therefore, the new functional beverage would be beneficial to human health, and this powdered quick drink may be produced commercially and help reduce pollution caused by the inappropriate disposal of these leftovers.

Keywords: Tangerine peel, Encapsulation, instant drink, spray drying, supercritical CO₂, GC-MS and HPLC.

1. Introduction

Large amounts of fruit waste by-products are created during the production and processing of food in developing countries, which is expensive and has an adverse impact on the environment. Globally, the agri-food industry produces over 190 million tones of byproducts per year. Molasses, oilseed cakes, bran, pomaces, leaves, seeds, shells, and ruined raw materials are a few examples of processing-related byproducts [1]. One side of the argument focuses on wastes and by-products from the agri-food business, especially the fruit industry, which ought to be categorized as wastes but aren't able to be disposed of as such because of environmental concerns. However, when viewed through the lens of sustainability and a circular economy, they provide valuable, bioactive, and healthy substances that may be recovered and utilized as raw materials for other products, helping to achieve the global goal of "zero waste" in the environment [2]. Because of their high concentration of essential oils and bioactive components, citrus peels that are left over after processing can be a significant source of nutrients and antioxidants. With growing awareness of environmental issues, there has been a lot of interest in using supercritical carbon dioxide extraction (SC-CO₂) to extract bioactive components, particularly essential oils, from different types of plant [3]. This method has some advantages over traditional approaches, primarily due to its distinct physical characteristics. SC-CO₂ can be used as a substitute for traditional solvents in a variety of industrial and scientific procedures. Compared to other traditional techniques like solvent extraction and distillation, SC-CO₂ offers the advantages of automation, fewer operational steps, and safe operations because it uses a moderate temperature in the critical range that is ideal for thermally labile compounds, which results in an exceptionally high-quality final product [4]. The edible and traditional medicinal raw material known as tangerine peel (TP), which comes from the tangerine fruit (*Citrus reticulata*), can be used to treat a variety of conditions, including indigestion, inflammation, and chest and abdominal fullness. Flavonoids, phenolic acid, pectic polysaccharides, and other components are abundant in TP and provide it with many of useful qualities, including antiviral, anti-inflammatory, and anticancer effects [5]. Water and electrolyte balance are maintained by beverages, which are an essential component of the human diet. Every socioeconomic group consumes them. Instant beverages have several significant benefits, such as a multi-component composition, a long shelf life, and a convenient commodity form. Because they may be utilized in nearly any situation, they are a crucial component of prepared meals for those who labour in harsh environments [6]. The most popular technique for turning liquid raw materials into food granules is spray drying. It is commonly recognized that using drying carriers is an effective way to speed up the drying process and enhance the properties

*Corresponding author e-mail: takwamagdy241@gmail.com; (Takwa M. Elgohary).

Receive Date: 03 February 2025, Revise Date: 25 March 2025, Accept Date: 26 March 2025

DOI: 10.21608/ejchem.2025.357835.11249

©2025 National Information and Documentation Center (NIDOC)

of powders [7]. Customers may be drawn to tangerine drink powder manufactured with TP bioactive extracts if it is promoted with an emphasis on flavour, taste, and eye-catching colour. As an alternative to the liquid form, powdered TP extracts have a longer shelf life, less mass and volume, and are easier to handle. Therefore, this study goals were to evaluate TP oil extracted by SC-CO₂, estimate TP antioxidant activity, and identify phenolic and flavonoid components in various TP extracts. It also aimed to create a popular quick TP drink. Our target is to produce instant drinks from tangerine byproducts (peels) with natural flavors (in the form of powder) free of preservatives and artificial colors as healthy alternatives to their industrial counterparts spread in the local markets.

2. Materials and methods

2.1. Chemicals and reagents

• Tangerine peels were acquired from the El-Marowa firm, a juice concentrate manufacturer located in the 6th of October industrial city in Giza, Egypt. Arabic gum (AG) and maltodextrin (MD) were acquired from Loba Chemie, a company based in Mumbai, India. The reagents DPPH (1,1-diphenyl-2-picrylhydrazyl), ABTS (2,2-azino-bis(3-ethyl-benzothiazoline-6-sulfonic acid), and Folin-Ciocalteu were acquired from Sigma-Aldrich, Inc. located in Louis, USA. Chemicals and reagents employed were of special analytical quality.

2.2. Extraction of the TP oil

- After rinsing with tap water, TP was sliced into tiny pieces and sun-dried for 48 hours. An electric grinder was used to reduce the dried tangerine peels to a uniformly fine powder. Subsequently, the powdered TP was kept out of direct sunlight at room temperature in an airtight container labeled clearly for later use.
- Supercritical carbon dioxide (Sc-CO₂) green extraction method (Speed-TM SFE-2, applied separation, built in conjunction with the USDA1 - USA) was used to extract TP oil, following the procedure outlined by Xiong and Chen [8]. To keep TP particles from clogging, 200g of dried samples were poured into a stainless steel extraction jar and covered with glass wool on both the top and bottom. The operation panel's micro-metering valve was adjusted to precisely regulate the CO₂ extraction temperature, pressure, and volume flow rate (10 ml/min). The extraction duration, pressure, and temperature were all set at 180 minutes, 140 bar, and 45°C, respectively. The yield of TP oil was calculated using the following equation:
- Yield (%) = $M_a/M_b \times 100$
- Where M_a is TP oil (g) while M_b is TP powder (g). The oil was stored at 4°C in a clean dark glass vial for further investigation.

2.3. Gas chromatography-mass spectrometry (GC-MS) analysis

- Gas chromatography system was used to analyze the tangerine peel oil. An HP-5MS fused silica capillary column (30 m, 0.25 mm i.d., 0.25 mm film thickness) and an Agilent 8890 GC System gas chromatography system were used to analyze the tangerine peel oil. The oven temperature was initially set to 50°C and was then programmed to rise to 220°C at a rate of 5°C/min, and then to 280°C at a rate of 15°C/min, before staying at 280°C for 7 minutes. Helium was employed as the carrier gas, with a flow rate of 1.1 mL/min. The essential oil was diluted in diethyl ether (30 µl essential oil / ml diethyl ether) before injecting 1 µl of the solution into the GC with a split ratio of 1:50. The injection temperature was 230°C, and mass spectra in the electron impact mode (EI) were recorded at 70 eV, with scan m/z spanning from 39 to 500 amu. The separated peaks were disclosed by cross-referencing them with mass spectra data from the National Institute of Standards and Technology's library [9].
- **2.4. Preparation of the ethanol extract**
- TP ethanol extract was made by liquefaction 25 g of the dried peels in 500 ml of 70% ethanol in a beaker. The sample was agitated and macerated for 48 hours at room temperature to dissolve all soluble components. The solvent was eliminated with a rotary evaporator (BÜCHI Labortechnik AG, Flawil, Switzerland) at $55 \pm 1^\circ\text{C}$, then at $60 \pm 1^\circ\text{C}$ under reduced pressure after being filtered with wattman filter paper (No. 1). The extract was then weighed and chilled in preparation for additional analysis.
- **2.5. Preparation of the aqueous extract**
- For preparation of the TP aqueous extract, 25 g of the dried peels were added to a beaker with 500 ml of hot water. After being shaken continuously for 24 hours, the sample was soaked to dissolve all soluble components. The mixture was concentrated after being filtered through wattman filter paper (No. 1). The extract was then weighed and chilled in preparation for additional analysis.

2.6. High performance liquid chromatography (HPLC) of TP extracts

The HPLC analysis was performed using an Agilent 1260 series. For quantitative determination of phenolic compounds in ethanol and aqueous extracts [10]. For separation, an Eclipse C18 column (4.6 mm x 250 mm i.d., 5 µm) was employed. The mobile phase consisted of water (A) and 0.05% trifluoroacetic acid in acetonitrile (B) at a flow rate of 0.9 ml/min. The following sequence of 0 minutes (82% A), 0–5 minutes (80% A), 5–8 minutes (60% A), 8–12 minutes (60% A), 12–15 minutes (82% A), 15–16 minutes (82% A), and 16–20 minutes (82% A) was programmed for the mobile phase. The multi-wavelength detector was observed at 280 nm. The injection volume for every sample solution was 5 µl. At 40 °C, the column temperature was maintained consistently.

2.7. Preparation of tangerine emulsion (non-encapsulated beverage)

- Distilled water (1 Liter) was mixed with 10 g of ethanol extract and 10 g of aqueous extract from TP. The use of Tween-80® (1.0% w/v) helped to produce the emulsion. Drop by drop, a volume of 1 ml of TP oil was supply to the aqueous solution.
- **2.8. Preparation of the instant powder of tangerine emulsion (encapsulated beverage)**
- The instant beverage powder was prepared through the encapsulation technique described by Mahdi, Al-Maqtari [11]. For the entire night, 5% w/v Arabic gum was soaked in deionized water at 4°C. The gummy solution was combined with

10% maltodextrin, equal amount (1%) of ethanol, and aqueous TP extract. Tween-80® (1.0 w/v) was then added to assist with generate the emulsion. TP oil (1 ml) was added drop by drop to the aqueous solution (1 litre). A high-speed homogenizer (Ingenieurbüro CAT, Germany) was employed to combine the emulsion for 5 min at 20,000 rpm. Using a two-fluid nozzle spray drier (Büchi Mini Spray drier B-290, Switzerland) with an external ring aperture of 1.5 mm and an internal tip opening of 0.5 mm, the emulsion was lyophilized immediately. One of the constant process variables was the drying air flow rate of 80% of the suction fan controller. The intake and output temperatures were 160 ± 5 °C and 80 ± 5 °C, in that order. The resulting powder was taken out of the drying chamber wall and the cyclone, and it was stored at 4°C for further study.

• **2.9. Dynamic Light Scattering (DLS) analysis**

• The polydispersity index (PDI), ζ -potential, and particle size distribution of the tangerine emulsion at 25 ± 0.1 °C were evaluated using Zeta-sizer (ZS90, Malvern Instrument, UK). The emulsion sample was diluted (1:10 v/v) with distilled water. Following that, the mixture was subjected to a little sonication. Measurements were conducted on the sample (ml) after it had been transferred to a disposable transparent PVC cuvette.

• **2.10. Encapsulation efficiency determination**

• According to Zahran, Catalkaya [12] and Dwivedy, Singh [13], the UV-Vis spectrophotometer (Jasco V-730, serial No. A 112361798, Japan) was used to calculate the encapsulation efficiency (EE). An assessment was made of the amount of tangerine peel oil present in the surface of the instant beverage powder by extracting it from 10 mg of lyophilized powder and comparing its absorbance to a reference curve created for the oil at 259 nm. The following equation was used to compute the EE:

• $EE (\%) = (\text{total amount of oil in the powder} - \text{the surface (nonencapsulated) oil content}) / \text{total amount of oil in the powder} \times 100$

• **2.11. Scanning electron microscope (SEM) of the instant beverage powder**

• In order to look into the surface structure of the instant beverage powder, a high-resolution scanning electron microscope (SEM) model with a field emission gun (TESCAN VEGA 3 in the Czech Republic) was used. Gold was applied to the samples using a Quorum Q 150 ES from the United Kingdom for 60 seconds.

• **2.12. Transmission electron microscope (TEM) of the instant beverage powder**

• After coating the sample with gold (DST3, Nanostructured Coating Co., Tehran, Iran), the shape of instant beverage powder was studied and seen using transmission electron microscopy (TEM) (JEM-2100 Electron Microscope Instruments, China).

• **2.13. Determination of the total phenolic, flavonoids and antioxidant activity**

• **2.13.1. Total phenolics assay:**

• Total phenolic content was estimated according to the method described by [14]. The extract (50 μ L) was mixed with 250 μ L of Folin-Ciocalteu reagent after the volume was adjusted to 3.5 mL with distilled water. After five minutes, 0.5 mL of a 20% aqueous sodium carbonate (NaCO₃) solution was added to the liquid to neutralize it. The absorbance was measured at 765 nm with respect to the solvent blank after 40 minutes. Using a calibration curve made using gallic acid, the total phenolic content was calculated and represented as μ g of gallic acid equivalent (μ g GAE) per ml of sample.

• **2.13.2. Total flavonoids assay:**

• Total flavonoid content was estimated according to [15]. In summary, 50 μ L of extract was combined with 300 μ L of 5% sodium nitrite (NaNO₂). After incubating for 6 minutes, 300 μ L of a 10% AlCl₃ solution was added, and distilled water was added to bring the amount to 1.80 mL. 1.5 mL of 1 M NaOH was added to the mixture, and after seven minutes, it was centrifuged at 5000 g for ten minutes. At 510 nm, the supernatant's absorbance was measured in relation to the solvent blank. Using a calibration curve created using catechin, the total flavonoid concentration was calculated and represented as micrograms of quercetin equivalent (μ g QE) per ml of sample.

• **2.13.3. DPPH radical scavenging activity:**

• DPPH (2,2-diphenyl-1-picrylhydrazyl) radical scavenging activity was assessed using the modified methods of (Shimada, Fujikawa et al. 1992) and (Mensor, Menezes et al. 2001). DPPH was finally concentrated to 200 mM, and the reaction volume was 3.0 mL. After 60 minutes of dark incubation, the absorbance was measured at 517 nm against a blank of pure methanol. The inhibitory percentage of DPPH radicals was calculated according to the following equation:

• $\text{Scavenging activity (\%)} = [1 - ((\text{absorbance of sample}) / (\text{absorbance of control})) \times 100\%]$

• **2.13.4. ABTS radical scavenging activity:**

• ABTS (2, 2'-azinobis (3-ethylbenzthiazolin-6-sulfonic acid) radical scavenging activity was assessed according to [16]. Equal parts of an aqueous solution of ABTS containing 7 mM was reacted with 2.45 mM potassium persulfate for 16 hours at room temperature (25°C) in the absence of light to create the stock solutions of ABTS reagent. Then, using a spectrophotometer, the working solution was created by diluting one milliliter of ABTS solution with sixty milliliters of ethanol: water (50:50, v/v) to achieve an absorbance of 1.0 ± 0.02 units at 734 nm. For one hour in the dark, extracts (50 μ L) were allowed to react with 4.95 mL of the ABTS solution. The spectrophotometer was then used to measure the absorbance at 734 nm. Trolox was used to prepare the standard curve. The inhibitory percentage of ABTS radicals was calculated according to the following equation:

• $\text{Scavenging activity (\%)} = [1 - ((\text{absorbance of sample}) / (\text{absorbance of control})) \times 100\%]$

• **2.13.5. Reducing power:**

• The reducing power was assessed using reducing power method described by [17]. Ascorbic acid was used as standard, and results were expressed as μ g ascorbic acid equivalents per mL (μ g ASCE/mL).

• **2.14. Sensory evaluation**

• Twenty participants assessed the freshly made non-encapsulated tangerine beverage's sensory characteristics (color, taste, flavor, and viscosity), according to [18]. The beverage was given to each participant in an unlabeled transparent cup under

white light, and they were instructed to assess each sensory quality on a scale of 1 to 25 on the accompanying provided sheet. The scores that were obtained were used to calculate each beverage's overall acceptability.

• 2.15. Statistical analysis

The SPSS software (version 21) was used in the statistical analysis. One-way ANOVA and the Duncan test were used to statistically examine the data, which were presented as mean \pm standard deviation (SD). When a difference was $P < 0.05$, it was considered statistically significant.

3.Results and discussion

3.1.GC-MS fingerprint of TP oil

Figure 1 and Table 1 identified 15 volatile compounds. The main ingredients in tangerine essential oil were d-limonene (70.16%) and γ -terpinene (12.65%). These findings are consistent with those of Naef [19], who discovered that the main component of TP oil was limonene (95.2%). Additionally, [20] discovered that the two main constituents of tangerine oil were γ -Terpinene (10.86%) and d-limonene (74.47%). In tangerine cold-pressed oil, the major hydrocarbon was limonene (95.1%) while γ -terpinene was not detected [21]

User Chromatograms

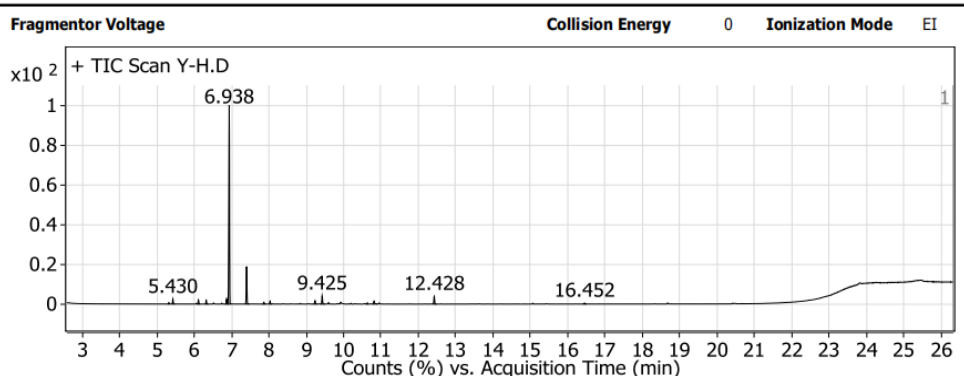


Fig. 1. GC-MS chromatogram of TP oil.

Table 1: Bioactive volatile compounds(%) of TP oil

Compounds	RT	%
Bicyclo[3.1.0]hex-2-ene, 2-methyl-5-(1-methylethyl)-	5.318	0.47
Alpha.-Pinene	5.43	1.54
Beta.-Pinene	6.113	1.32
Beta.-Myrcene	6.321	1.21
p-Cymene	6.867	1.69
D-Limonene	6.938	70.16
Gamma.-Terpinene	7.401	12.65
Cyclohexene, 1-methyl-4-(1-methylethylidene)-	7.864	0.65
Linalool	8.03	0.95
Terpinen-4-ol	9.235	1.25
Alpha.-Terpineol	9.425	2.89
1-Cyclohexene-1-carboxaldehyde, 4-(1-methylethenyl)-	10.63	0.42
Thymol	10.81	1.46
Benzoic acid, 2-(methylamino)-, methyl ester	12.42	2.89
Alpha.-Sinensal	16.45	0.44

3.2. The constituents of TP ethanol and aqueous extracts

The HPLC detected 17 and 18 distinct phenolic components in the TP aqueous and 70% ethanol extracts, respectively. The results in Figure 2 and Table 2 show that TP has a considerable amount of total phenolics. Chlorogenic acid, quercetin, coumaric acid, catechin, hesperetin, and rosmarinic acid were the main constituents in the aqueous extract. whereas naringenin, gallic acid, rosmarinic acid, chlorogenic acid, and catechin were the main constituents in the ethanol extract. According to the findings, the ethanol extract had greater levels of phenolic chemicals than the aqueous extract [22] verified that, when compared to using water or organic solvents alone, the combination of an aqueous solvent (distilled water) with an organic solvent (methanol or ethanol) increases the yield of bioactive chemicals. When it comes to extracting phenolic chemicals, alcohol mixtures containing varying amounts of water have proven to be more successful than mono-component solvent systems [23]. Addition of small quantity of water to organic solvent usually facilitates the polyphenols extraction [24]

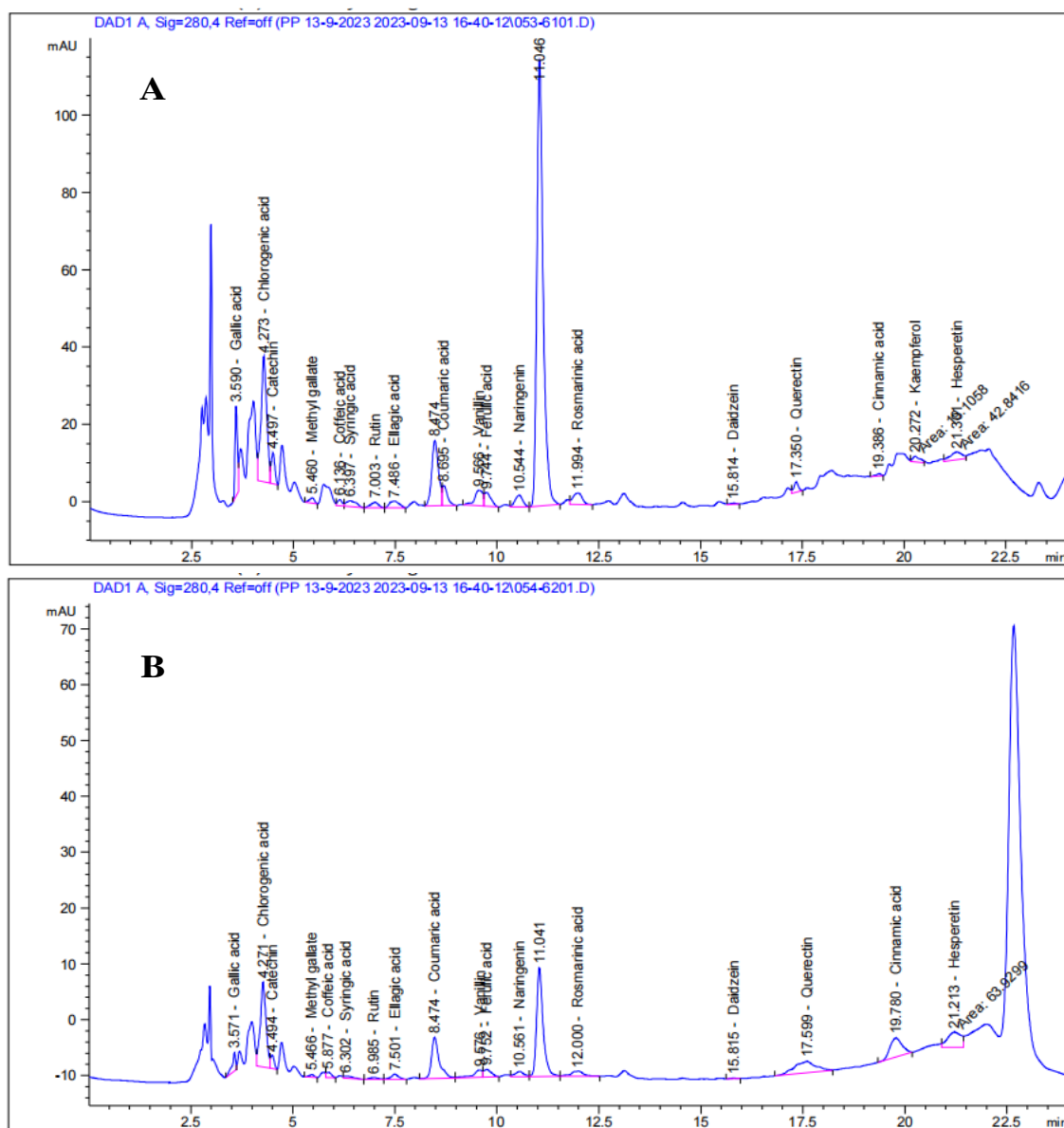


Fig. 2. HPLC chromatograms of TP ethanol (A) and aqueous (B) extracts.

Table 2: Chemical constituent's concentration ($\mu\text{g/g}$) of TP ethanol and aqueous extracts using HPLC

	Aqueous extract ($\mu\text{g/ml}$)	70% Ethanol extract ($\mu\text{g/ml}$)
Gallic acid	87.37	422.56
Chlorogenic acid	1008.47	2156.56
Catechin	167.38	567.00
Methyl gallate	10.25	31.99
Coffeic acid	34.51	50.97
Syringic acid	23.08	98.23
Rutin	34.91	154.89
Ellagic acid	48.80	129.06
Coumaric acid	179.18	86.30
Vanillin	37.84	108.23
Ferulic acid	52.46	121.71
Naringenin	66.19	191.28
Rosmarinic acid	114.41	254.61
Daidzein	3.88	11.89
Quercetin	474.32	130.12
Cinnamic acid	65.48	4.30
Kaempferol	0.00	62.87
Hesperetin	164.48	110.22

3.3. The tangerine emulsion's ξ -potential, polydispersity index (PDI), and particle size distribution

Emulsion factors like z-average, ξ -potential, and polydispersity index (PDI) are all closely related to encapsulation skillfulness, according to Baranauskaite, Ockun [25]. The tangerine emulsion's particle size, ξ -potential, and PDI were 249.3 nm, -24 mV, and 0.145, respectively, based on the results shown in Table 3. Z-averages, PDI, and ξ -potential are important markers of an emulsion's physical stability over time. The ξ -potential plays a crucial role in maintaining the physical stability of emulsions. Regardless of their sign, higher ξ -potential values indicate that the emulsion is more stable. The PDI number, which represents the width of the droplet size distribution, indicates the homogeneity of the resulting droplets in emulsions. PDI values range from 0 to 1, and the closer the number gets to zero, the more uniform the distribution [25].

Table 3: The tangerine emulsion's ξ -potential, polydispersity index (PDI), and particle size distribution

Particle size (nm)	ξ -Potential (mV)	PDI
249.3 \pm 49.07	-24 \pm 3.91	0.145 \pm 0.00

3.4. Transmission and Scanning electron microscopy (TEM&SEM)

TEM analysis of the TP emulsion's morphological structure revealed a smooth, rounded, spherical shape (Figure 3 A). The droplet dimensions seen by TEM and those ascertained by dynamic light scattering were in agreement. Additionally, the size distribution of the droplets was found to be varied, indicating the presence of certain aggregates. Overall, our results confirm that the microstructures of the emulsions varied according to the composition of the droplet-encircling layers. The SEM microphotographs of the powders made at the chosen ideal inlet air temperature of 160 °C are displayed in Figure 3 (B). As is common with spray-dried powders, the particles were spherical in shape and varied in size. Although some particles had rough surfaces, the majority of them had smooth surfaces. Particle shrinking during drying and cooling is what causes the depressions. Particle shrinkage as a result of the extreme moisture loss and subsequent cooling is typically ascribed to spray-drying. Similar morphology was noted in numerous earlier investigations [26] [27] all of them produced by spray drying using gum Arabic and maltodextrin as wall material. Addition of maltodextrin facilitated the drying process and improved the properties of the powder [7].

There were no fractures, cracks, or porosity on the continuous exterior particle surfaces. According to Abdel-Razek [28] these characteristics are critical for ensuring active substances' considerable protection and retention.

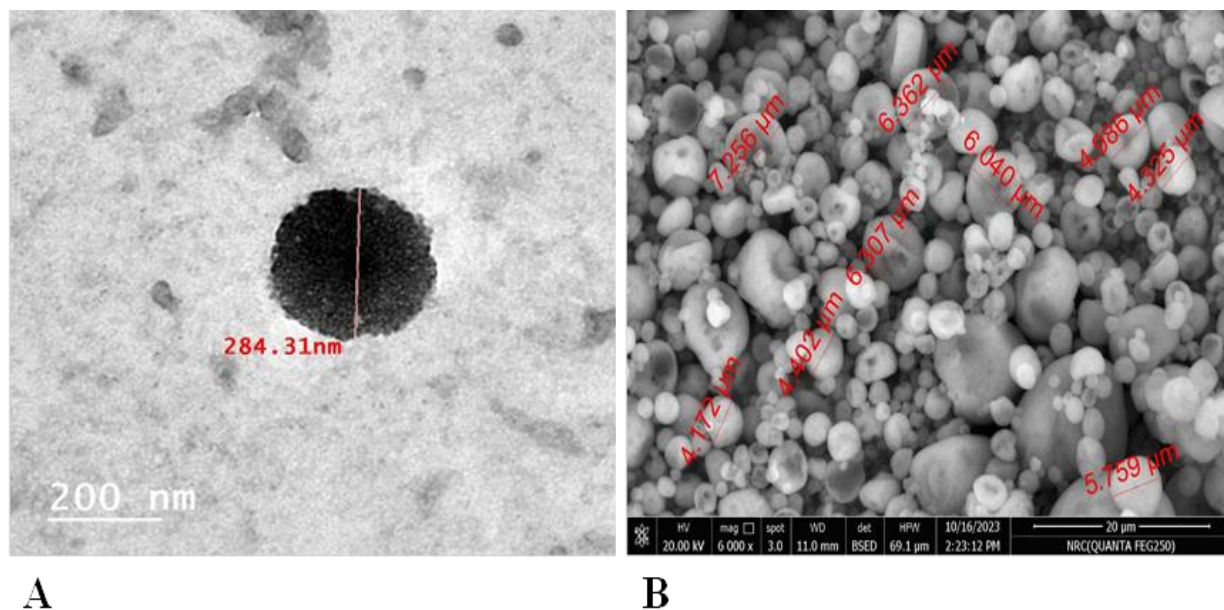


Fig. 3. TEM micrograph (A) and SEM image (B) of the encapsulated tangerine beverage

3.5. Encapsulation efficacy (EE)

By improving their solubility and preserving their functional stability throughout processing and storage, encapsulation successfully shields essential oils and bioactive chemicals from chemical reactions and adverse interactions with other food ingredients. Encapsulation is also believed to assist control the release of substances that are encapsulated, as well as their bioavailability and bioaccessibility. Therefore, encapsulation was employed in this work to create microcapsules of the bioactive chemicals found in tangerine peel. Tangerine peel bioactive components had an encapsulation efficiency percentage (EE%) of 88.54 ± 1.35 . The amount of free TP oil in microcapsules was calculated to determine encapsulation efficiency (EE). A higher encapsulation efficiency is associated with a lower free oil content.

3.6. Total phenolic, Total flavonoid content and antioxidant activity of TP oil, extracts, emulsion, and microcapsules'

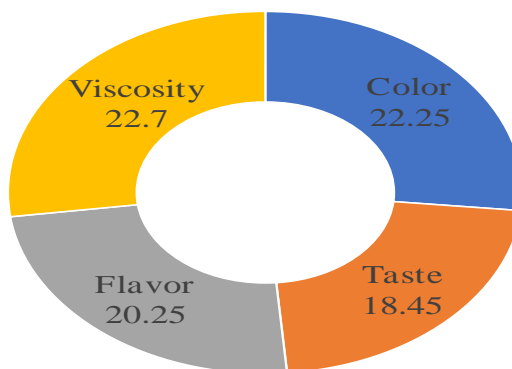
Tangerine oil, aqueous and ethanol extracts, emulsion, and microcapsules were shown in Table 4 along with their total phenolic content (TPC), total flavonoids (TFC), and antioxidant activity. The findings of this investigation shed important light on the antioxidant characteristics of several PT extracts (oil, aqueous, and ethanol) as well as the part that flavonoid and phenolic chemicals play in these activities. The phenol and flavonoid levels of the water extract, which is the primary extract used to prepare PT drinks, are responsible for the protective effect against free radical oxidation that was found [29]. It is crucial to remember that the use of water as a solvent for the extraction of bioactive chemicals was the main focus of this investigation. Because water extraction is a polar solvent, some chemicals may have been selectively removed while others were ignored [30]. Different solvents with varying polarities, such as ethanol and methanol may yield different results [31, 32]. According to our findings, ethanol extract has a higher phenolic and flavonoid content than both oil and aqueous extract. As a result, it was essential to utilize their synergistic activity in order to produce an emulsion with various tangerine peel extracts that had the highest antioxidant activity. Phenolic acids and flavonoids' redox characteristics and capacity to squelch singlet oxygen are linked to their antioxidant qualities [33]. Antioxidant activity is characterized as increased DPPH and ABTS radical scavenging activity [34]. DPPH and ABTS radical inhibition percentages were found to be higher in tangerine microcapsules than in the emulsion and other separate extracts. The higher concentration of phenolic chemicals, which can release free electrons or hydrogen atoms, may be the cause of this. Additionally, it was found by Jayari, Donsi [35], that the ability of the encapsulating structure to retain essential oils and their dispersion in water as an emulsion both increase their antioxidant impact. Because of their high polyphenol content, which slows down the degradation of physiologically active substances during storage and consumer presentation, products rich in phenolics and their derivatives have a longer shelf life [36].

Table 4: Total phenolic content (TPC), total flavonoids (TFC) and the antioxidant activity (DPPH, ABTS and reducing power) of the tangerine oil, aqueous and ethanol extracts, emulsion, and microcapsules.

Tangerine peel extract	TPC ($\mu\text{gGAE/m}$)	TFC ($\mu\text{gQE/mL}$)	ABTS (TE)/g	DPPH (%) (IC50)	Reducing power ($\mu\text{g ASC/mL}$)
Tangerine oil	96.41 ^b \pm 4.42	10.34 ^a \pm 0.38	63.35 ^b \pm 5.60	26.68 ^a \pm 1.63	65.02 ^b \pm 2.36
Ethanollic extract	48.27 ^a \pm 2.32	19.43 ^a \pm 1.25	41.38 ^a \pm 1.53	32.96 ^b \pm 2.96	48.57 ^a \pm 2.15
Aqueous extract	146.29 ^c \pm 3.50	195.68 ^c \pm 13.50	98.99 ^d \pm 0.61	44.88 ^c \pm 1.36	145.67 ^d \pm 8.89
Emulsion	313.98 ^d \pm 4.73	133.34 ^b \pm 0.63	79.72 ^c \pm 3.85	65.73 ^d \pm 1.09	108.26 ^c \pm 7.57
Capsules	449.18 ^e \pm 13.00	231.23 ^d \pm 3.36	96.69 ^d \pm 1.00	79.10 ^e \pm 4.87	191.22 ^e \pm 9.63

3.7. Sensory attributes of the functional tangerine beverages

A critical step in assessing the functional property improvement study is sensory evaluation since products meant for consumers must first appeal to their senses [26]. In addition to serving as a gauge for food quality, flavour has a significant impact on consumer happiness and food products' market value. After 5% recovery with cold water, the sensory evaluation of tangerine peel microcapsules as an instant drink was conducted. The sensory qualities of the tangerine beverage, including colour, flavour, taste, and viscosity, were evaluated. Figure 4 displays the findings. Each attribute was given a score on a hedonic scale from 1 to 25. The beverage sample was superior in terms of taste, colour, flavour, and viscosity.

**Fig. 4.** Sensory attributes of the tangerine beverages.

4. Conclusion

In the present study, the utilization of tangerine peels, as agricultural wastes, was maximized by extracting the bioactive compounds from these peels and converting these extracts into instant beverages. The functional beverages produced by spray drying technique showed antioxidant activity and recorded good values for the sensory attributes. The current study suggests the possibility of including the prepared functional beverages as healthy alternatives to instant drinks available in the market, which contain artificial flavoring substances that have health risks, in addition to their high sugar content.

Acknowledgements

The authors express their gratitude to the Academy of Scientific Research and Technology (ASRT) in Egypt for their valuable support in facilitating this work.

References

1. Raţu, R.N., et al., *Application of Agri-Food By-Products in the Food Industry*. Agriculture, 2023. **13**(8): p. 1559.
2. Lucarini, M., et al., *Fruit wastes as a valuable source of value-added compounds: A collaborative perspective*. Molecules, 2021. **26**(21): p. 6338.
3. Sicari, V. and M. Poiana, *Recovery of bergamot seed oil by supercritical carbon dioxide extraction and comparison with traditional solvent extraction*. Journal of Food Process Engineering, 2017. **40**(1): p. e12341.
4. Frascareli, E., et al., *Effect of process conditions on the microencapsulation of coffee oil by spray drying*. Food and bioproducts processing, 2012. **90**(3): p. 413-424.
5. Yue, F., et al., *Effects of ageing time on the properties of polysaccharide in tangerine peel and its bacterial community*. Food Chemistry, 2023. **417**: p. 135812.
6. Badsha, M., et al., *Quality evaluation of commercially available instant mango drinks powder in local market of Bangladesh*. International Journal of Agricultural Research, Innovation and Technology (IJARIT), 2020. **10**(2): p. 54-58.
7. Samborska, K., et al., *Powdered plant beverages obtained by spray-drying without carrier addition-physicochemical and chemometric studies*. Scientific Reports, 2024. **14**(1): p. 4488.
8. Xiong, K. and Y. Chen, *Supercritical carbon dioxide extraction of essential oil from tangerine peel: Experimental optimization and kinetics modelling*. Chemical Engineering Research and Design, 2020. **164**: p. 412-423.
9. Idowu Oyeleye, S., et al., *GC characterization and erectogenic enzyme inhibitory effect of essential oils from tangerine and lemon peels: A comparative study*. Flavour and Fragrance Journal, 2022. **37**(1): p. 33-42.
10. El-Bilawy, E.H., et al., *Antifungal, antiviral, and HPLC analysis of phenolic and flavonoid compounds of Amphiroa anceps extract*. Sustainability, 2022. **14**(19): p. 12253.
11. Mahdi, A.A., et al., *Nanoencapsulation of Mandarin essential oil: Fabrication, characterization, and storage stability*. Foods, 2021. **11**(1): p. 54.
12. Zahran, H.A., et al., *Determination of the optimum conditions for emulsification and encapsulation of echium oil by response surface methodology*. ACS omega, 2023. **8**(31): p. 28249-28257.
13. Dwivedy, A.K., et al., *Nanoencapsulated Illicium verum Hook. f. essential oil as an effective novel plant-based preservative against aflatoxin B1 production and free radical generation*. Food and Chemical Toxicology, 2018. **111**: p. 102-113.
14. Quettier-Deleu, C., et al., *Phenolic compounds and antioxidant activities of buckwheat (Fagopyrum esculentum Moench) hulls and flour*. Journal of ethnopharmacology, 2000. **72**(1-2): p. 35-42.
15. Chang, C.-C., et al., *Estimation of total flavonoid content in propolis by two complementary colorimetric methods*. Journal of food and drug analysis, 2002. **10**(3).
16. Re, R., et al., *Antioxidant activity applying an improved ABTS radical cation decolorization assay*. Free radical biology and medicine, 1999. **26**(9-10): p. 1231-1237.
17. Oyaizu, M., *Studies on products of browning reaction antioxidative activities of products of browning reaction prepared from glucosamine*. The Japanese journal of nutrition and dietetics, 1986. **44**(6): p. 307-315.
18. Hussein, A.M., et al., *Fortified vegetarian milk for prevention of metabolic syndrome in rats: impact on hepatic and vascular complications*. Heliyon, 2020. **6**(8).
19. Naef, R.A.V., A. , *Volatile Constituents in Extracts of Mandarin and Tangerine Peel*. Journal of Essential Oil Research., 2001. **13**:3: p. 154-157.
20. Chandharakool, S., et al., *Effects of tangerine essential oil on brain waves, moods, and sleep onset latency*. Molecules, 2020. **25**(20): p. 4865.
21. Minh Tu, N., et al., *Volatile constituents of Vietnamese pummelo, orange, tangerine and lime peel oils*. Flavour and fragrance journal, 2002. **17**(3): p. 169-174.
22. Musa, K.H., et al., *Antioxidant activity of pink-flesh guava (Psidium guajava L.): effect of extraction techniques and solvents*. Food Analytical Methods, 2011. **4**: p. 100-107.
23. Garcia-Salas, P., et al., *Phenolic-compound-extraction systems for fruit and vegetable samples*. Molecules, 2010. **15**(12): p. 8813-8826.
24. Spigno, G., L. Tramelli, and D.M. De Faveri, *Effects of extraction time, temperature and solvent on concentration and antioxidant activity of grape marc phenolics*. Journal of food engineering, 2007. **81**(1): p. 200-208.
25. Baranauskaite, J., et al., *Effect of the amount of polysorbate 80 and oregano essential oil on the emulsion stability and characterization properties of sodium alginate microcapsules*. Molecules, 2021. **26**(20): p. 6304.
26. Ramadan, M.M., et al., *Development of a functional cake with probiotics and micro-encapsulated essential oils: Evaluation of nutritional properties, liver protection, and immune boosting*. Heliyon, 2024. **10**(1).
27. El Haggag, E.F., et al., *Tomato-Free wonder sauce: A functional product with health-boosting properties*. Journal of Functional Foods, 2023. **109**: p. 105758.
28. Abdel-Razek, A., *Non-traditional Oils Encapsulation as Novel Food Additive Enhanced Yogurt Safety Against Aflatoxins*. Pakistan Journal of Biological Sciences: PJBS, 2019. **22**(2): p. 51-58.
29. Bouloumpasi, E., et al., *Assessment of antioxidant and antibacterial potential of phenolic extracts from post-distillation solid residues of oregano, rosemary, sage, lemon balm, and spearmint*. Processes, 2024. **12**(1): p. 140.

30. Vilková, M., J. Płotka-Wasyłka, and V. Andruch, *The role of water in deep eutectic solvent-base extraction*. Journal of Molecular Liquids, 2020. **304**: p. 112747.
31. Awang, M.A., et al., *Comparison of different solvents on the extraction of Melastoma malabathricum leaves using soxhlet extraction method*. Der Pharm Lett, 2017. **8**(4): p. 153-7.
32. Ghanem, K.Z., et al., *Enhancing the antioxidant properties of functional herbal beverages using Ultrasonic-Assisted extraction: Optimized formulation and synergistic combinations of taurine and vit. C*. Heliyon, 2024. **10**(15).
33. Simunkova, M., et al., *Antioxidant vs. prooxidant properties of the flavonoid, kaempferol, in the presence of Cu (II) ions: A ROS-scavenging activity, fenton reaction and DNA damage study*. International journal of molecular sciences, 2021. **22**(4): p. 1619.
34. Zhou, K. and L. Yu, *Effects of extraction solvent on wheat bran antioxidant activity estimation*. LWT-Food science and Technology, 2004. **37**(7): p. 717-721.
35. Jayari, A., et al., *Nanoencapsulation of thyme essential oils: Formulation, characterization, storage stability, and biological activity*. Foods, 2022. **11**(13): p. 1858.
36. Ahmad-Qasem, M.H., et al., *Influence of freezing and dehydration of olive leaves (var. Serrana) on extract composition and antioxidant potential*. Food Research International, 2013. **50**(1): p. 189-196.