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Encapsulation of carotenoids extracted from Navel orange and Clementine Tangerine peel extract as a by-product.

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Abstract

Carotenoids are pro-vitamin A, natural pigments and strong antioxidants. Encapsulation technology protects carotenoids against degradation and increases stability in food products. In this work, the carotenoid extract from Navel Orange peel (NOP) and Clementine Tangerine peel (CTP) was used. The study was designed for encapsulation in microstructures by three concentrations of carotenoid (3, 7 and 9 %) using the same coating agent (maltodextrin with Arabic gum15% w/v).The evaluation of physicochemical carotenoid stability of the encapsulated carotenoid extract. The result showed that the total carotenoids after microencapsulation for NOP and CTP, 3 and 7% were significantly higher than 9%. The antioxidant activity of CTP is higher than NOP when measured by $ABTS^{\bullet+}$ assays. The encapsulation efficiency (EE) of microcapsules content of 7% carotenoid was the highest efficiency for NOP and CTP extract. The size of the micro-particles was determined by transmission electron microscope (TEM) which showed it less than 1µm after encapsulation, ensuring good solubility in an aqueous solution. Shape microcapsules had an irregular shape and were amorphous with porous and slab forming a structure that was similar to each other in morphology under scanning electron microscopy (SEM). Assessment of β-carotene content in NOP and CTP encapsulated extract by HPLC the β-carotene of the CTP extract was twice the β-carotene of the NOP extract. Also, the assessment of moisture and water activity for NOP and CTP encapsulated ranged between 4.59–6.07%, 0.26–0.32, 5.72–6.81% and0.303–0.32 respectively. The purpose of the study was to extract natural carotenoids from Navel Oranges and Clementine Tangerine Peels and to study their functional properties and their possible use as additives with certain foods. It can be used in foods as pro-vitamin A, nature pigment, and antioxidants.

Keywords: Micro-capsulation; β-carotene; Encapsulation efficiency; Scanning electron microscopy; Arabic gum; Maltodextrin (MD)

1.Introduction

Citrus fruits are the world's most powerful fruit crop, producing more than 100 million tonnes of fruit every year **[1].** Egypt is the sixth most important supplier of oranges throughout, producing a variety of citrus fruits, especially oranges (70 % of citrus output) for instance. Navel orange, whose production had rapidly expanded to 4.5 million tons, including about 1.6 million tons in exports **[2].** Even though citrus peels are not tasty, they do provide significant biological benefits

Tangerines come from the citrus fruit group and have loose skin that can be easily peeled **[3]**. It has some properties such as antibacterial, antioxidant, and anti-cancer. **[4]**

According to Juliana Tamayo, [**5**] there are numerous varieties of tangerines, including clementines, tangelos, and temples.

Orange and Tangerine peels are extremely pigment-rich and weight nearly half as much as the total fruit. Carotenoids and phenolic compound are usually responsible for the orange peel's color **[6].** Significant quantities of orange peel waste have been tracked in the technological circumstances when the juice is removed from the remainder of the fruit. The peels contain phenols, amino acids, essential oils, carotenoids, pectin, flavonoids, sugars, vitamins, and minerals, which make them protective against many diseases **[4]**.

Carotenoids, for example, are among the most commonly applied natural colorants, despite their low water solubility and durability **[7]**. Carotenoids are lipid molecules that are naturally occurring pigments that are typically made up of eight isoprenoid units. As a result, they can be dissolved in nonpolar organic solvents but not in water **[8].** Carotenoids' stability will be improved by encapsulating agents, which also create a barrier between the wall and core materials.

The Freeze drying technique yields the best quality for the finished product and keeps the

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bioactive because of the low temperature used. Because the coating materials provide a barrier of protection for the bioactive compounds, suitable coating agents are required during the encapsulation proc**ess [9].**

The food industry is very interested in applying these encapsulations in natural extracts because they produce color stability as colorants and alternatives to artificial colorants. Natural colorants are now increasingly used in a variety of meals, beverages, cosmetics, pet food, foods, and pharmaceutical industries have increased. This is because the safety of natural colorants is more credible than that of synthetic colors **[10]**

Although powerful antioxidants, carotenoids are very unstable and prone to damage when processed or stored. Using natural ingredients in food products helps increase the protection against bioactivity **[11 and 12]**.

This study aims to produce micro-capsuled carotenoid pigment from the citrus peel (Navel orange and Clementine tangerine) extract and to evaluate their characterizations and possible use as bioactive in bakery product foods.

2. MATERIALS AND METHODS

2.1. Materials

2.1.1. Plant materials

Citrus (Clementine tangerine and Navel orange) were collected from Egypt's Horticultural Research Institute Agriculture Research Center, Giza, (2021). Citrus fruits (Clementine tangerine and navel orange) were carefully peeled, cleaned under running water, and divided into edible pieces. Before the drying process, the peels were cut into small pieces. The fresh peel pieces were dried for 48 hours in an air oven (Shellab-Model 1350FX.-Made in the USA), Then crushed to a fine powder using a mechanical grinder (Moulinex TYPE R 43 Moodle DEPOSE – Made in France) and Each type of peels has been individually packed was packaged in plastic bags,and stored at 4±1°C until needed.

2.1.2. Chemicals

Acetone and petroleum ether were purchased from Piochem Chemicals Co., Egypt. Scorbic acid, methanolic potassium hydroxide solution, dihydrogen potassium phosphate solution, anhydrous sodium sulphate, acetonitrile, tetrahydrofuran, ammonium-acetate, sodium chloride, Tween 80, ethyl alcohol, methyl alcohol, hexane and petroleum ether were purchased from El-Nasr Pharmaceutical Chemicals Co., Egypt. Gum Arabic (GA), Maltodextrin (MD) were used Alhoda Co. in Cairo, Egypt. ABTS•+ [2, 2 - azinobis -3ethyl benzothiazoline - 6-sulfonic acid] and potassium persulfate ($K_2S_2O_8$) Purchased from Sigma Aldrich, USA, provided the carotene standards used for HPLC identification.

2.2. Methods

2.2.1. *Extraction of carotenoid of Navel orange and Clementine tangerine peels powder*

Navel orange and Clementine tangerine peels powder (500grams) were mixed and macerated in 1 L of acetone and added 0.1% ascorbic acid in a blender. After filtering the extract, the leftover material underwent two further extractions using the same solvent. At 40 ±1°C, the crude-colored extract was quickly highly concentrated using a rotary. Saponification is carried out to remove oil and chlorophylls extracted with the natural yellow pigments. The concentrated extract was washed twice with a 200 mL portion of (100 g potassium hydroxide solved in 750 mL of methanol+ 250 mL water) in a separator funnel. A suitable amount of hexane was added and the upper layer was washed several times with a 10% dihydrogen potassium phosphate solution. The extract was dried over anhydrous sodium sulphate and kept under cover in dim bottles refrigerated at 5 ± 2 °C until used, according to Megahed **[13].**

2.2.2. Analysis β-Carotene

For extraction, the Gentili and Caretti **[14]** method was used with some modifications. Acetone (1 mL) was added to extract samples (50 mg). The extract was centrifuged by Labnet Internatonal Edison, NJ USA at 10,000 RCF for 10 minutes at 4ºC. After being vortexed for 30 seconds and incubated in the dark for 30 minutes at 4º C with vortexing every 5 minutes. The supernatant was separated with a glass syringe, and the extract was placed into an amber 1.8 mL glass vial with a low-volume vial insert. The glass vial was then refrigerated at -20 °C until analysis.

2.2.3. Separation and identification of β - Carotene using HPLC

The β-carotene content was determined using Hart and Scott's **[15]** methods, with some modifications. Five grams of sample were saponified with 20ml of potassium hydroxide (25%) and 0.5 gm. BHT was added, then stirred for about 3 hr. at room temperature under nitrogen. β carotene was extracted from the saponified mixture by careful shaking with 20 ml of hexane and petroleum ether (1:1) and 20 ml of sodium chloride solution by using a separated funnel. The lower phase was removed to another separating funnel and extracted two times with 20 ml of hexane and petroleum ether (1:1). The petroleum ether and hexane phases were thoroughly cleaned with distilled water to remove all residues of the base. Rotary evaporation and a water bath at 30°C were used to concentrate the extract, and quantitatively transferred into an amber vial with the mobile phase. HPLC analysis was performed using Chiosa *et al.*

[16] methods. The investigations were performed using an HPLC (Agilent) model-LC 1100 series equipped with a degasser, an auto sampler automated injector, a high-pressure pump, and a UV/Visible detector at different wavelengths. Acetonitrile, methanol, 1,2-dichloroethane (60:35:5, v/v/v), 0.1% BHT, 0.1% triethylamine, and 0.05 M ammonium acetate made up the mobile phase (in methanol). The compound was divided on an Alltima C18 5 m guard column with a C18 column (2504.6 mm id., Vydac) (7.54.6 mm id., Alltech). The column was maintained at room temperature (about 22°C) and the flow rate was 1 ml min-1. The column was held at room temperature (about 22°C) and the flow rate was 1 ml min^{-1}. 450 nanometers was chosen as the wavelength. The peak regions were measured using Millennium Software V. 2.0. (waters).

2.2.4. Microencapsulation techniques

2.2.4.1. Emulsion formulation

As the first stage in microencapsulation, a carotenoid emulsion was created using freeze-drying. To produce the emulsion, 15 g of gum Arabic (AG) was combined with 100 mL of deionized water. This solution was allowed overnight at 4ºC to thoroughly hydrate. Then, in the same gum solution, 15 g of maltodextrin (MD) was dissolved to obtain a 1:1 GA: MD (w/w) ratio. Tween 80 (1.0% w/v) was appropriately mixed into the same ratio solution of Arabic gum and maltodextrin before three different concentrations of carotenoids were added to generate an emulsion that was (3, 7, and 9%) w/v. An IKA Hielscher GmbH UP200S ultrasound homogenizer (IKA Hielscher GmbH, Berlin, Germany) fitted with a 14 mm diameter impulse generator was used to homogenize the entire mixture for 20 minutes at full sonicator power (200.0 Watt, 24 kHz) after being magnetically stirred for 5 minutes to create a coarse pre-emulsion. The emulsion vessel was submerged in a cold water bath during the ultrasonic homogenization procedure to prevent the emulsion temperature from rising over ambient, with minor changes to the method reported by Edris *et al.* **[17].** Finally, six formulas were developed in this study, which are listed in Table (1):

Table 1. The formulas which developed before microcapsule treatments.

	Navel orange peel extract		Clementine tangerine peel extract
T_A	3%Carotenoid + MD + AG	T_D	3% Carotenoid + MD + AG
T_B		T_E	
T_C	7%Carotenoid + MD + AG	T_F	7%Carotenoid + MD + AG
	9% Carotenoid + MD + AG		9%Carotenoid + MD + AG

MD: 15% maltodextrin and AG: 15% Arabic gum.

2.2.4.2. Freeze dryer technique

Freeze drying was carried out at the National Research Centre. Labconco freeze dryer, Console, 12L, -50ºC, Stoppering Tray Dryer, Freezon, 240V, Catalog No. 7754030, Serial No. 100931482 D, U.S.A. 12L, Condition:-50C 0.1mbar Time 48h. **[18]**

2.2.5.1. Encapsulation Efficiency (EE)

To calculate the amount of EE in the carotenoids, divide the amount of carotenoids in the microparticle by the amount of carotenoids in the carotenoid extract before freeze-drying.

$\% EE = ({}^{Ca}/_{Cb}) \times 100$

Where Ca represents the concentration of carotenoids in the micro-particle, and Cb represents the concentration of carotenoids before freeze drying. By adding 2 mL of a methanol/water solution (50:50 v/v) to the microparticles (0.4 g), the total carotenoid content was extracted. The vortex dispersion was homogenized for 1 minute and then subjected to a 20-minute ultrasonic bath (Cleaner Kondentech, So Paulo, Brazil). It was centrifuged for 15 minutes at 7200 rpm. The supernatant was filtered through 0.45µm 19 pore diameter membranes for carotenoids concentration determination at a 470 nm UV-VIS (Shimadzu® UV 1800, Kyoto, Japan) **[19].**

2.2.5.2. Assessment of total carotenoids

The UV/Vis spectrophotometric technique was used to remove the microparticles and determine the total carotenoid content (Shimadzu UV 1800, Kyoto, Japan). Weighed 3 g of the sample and it was then dispersed in 10 mL of hexane. The measurement was made using the carotene-specific coefficient (Eo = 2592) at a wavelength of 470 nm. The extract was also subjected to analysis before drying. The findings, which were carried out in triplicate, were presented as mg carotenoids/100 g **[20].** An equation was used to calculate the outcomes:

$$C = (\text{Absorbance} \times \text{solution volume} \times 10^6) / (100 \times 2592 \times \text{sample weight}).$$

2.2.5.3. Antioxidant activity of Navel Orange, Clementine peel extracts carotenoid extracts and microcapsule by ABTS Assay

The antioxidant activity of Navel Orange, Clementine peel extract carotenoid extracts and microcapsule described by Re et al. **[21]** methods with some modifications. We used 7 mM ABTS• + ([2, 2 - azinobis -3 ethylbenzothiazoline-6-sulfonic acid]; was mixed with 2.45 mM $K_2S_2O_8$ and leaves at room temperature for 16 hours to produce the ABTS•+ radical cations. After that, the solution was diluted with ethanol to give an initial absorbance of 0.71 ± 0.03 at 734 nm. The absorbance was measured after 6 minutes after combining 20µL of carotenoid

and microcapsule solutions with 980 µL of diluted $ABTS^{\bullet+}$ Calculated the percentage of inhibition.

$$\text{Inhibition (\%)} = \frac{\text{Initial absorbance} - \text{Final absorbance}}{\text{Initial absorbance}} \times 100\%$$

2.2.6. Characteristics and properties of the microcapsules

2.2.6.1. Determination of moisture content (M)

Determination moisture according to [**22]** 2g of a sample was dried at 130°C in an oven for 3 hours then the sample was reweighed. The calculated difference between the two weights corresponded to the amount of moisture content which was expressed as a percentage of oven dry sample.

2.2.6.2. Determination of water Activity (a_w)

According to Vuong *et al*. [**23**] water activity was measured by using (Rotronic Hygro Labe water activity (aw) EA10-SCs (Switzerland).

2.2.6.3. Determination of water solubility

To dissolve 200 mg of each powder, 10 mL of distilled water was used. Then centrifuged the mixture at 760 g for 15 minutes (HERMLE Labortechnik, Germany), 9 mL of the suspension was then taken out, and the remaining material was dried at 105 °C for 24 hours**[24].** We used Eq. (1) to determine the solubility in water. The determination of solubility (%) was done in triplicate.

$$\text{Solubility (\%)} = \frac{\text{Final weight} \times 10/9}{\text{Initial weight}} \quad (1)$$

2.2.6.4. Percentage of carotenoid pigment retention

The (% R) was used to determine the percentage of carotenoid pigment retention reported by Wang *et al***. [25]**

$$\%\ R = Ca/Cb \times 100$$

Where Ca represents the amount of carotenoid in the sample after capsulation and Cb represents the amount of carotenoid in the sample before freeze drying (emulsion).

2.2.6.5. Assessment Differential Scanning Calorimetry (DSC)

Utilizing Differential Scanning Calorimetry (DSC) (Shimadzu®, Kyoto, Japan), it was assess the thermal stability of carotenoid extract forms that were both non-encapsulated and micro-encapsulated. In an aluminum crucible, 3 mg of the materials were added. The samples were examined in a 50 mL/min nitrogen gas environment. Da Costa *et al***. [26]** state that a dynamic scan was performed over the range of 0 to 300^o C at a heating rate of 10^o C per minute by peak area integrating DSC profiles, evaporation enthalpies were calculated.

2.2.7. Morphology and size distribution

2.2.7.1. Measuring the particles size of microcapsules by Scanning Electron Microscopy (SEM)

The SEM was used to evaluate the exterior topographies of the freeze-dried, encapsulated Navel orange and Clementine Tangerine) extracts (SEM, Quanta FEG 250 with FEI Company's Netherlands-based field emission gun). The samples were first cast-dried on an aluminum pan, then cut to the proper size and attached to the conductive carbon tapes. After that, using a sputter coater to apply a thin (20 nm) conductive gold and platinum coating.

2.2.7.2. Transmission Electron Microscope (TEM) analysis of micro-particles

High-resolution (TEM) was used to observe the size of the micro-particles in the freeze-dried powder. 200 µl of diluted samples were put on a film covered with a 200-mesh copper piece of work for 10 minutes. The extra fluid was then filtered with paper. The grid was colored and allowed to cure for three minutes after one drop of 3% phosphotungstic acid was applied to it. The observation of the samples at 160 kV allowed for the TEM microscope **[27].**

2.2.7.3. Color measurements

The Color parameters (*L**, *b**, *a**, h^o and *c**) of samples microencapsulated Navel orange and Clementine Tangerine peel extract) were measured using a Chroma meter at three separate locations on the sample (Minolta CR 400, Minolta Camera, Co., Osaka, Japan). The Abonyi *et al*. [**28]** formula was used to calculate the *L** value (color lightness from 0 (black) to 100 (white), *a** value (degree of redness (0-60) or greenness (0-60), *b** value (degree of yellowness (0 - 60) or blueness (0 - 60), hue angle (*h**) which distinguishes between vivid and dull colors. While h^o is a measure of color hue angle *(h^o)* = 0, red; h^o = 90, yellow; h^o = 180, green; h^o = 270, blue), high C* values indicate brighter, more vivid colors.

2.2.8. Statistical Analysis

Duncan's multiple range tests was used to evaluate if a one-way analysis of variance (ANOVA) with multiple ranges significant difference ($p < 0.05$) had been done in SPSS version 26.0. (IBM corp., Armonk, NY). The data is reported using the mean value and standard deviation (±SD) for three replicates.

3. Results

Carotenoids are nutrients that range in color from yellow to red and are powerful antioxidants, but they are highly unstable and insoluble in water. The Navel orange and Clementine tangerine peel are a high source of carotenoids and antioxidants **[4].** Table (2) showed the total carotenoids and antioxidant values for citrus (Navel Orange and Clementine Tangerine)

peel extract and microencapsulated. Before microencapsulation, the total carotenoid content of the Navel orange and Clementine Tangerine peel extracts were the highest significant, at 58.94± 0.63 and 87.51± 0.15 mg/100g, respectively. On the other hand, decreased carotenoids value after microencapsulation are 52.70 ± 0.22, 52.76 ± 0.23, 40.26 ± 0.14, 79.53±0.12, 80.39±0.14 and 71.64 ± 0.05mg/100g for T_A, T_B, T_C, T_D, T_E and T_F respectively. The same table showed that the total carotenoids after microencapsulation for Navel Orange peel extract T_A was higher significant than T_B and T_C. Also, T_D was higher significant than T_E and T_F for Clementine Tangerine peel extract. Paini *et al.* [**29**] confirmed the finding by reporting that this disparity might be explained by a slight carotenoid loss during freeze-drying, also Abdelwahed *et al.* **[30]** revealed that the encapsulation procedure, the grinding following lyophilization, may have caused extra dehydration stressors. The antioxidant activity of citrus (Navel Orange and Clementine Tangerine) peel extract and microencapsulated carotenoid powders was determined by $ABTS^{\bullet+}$ assay. All microencapsulated products showed decreased antioxidants by $ABTS^{\bullet+}$ assay with decreased carotenoid. Carotenoids were lost as a result of carotenoid oxidation and the double bond breakdown and this was directly related to lower antioxidant activity. **[31]** According the antioxidant activity of T_A and T_B was nonsignificant and higher than T_C for navel orange peel extracts T_D and T_E were significant but also higher than T_F for Clementine Tangerine peel extract. Additionally, table (2) showed that due to their higher carotenoid content, Clementine Tangerine peels have stronger antioxidant activity than Navel Orange peels. This result was confirmed by Lindalva Maria *et al.* **[32].**

Table (2) Total carotenoid and antioxidants activity of encapsulation carotenoid pigment of citrus (Navel orange and Clementine tangerine) peel extract.

Treatments	Total carotenoid mg/100g	Antioxidant $ATBS^{\bullet+}$ %
A*	58.94 ± 0.63 [a]	53.44 ± 0.63[a]
T_A	52.70 ± 0.22 [b]	47.34 ± 0.36[b]
T_B	52.76 ± 0.23[b]	47.65 ± 1.63[b]
Tc	40.26 ± 0.14 [c]	37.38 ± 1.28[c]
B*	87.42 ± 0.15 a	79.88 ±1.07 [a]
T_D	79.53 ± 0.12 [b]	62.44 ±1.24 [c]
T_E	80.39 ± 0.14 [b]	66.49 ± 0.46[b]
T_F	71.64 ± 0.05 [c]	54.75 ± 0.11[d]

A*: Navel orange peel extract before microcapsule, B*: Clementine Tangerine peel extract before microcapsule. T_A: 15% MD + 15%AG +3% Carotenoid extract, T_B: 15%MD + 15%AG +7% Carotenoid extract, Tc: 15% MD +15%AG+9% Carotenoid extract for orange peel. T_D: 15%MD + 15%AG +3% Carotenoid extract, $_{15\%}$MD +15%AG +7% Carotenoid extract and T_F: 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel. The results were expressed as mean ± standard deviation (n = 3). Values followed by different letters in the same column are significantly different at $P < 0.05$, according to Duncan's test.

3.2. The Encapsulation efficiency (EE)

The ability of the carrier substance to encapsulate the target molecule is known as encapsulation efficiency. Table (3) showed that microencapsulated extracts encapsulation efficiency values of 89.74 ± 0.09 %, 90.21± 0.2 %, 81.84 ± 0.04 %, 90.85 ± 0.12%, 91.64 ± 0.11% and 90.88 ± 0.09 % for T_A, T_B, T_C, T_D, T_E and T_F respectively. Table (3) revealed that the Clementine Tangerine peel extract was higher than the Navel Orange peel extract. The Encapsulation Efficiency (EE%) of the carotenoids was still better than 80% in the microparticles. Our outcomes agreed with those of Alvarez-Henao *et al*. **[33],** who reported that encapsulation efficiencies ranged from 91.94 - 65.72 %. This final result is similar to Lindalva Maria et al**. [32],** who discovered EE of maltodextrin and Arabic gum was equally effective as carrier materials and a good film-forming agent. From Table (3) we noted that the Encapsulation Efficiency (EE %) of microcapsules increased until a 7% concentration of carotenoid. On the other hand, 9 % of carotenoids had lower efficiency (EE %). This result was due to the higher concentration of carotenoids which decreased the microcapsule loading of carotenoids used with the same carrier agent. Thamaket and Raviyan [**34**] found that adding carotenoids at high concentrations did not result in improved encapsulation with the same coating agent in the carotenoid extract. The overloading of encapsulated components caused a little reduction in the encapsulation efficiency, and this phenomenon was also seen in other polymeric matrices Liu and Park **[35].** We concluded that the concentration of 7% carotenoid with mixture (15% maltodextrin + 15% Arabic gum) was sufficient for coating carotenoid encapsulation efficiency.

Table (3) Encapsulation Efficiency % of microparticle

Encapsulation Efficiency (%)			
Navel orange peel extract treatments		**Clementine Tangerine peel extract treatments**	
T_A	**89.74 ± 0.09 [b]**	**T_D**	**90.85± 0.12 [b]**
T_B	**90.28± 0.21[a]**	**T_E**	**91.64 ± 0.1[a]**
Tc	**81.84 ± 0.04[c]**	**T_F**	**90.88 ± 0.10 [b]**

T_A: 15% MD + 15%AG +3% Carotenoid extract, T_B: 15%MD + 15%AG +7% Carotenoid extract, Tc: 15% MD +15%AG+9% Carotenoid extract for orange peel. T_D: 15%MD + 15%AG +3% Carotenoid extract, $_{15\%}$MD +15%AG +7% Carotenoid extract and T_F: 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel.. The results were expressed as mean ± standard deviation (n = 3). Values followed by different letters in the same column are significantly different at $P < 0.05$, according to Duncan's test.

3.3. The percentage of carotenoid pigment retention

Figure (1) showed that the comparison percentage of carotenoid pigment retention of encapsulation Navel Orange and Clementine Tangerine peel extract T_A, T_B, T_C, T_D, T_E, and T_F were 14.57, 11.58, 54.58,73.66, 57.3, and 87.36 % respectively. Luis *et al.* [**36]** found the encapsulation efficiency of β-carotene with a coating agent is retained between 60 and 99% at 100ºC. Guadarrama-Lezama, *et al.* [**37**]. We obtained carotenoid pigment retention rates ranging from 84 to 86 % when we used maltodextrin and an Arabic gum coating agent. Figure (1) showed that the retention of encapsulation (R %) of Clementine Tangerine peel extract was higher than the retention of encapsulation (R %) of orange peel extract. We noted that from Table (3) and Figure (1) the microencapsulation with higher encapsulation efficiency (EE) was a lower percentage of carotenoid retention. This result was confirmed by Luis et al. **[36]** who found that the decreased R% with increased EE% due to the coating agent before freeze drying.

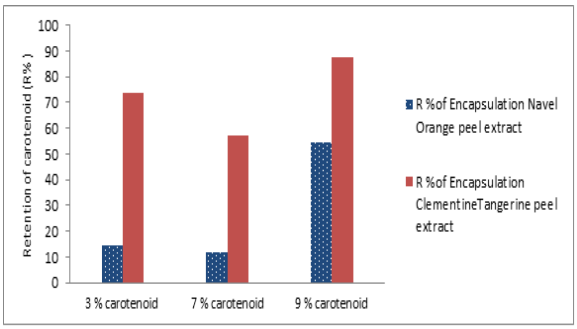

Figure (1) the comparison between carotenoid pigment retention of encapsulation Navel Orange and Clementine Tangerine peel extract(R %).

3.4. Thermal stability as measured by Differential Scanning Calorimetry (DSC)

DSC is defined as a thermal-analytical method that counts the physical and chemical alterations brought on by temperature variations in a substance. Figure (2) shows that the melting temperature of Navel Orange and Clementine Tangerine peels (without encapsulation) were 98.89 and 103ºC, respectively. The Navel Orange peel extract and Clementine Tangerines were unable to withstand high baking temperatures and lost bioactive components. The DSC of Navel Orange and Clementine Tangerine peels extract micro-encapsulated shown in Figure (2) the melting point reaches 215.75ºC for T_A, the melting point is 217.76ºC for T_B, melting point of 220.10ºC for T_C, melting point of 217.45ºC for T_D, melting point 217.35ºc for T_E and the melting point 217.13ºc for T_F. The results showed that the encapsulation increased melting point at the high-temperature process, due to the heat resilience of carotenoids being improved. The micro-encapsulated carotenoid pigment in peel extract from Navel Oranges and Clementines Tangerine increases the potential for use in the Food industry that is exposed to high temperatures while maintaining its antioxidant function. This result was consistent with that of Giles *et al.* **[38]** and suggests that it will be used as an antioxidant in cakes if heated to a high temperature.

3.5. Transmission Electron Microscope (TEM)

Figures (3) and (4) demonstrate the particle sizes of encapsulated and non-encapsulated Navel Orange and Clementine Tangerine peels that were re-dispersed in distilled water, respectively. The diameters of all treatment particles were 0.85µm before micro encapsulation carotenoid of Naval Orange peels and changed to 0.24, 0.03, and 0.08 µm after microencapsulation for different concentrations of carotenoid extracts T_A, T_B, and T_C as shown in Figure (3) (a - b - c and d) respectively.

While the particle size of the encapsulated carotenoid pigment Clementine Tangerine peels changed from 0.67 µm before micro capsulation to 0.02,0.03 and 0.09 µm after microencapsulation for T_D, T_E, and T_F Figure (4) (f-g-h and i) respectively.

After encapsulation, we noticed that all treatment particle sizes were reduced. Khaled *et al.* **[39]** discovered that some of the microparticles obtained by dispersion were collected following homogenization. The samples were kept to a thickness of less than 1µ ensuring good solubility and use in aqueous solutions.

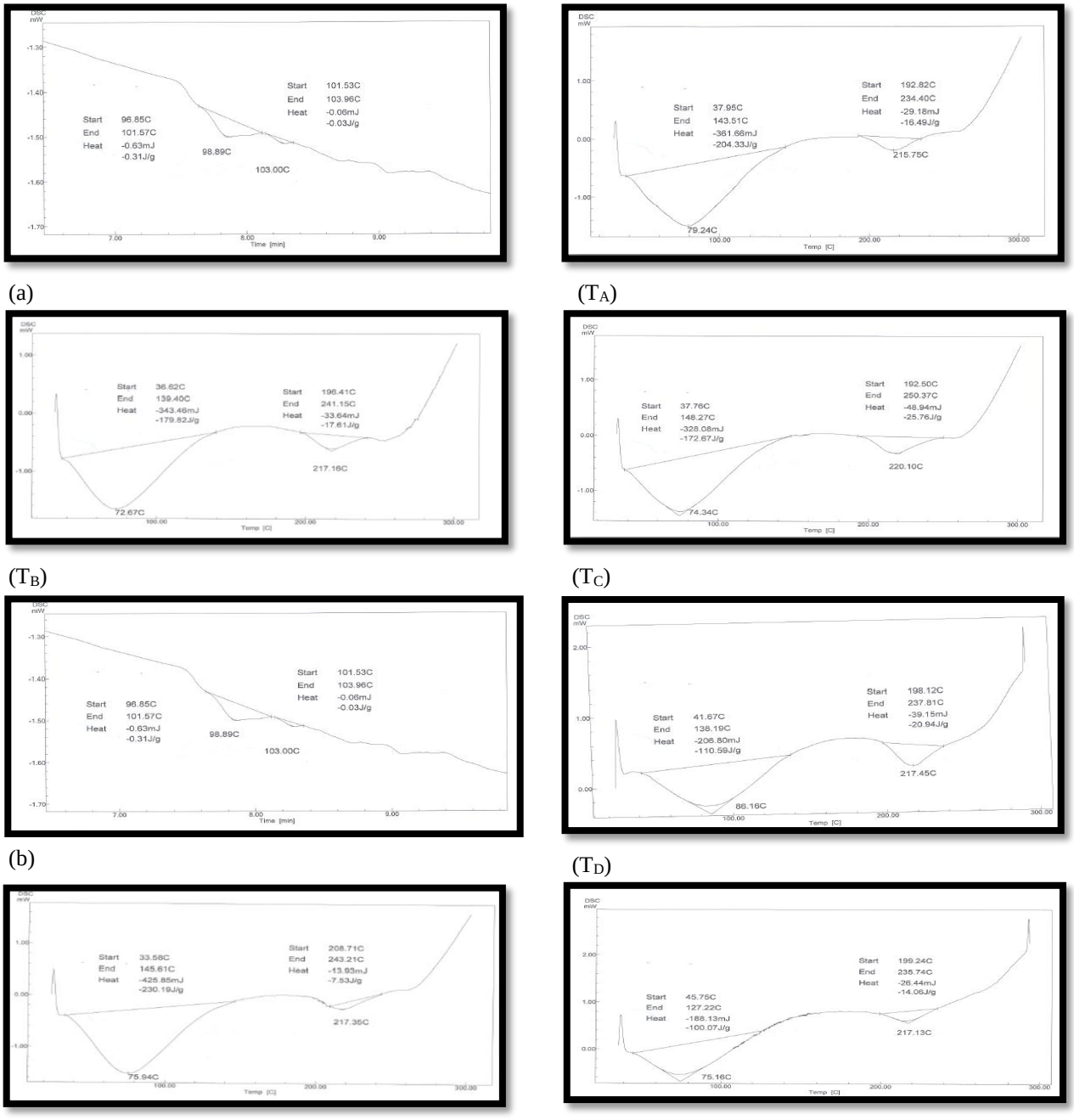

Figure (2) a: DSC thermogram of Carotenoids of naval orange peel extract before encapsulation, b: Carotenoids of clementine tangerine peel extract before encapsulation T_A: 15% MD + 15%AG +3% Carotenoid extract, T_B: 15%MD + 15%AG +7% Carotenoid extract, Tc: 15% MD +15%AG+9% Carotenoid extract for orange peel, T_D: 15%MD + 15%AG +3% Carotenoid extract. $T_{E:}$ 15%MD +15%AG +7% Carotenoid extract and $T_{F:}$ 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel.

3.6. Characteristics of microcapsules by (SEM)

Analyzing electron microscope image is a critical step in the evaluation of microcapsules. The encapsulated samples appeared to be very fine visually. SEM was used to evaluate the exterior topographies of the freeze-dried, encapsulated Navel orange and Clementine Tangerine) extracts. Figure (5) showed the similarities in shape between the powders made by freeze-drying peel extract from different citrus fruits (Navel Orange and Clementine Tangerine) peel to extract pigment with different carotenoid concentrations. The surface morphology of freeze-dried microcapsule powders was observed and related to the same qualities of the coating agents. The interactions of carotenoids with Arabic gum and maltodextrin in the feed solution may be the cause of the microcapsules produced after freeze-

drying. These SEM findings concurred with those of Naomi Mulyadi *et al.* [**40**] claimed that the form and slab-like structure of freeze-dried microcapsules were uneven. These morphological features came from the freeze-dried Navel Orange and Clementine Tangerine peel extract crushed by mortar. All of our products are amorphous with a dent and shrinkage-free structure, which is a desired attribute for such encapsulation tasks since it protects the entrapped molecules from heat and oxygen exposure**. [41]**

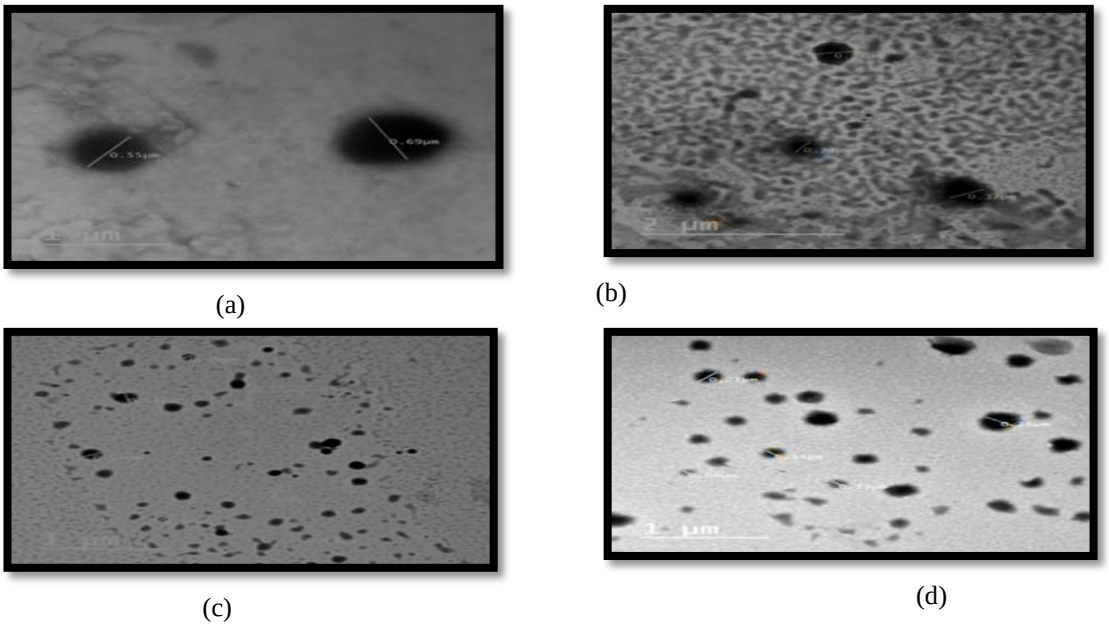

Figure (3) TEM of a: Carotenoids of naval orange peels extract before encapsulation, b: 15% MD + 15%AG +3% Carotenoid extract, c: 15%MD + 15%AG +7% Carotenoid extract and d: 15% MD +15%AG+9% Carotenoid extract for naval orange peel after encapsulation.

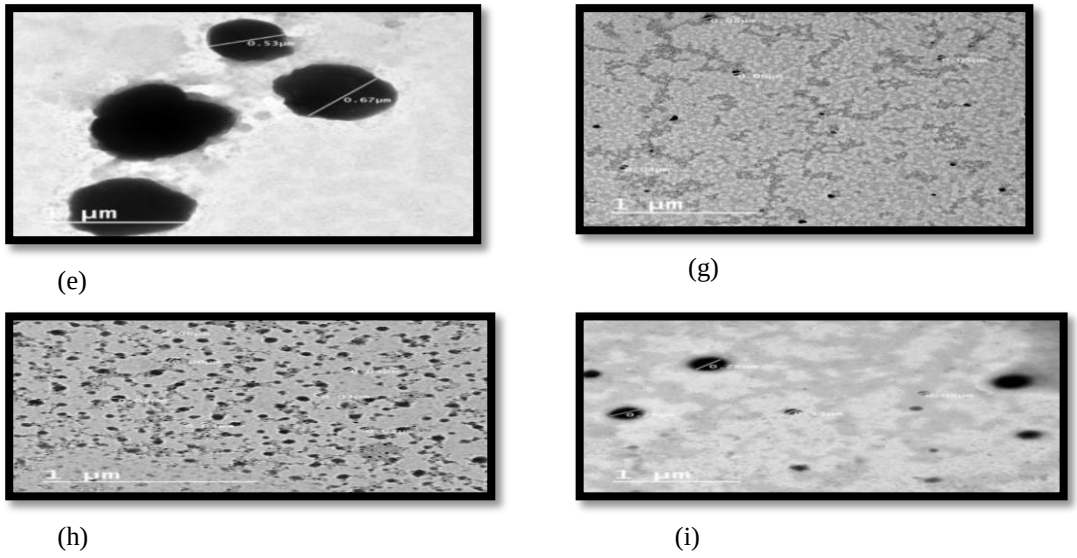

Figure (4) TEM: of e: Carotenoids of clementine tangerine peels extract before encapsulation, g: 15%MD + 15%AG +3% Carotenoid extract. h: 15%MD +15%AG +7% Carotenoid extract and i: 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel after encapsulation.

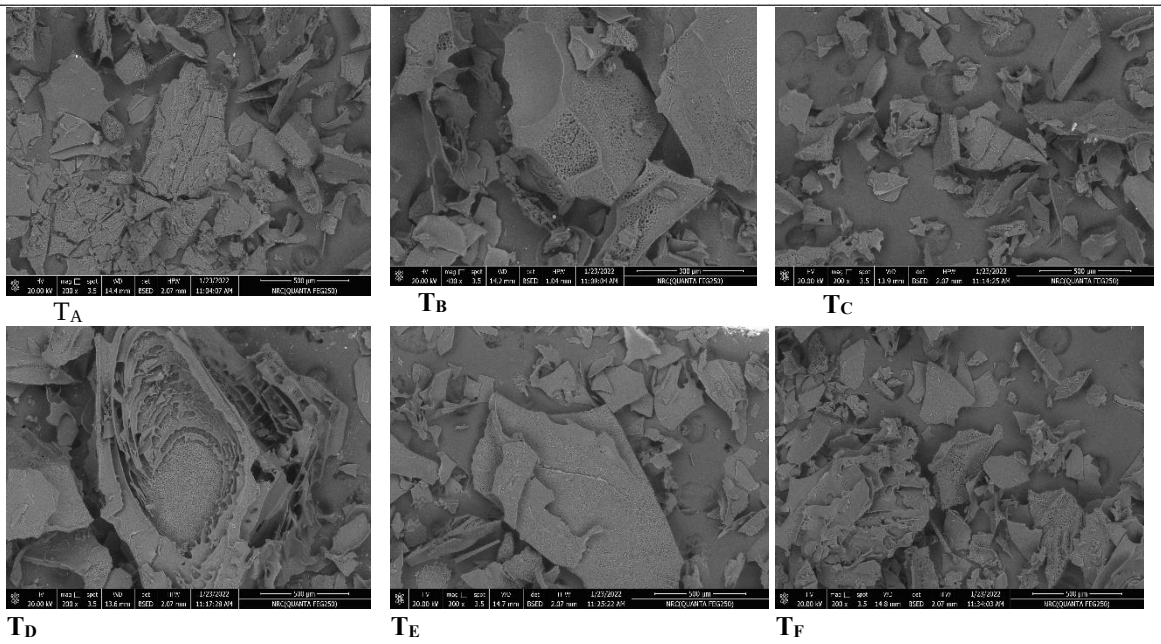

Figure (5) SEM microencapsulated of T_A: 15% MD + 15%AG +3% Carotenoid extract, T_B: 15%MD + 15%AG +7% Carotenoid extract, Tc: 15% MD +15%AG+9% Carotenoid extract for orange peel. T_D: 15%MD + 15%AG +3% Carotenoid extract. $_{15\%}$MD +15%AG +7% Carotenoid extract and T_F: 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel.

3.7. The water solubility of microencapsulated

The none capsulated carotenoids were water-insoluble. Figure (6) showed the comparison between the water solubility of microencapsulated Navel orange and Clementine tangerine peel extract. The water solubility of carotenoids encapsulated was higher than that without encapsulation. The solubility of microencapsulated Navel orange peel extract pigment T_A, T_B and T_C were 95.5, 83.5 and 97% respectively, and nearly similar solubility of microencapsulated Clementine Tangerine peel extract T_D, T_E, and T_F, are 91.5, 89.5 and 90.5% respectively. According to Lara Etzbach *et al. [***42**] investigations, the solubility of powders made from maltodextrin in cold water is high (>90%). This result reflects that indicate the hydrophilic characteristic of maltodextrin contributes to its high water solubility.

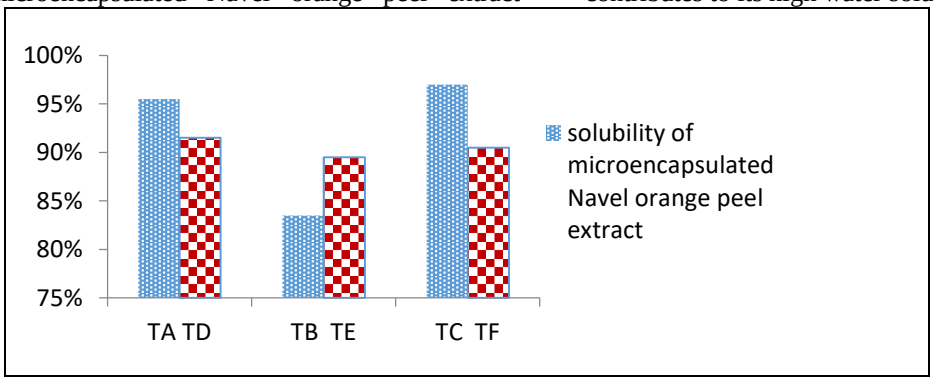

Figure (6) Comparison between water solubility of microencapsulated Navel orange and Clementine tangerine peel extract. *T_A: 15% MD + 15%AG +3% Carotenoid extract, T_B: 15%MD + 15%AG +7% Carotenoid extract, Tc: 15% MD +15%AG+9% Carotenoid extract for orange peel. T_D: 15%MD + 15%AG +3% Carotenoid extract. $_{15\%}$MD +15%AG +7% Carotenoid extract and T_F: 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel.*

3.8. Moisture and Water Content Activity of microencapsulated

The drying process is efficient in the food product's shelf life, and the longevity of food products was all impacted by the amount of moisture present and the water's activity. Table (4) displayed the relevance of the microencapsulated products' moisture content at various carotenoid concentrations. For Navel Orange for three treated T_A, T_B and T_C were 4.59 ± 0.91, 5.31 ± 0.05 and 6.07 ± 0.14respectavaly. Table (4) showed that the moisture content of the microencapsulated products with the different concentrations of carotenoid for Clementine Tangerine T_D, T_E, and T_F were 5.72 ± 0 .02, 6.25 ± 0.65 and 6.81 ± 0.16 % respectively. According to the findings in table (4), the water activity (a_w) value for microencapsulated Navel orange and Clementine Tangerine peel extract (T_A, T_B, T_C, T_D, T_E and T_F) were nonsignificant for T_A, T_B and T_C (0.26 ± 0.03, 0.26±0.00 and 0.32 ±

0.01respectively. According to Gamboa-Santos *et al.* **[43],** A water activity rating approaching 0.3 implies that the food will be stable against non-enzymatic browning, microbiological development, and enzymatic activity when stored properly. Our findings agreed with those of Avarez-Henao *et al*. [**44]**, who discovered microencapsulation (water activity ranging from 0.12 to 0.33 and moisture content ranging from 4.21% to 9.01%). Yamashita *et al*. [45] furthermore discovered that the moisture content ranged from 8.21% to 11.17% and that the water activity was between 0.27 and 0.35. **[44 and 46].**

Table (4) moisture and water activity of microencapsulated Navel orange and Clementine Tangerine peel extract.

Treatments	Moisture (%)	Water activity (a_w)
T_A	4.59 ± 0.09^c	0.26 ± 0.03^b
T_B	5.31 ± 0.05^b	0.26 ± 0.01^b
T_C	6.07 ± 0.14^a	0.32 ± 0.01^a
T_D	5.71 ± 0.02^b	0.32 ± 0.01^a
T_E	6.25 ± 0.65^b	0.26 ± 0.01^c
T_F	6.81 ± 0.19^a	0.30 ± 0.00^b

The results were expressed as mean ± standard deviation (n = 3). Values followed by different letters in the same column are significantly different at P < 0.05, according to Duncan's test. ***T_A: MD +AG +3% Carotenoid extract, T_B: MD +AG +7% Carotenoid extract, Tc: MD +AG+9% Carotenoid extract for orange peel. T_D: MD +AG +3% Carotenoid extract: MD +AG +7% Carotenoid extract and T_F: MD +AG+9% Carotenoid extract for Clementine Tangerine peel.***

3.9. Determination of β-carotenes (mg/100g) by High-pressure liquid Chromatograph (HLPC)

The β- carotene component in the samples was identified using HPLC. Table (5) showed that the β-Carotene content of Navel Orange and Clementine Tangerine peel extract before encapsulation was more than the β-carotene contained in microparticles after capsulation this difference could be attributed to a minor loss due to carotenoid degradation during the drying process. These results according to research by Lindalva *et al.* **[32]** β-Carotene Navel orange(A*) and Clementine Tangerine peel extract (B*) 8.79 and 19.47mg/100g, respectively. The β-Carotene content of the carotenoid difference concentration (T_A, T_B, T_C, T_D, T_E, and T_F) for Navel Orange and Clementine Tangerine peel extract after encapsulation with maltodextrin and Arabic gum was 7.77, 7.43, 2.22,13.83,16.97 and 10.72 mg/100g, respectively. From Table 5, it is clear that the β-carotene content of the Clementine Tangerine peel extract is twice the β-carotene of the Navel Orange peel extract.

Table (5) β- carotenes (mg/100g) by High-pressure liquid Chromatograph (HLPC)

Treatments	β- carotenoids mg/100g
A*	**8.79**
T_A	**7.77**
T_B	**7.43**
T_C	**2.22**
B*	**19.47**
T_D	**13.83**
T_E	**16.97**
T_F	**10.71**

A*: Navel orange peel extract before encapsulation, B*: Clementine Tangerine peel extract before encapsulation. ***TA: MD +AG +3% Carotenoid extract, TB: MD +AG +7% Carotenoid extract, Tc: MD +AG+9% Carotenoid extract for orange peel. TD: MD +AG +3% Carotenoid extract: MD +AG +7% Carotenoid extract and T_F: MD +AG+9% Carotenoid extract for Clementine Tangerine peel.***

Figure (7) Stander of β- carotene by HPLC.

3.10. Color microencapsulated carotenoid powders

The microencapsulated carotenoid powders might be seen in figure (9) To range from yellow to orange visually (Figure.9). The color of the microencapsulated in Table (6) changed as a result of the carotenoids. *L**, *a**, *b**, *C**, and *hue^o* are used to summarize color values. From a table (6) the values of *L** were no segniafant between all treatments. The values of *L∗* (luminosity) tended to decrease as carotenoid concentration increased. [**40**] the mean color space values *a** (+*a** red direction; -*a** green direction) for navel orange peel extract ranged from -4.94 ± 1.20 to -2.61 ± 0.29, while for clementine tangerine peel extract, they ranged from -1.90 ± 0.01 to 1.44 ± 0.20. According to Rachel *et al.* [**47**] negative values refer to light yellow. Although the value of b* presented nonsignificant differences for Navel Orange peel extract and significant for Clementine Tangerine peel extract. The color values,

however, shifted around *b**, which in positive levels represents the yellow color. According to [48], *C** values for encapsulated extracts indicate more colorful and vivid colors. Furthermore, the Chroma and hº on *a** and *b** **[49]** can explain this observation color. About h^o values were bright yellow (90º).

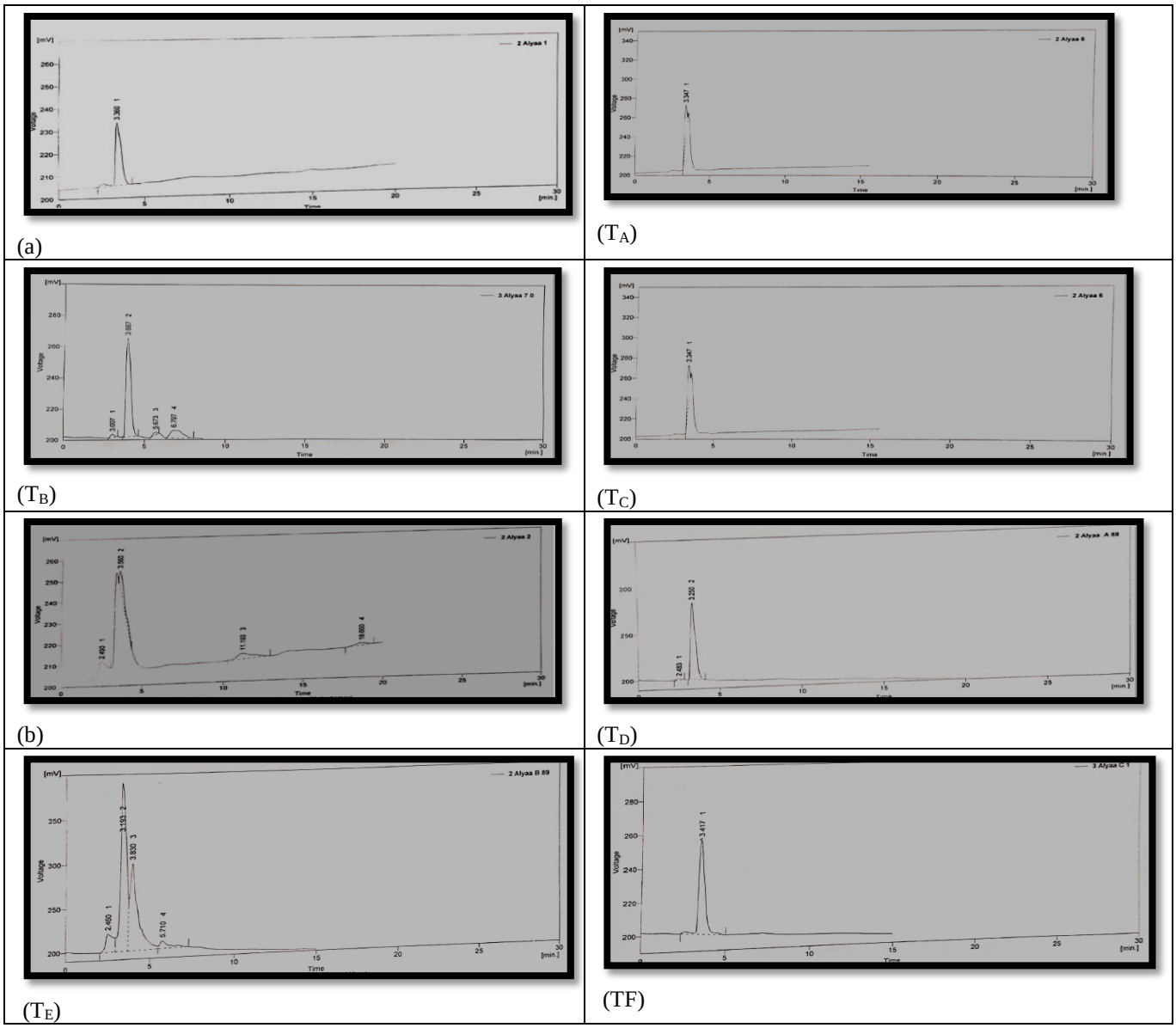

Figure (8) a:Identification of β- carotene (mg/100g) extracted from navel orange peel before encapsulation, b: Identification of β- carotene (mg/100g) extracted from clementine tangerine peel before encapsulation, microencapsulated T_A: 15% MD + 15%AG +3% Carotenoid extract, T_B: 15%MD + 15%AG +7% Carotenoid extract, Tc: 15% MD +15%AG+9% Carotenoid extract for orange peel after encapsulation, T_D: 15%MD + 15%AG +3% Carotenoid extract. 15%MD +15%AG +7% Carotenoid extract and T_F: 15% MD + 15%AG+9% Carotenoid extract for Clementine Tangerine peel after encapsulation by HPLC.

Table (6) Color microencapsulated carotenoid powders.

Treatments	*L**	*a**	*b**	*C**	h^o
T_A	78.08 ± 6.57 [a]	-4.94 ± 1.20 [b]	23.65 ± 1.61[a]	31.07 ± 1.25[b]	99.63± 1.79 [a]
T_B	72.86 ± 3.93 [a]	-3.10 ± 0.214 [a]	28.03 ± 3.85 [a]	38.59 ± 4.39 [a]	94.70 ± 0.17 [b]
T_C	71.76 ± 6.30 [a]	-2.61 ± 0.29 [b]	25.48 ± 2.35 [a]	32.45 ± 2.12[b]	99.93 ± 0.35 [a]
T_D	74.23 ± 2.13 [a]	-1.90 ± 0 .01[c]	29.99 ± 1.45 [a]	40.91 ± 1.7 [a]	92.80 ± 0.10 [a]
T_E	52.82 ± 2.63 [a]	- 0.72 ± 0.75 [b]	19.22 ± 1.24[c]	27.96 ± 1.17[b]	91.80 ± 1.30 [a]
T_F	63.94 ± 2.50 [a]	1.44 ± 0.20 [a]	26.73 ± 1.57 [b]	39.40 ± 2.04 [a]	88.01± 0.96 [b]

T_A: MD +AG +3% Carotenoid extract, T_B: MD +AG +7% Carotenoid extract, Tc: MD +AG+9% Carotenoid extract for orange peel. T_D: MD +AG +3% Carotenoid extract MD +AG +7% Carotenoid extract and T_F: MD +AG+9% Carotenoid extract for Clementine Tangerine peel.

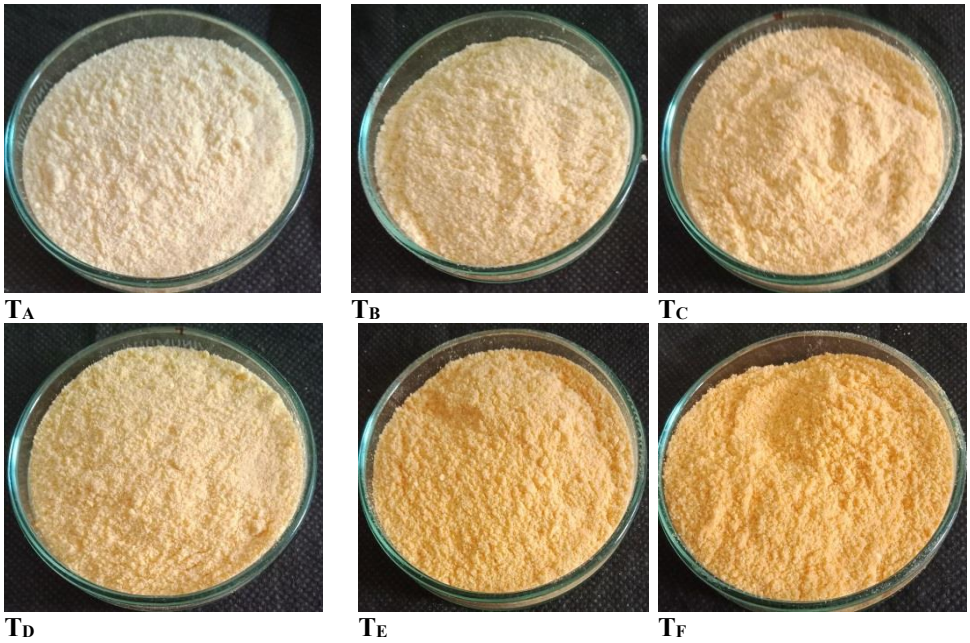

Figure (9). Microencapsulated carotenoids powder with *the (MD and AG) formulation using different addition of carotenoid concentrations* *T_A: MD +AG +3% Carotenoid extract, TB: MD +AG +7% Carotenoid extract, Tc: MD +AG+9% Carotenoid extract for orange peel. T_D: MD +AG +3% Carotenoid extract: MD +AG +7% Carotenoid extract and T_F: MD +AG+9% Carotenoid extract for Clementine Tangerine peel.*

4. Conclusion

This study focuses on the encapsulation of carotenoids sourced from Navel orange and Clementine tangerine peels by freeze-drying. The results showed that the encapsulation increased the melting point during the high temperature process due to the improved heat resilience of carotenoids. And resulting in micro particles, several studies can be applied in the near future. The micro-encapsulated carotenoid pigment in peel extract from Navel oranges and Clementine tangerine increases its potential for use in the food industry that is exposed to high temperatures while maintaining its antioxidant function. Microencapsulation is a promising technology we suggest using in food products to improve their nutritional characteristics, health benefits, technological, rheological, sensory properties and shelf life.

5. Acknowledgments

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6. Conflict of interest

According to the authors, there is no conflict of interest.

7. References

[1] Mohamed G. Shehata , Tarek S. Awad b, Dalal Asker, Sobhy A.El Sohaimy, Nourhan M. Abd El- Aziz, Mohammed M. Youssef Antioxidant and antimicrobial activities and UPLC-ESI-MS/MS polyphenolic profile of sweet orange peel extracts. Current Research in Food Science 2021,4, 326–335.

[2] Omran, B.A., Nassar, H.N., Fatthallah, N.A., Hamdy, A., El-Shatoury, E.H., El-Gendy, N.S., Waste upcycling of Citrus sinensis peels as a green route for the synthesis of silver nanoparticles, Energy Sources, Part A Recovery, Util. Environ. Eff. 2018, 40, 227–236.

[3] Singh, B., Singh, J.P., Kaur, A., Singh, N., Phenolic composition, antioxidant potential and health benefits of citrus peel. Food Res. Int., 2020,132, 109114.

[4] Wang, L. J.; Borradaile, N. M. ; Dreu, L. Huff, M. W. Secretion of hepatocyte apo-β-carotene is inhibited by the flavonoids, naringenin and hesperetin, via reduced activity and expression of Acat2 and Mtp, 2014, Res.;42:725–734.

[5] Juliana Tamayo, Tangerine vs clementine what are the difference? Food Guides July 8, 2021. From https:/www.fitibility.com/ clementine vs Tangerine.

[6] Oroian, M. Escriche, I. Antioxidants: Characterization, natural sources, extraction and analysis, Food Res Int., 2015, 74, 10-36.

[7] Solymosi, K.; Latruffe, N.; Morant-Manceau, A.; Schoefs, B. Food colour additives of natural origin. In Colour Additives for Foods and Beverages; Scotter, M.J., Ed.; Elsevier BV: AmsterdamThe Netherlands,2015; pp. 3–34.

[8] Rehman, A.; Tong, Q.; Jafari, S.M.; Assadpour, E.; Shehzad, Q.; Aadil, R.M.; Iqbal, M.W.; Rashed, M.M.A.; Mushtaq, B.S.; Ashraf, W. Carotenoid-loaded nanocarriers: A comprehensive review. Adv. Colloid Interface Sci. 2020, 275, 102048.

[9] Charikleia Chranioti, Aspasia Nikoloudaki, Constantina Tzia Saffron and beetroot extracts encapsulated in maltodextrin, gum Arabic, modified starch and chitosan: Incorporation in a chewing gum system Carbohydrate Polymers 127 2015, 252–263.

[10] Petito, N.; Dias, D.D.S.; Costa, V.G.; Falcão, D.Q.; Araujo, K.G.D.L.Increasing solubility of red bell pepper carotenoids by complexation with 2-hydroxypropyl-_-cyclodextrin. Food Chem.2016, 208, 124–131.

[11] Jong-Bang Eun, Ahmed Maruf, Protiva Rani Das, Seung-Hee Nam , A review of encapsulation of carotenoids using spray drying and freeze drying. Critical Reviews in Food Science and Nutrition, 2019, DOI: 10.1080/10408398.2019.1698511

[12] Nitin Mehta, Pavan Kumar , Akhilesh K. Verma , Pramila Umaraw , Yogesh Kumar ,Om Prakash Malav, Awis Qurni Sazili, Rubén Domínguez José M. Lorenzo, Microencapsulation as a Noble Technique for the Application of Bioactive Compounds in the Food Industry, 2020, A Comprehensive Review

[13] Megahed, M.Y.Studies on composition and antimicrobial effect of carrot carotenes. J. Food Sci., 1985, 60 (5), 1048-1053.

[14] Gentili, A.; Caretti, F. Evaluation of a method based on liquid chromatography–diode array detector–tandem mass spectrometry for a rapid and comprehensive characterization of the fat-soluble vitamin and carotenoid profile of selected plant foods. J. Chromatogr. A 2011, 1218, 684–697. [CrossRef] [PubMed]

[15] Hart, D. J., & Scott, K. J. Development and evaluation of an HPLC method for the analysis of carotenoids in foods and measurement of the carotenoid content of vegetables and fruits commonly consumed, in the UK. Food Chemistry,1995, 54, 101-111.

[16]Chiosa,Valentina.;Mandravel,Cristina.;Kleinjans, J.C.S.andMoonen, E.Determination of β-carotene concentration in orange and apple juice and in vitamin supplemented drinks.2005, AnaleleUniversităNii din București – Chimie, Anul XIV (serienouă),2005, vol. I-II, pg. 253-258.

[17] Edris A.E.,KalembaD .,Adamiec J.and Piątkowski M.Microencapsulation of Nigella sativa oleoresin by spray drying for food and nutraceutical applications, FoodChemistry,2016.204:326333.Available at:doi:10.1016/j.foodchem.2016.02.143.

[18] Norulfairuz, D.; Zaidel, A.; Aqilah, N.; Mohd, Y.M. Efficiency and Thermal Stability of Encapsulated Anthocyanins from Red Dragon Fruit (Hylocereus polyrhizus (Weber) Britton & Rose) using Microwave-assisted Technique. Chem. Eng. Trans. 2015, 43, 127–132. [CrossRef]

[19] Robert, P.; Gorena, T.; Romero, N.; Sepulveda, E.; Chavez, J.; Saenz, C. Encapsulation of polyphenols and anthocyanins from pomegranate (Punica granatum) by spray drying. Int.J. Food Sci.Technol.2010,45, 1386–1394. [CrossRef]

[20] Ogawa, M.; Maia, E.L.; Fernandes, A.C.; Nunes, M.L.; de Oliveira, M.E.B.; Freitas, S.T. Resíduos do beneficiamento do camarão cultivado: Obtenção de pigmentos carotenóides. Ciência e Tecnol. Aliment. 2007, 27, 333–337. [CrossRef]

[21] Re, R.; Pellegrini, N.; Proteggenete, A.; Pannala, A.; Yang, M.; Rice-Evans, C. Trolox Antioxidant activity applying an improved ABTS radical cation decolorization assay. Free Radic. Biol. Med. 1999, 26, 1231–1237. [CrossRef]

[22] A.O.A.C. Association of Official Analytical Chemists, 2005, "Official Method of Analysis, 18th Edition, AOAC, USA: Washington, USA

[23] Vuong, Q.V.; Golding, J.B.; Nguyen, M.H.; Roach, P.D. Production of caffeinated and decaffeinated green tea catechin powders from underutilised old tea leaves. J. Food Eng. 2012,110, 1–8. [CrossRef]

[24] Montero P, Calvo MM, Gómez-Guillén MC, Gómez-Estaca J Microcapsules containing astaxanthin from shrimp waste as potential food coloring and functional ingredient: Characterization, stability, and bioaccessibility. LWT - Food Sci Technol 2016, 70:229–236

[25] Wang, Y., Ye, H., Zhou, C., Lv, F., Bie, X. Lu, Z., Study on the spray-drying encapsulation of lutein in the porous starch and gelatin mixture. Euro Food Res Technol., 2012,

234(1), pp. 157-163. DOI: 10.1007/s00217-011-1630-6

[26] Da Costa, R.S.; Negrão, C.A.B.; Camelo, S.R.P.; Ribeiro-Costa, R.M.; Barbosa, W.L.R.; Da Costa, C.E.F.; Silva Júnior, J.O.C. Investigation of thermal behavior of Heliotropium indicum L. lyophilized extract by TG and DSC. J. Therm. Anal. Calorim. 2013, 111, 1959–1964. [CrossRef]

[27] Saloko, S., P. Darmadji, B. Setiaji, Y. Pranoto A.K. Anal, Encapsulation of coconut shell liquid smoke in chitosan-maltodextrin based nanoparticles. Int. Food Res. J., 2013,20: 1269-1276.

[28] Abonyi, B.I.Feng, H. Tang, J. Edwards, C.G. Chew, B.P. Mattinson, D.S. Fellman, J.K. Quality Retention in StrawberryandCarrotPureesDriedwith Refractance,2002 WindowTM Systemhttps://doi.org/10.1111/j.1365-2621.2002.tb09452.

[29] Paini, M.; Aliakbarian, B.; Casazza, A.A.; Lagazzo, A.; Botter, R.; Perego, P.. Microencapsulation of phenolic compounds from olive pomace using spray drying: A study of operative parameters. LWT Food Sci. Technol. 2015, 62, 177–186.

[30] Abdelwahed,W.; Degobert, G.; Stainmesse, S.; Fessi, H. Freeze-drying of nanoparticles: Formulation, process and storage considerations. Adv. Drug Deliv. Rev. 2006, 58, 1688–1713. [CrossRef] [PubMed]

[31] Jimenez-Aguilar DM, Ortega-Regules AE, Lozada-Ramirez JD, Perez-Perez MCI, Vernon-Carter EJ, Welti-Chanes J Color and chemical stability of spray-dried blueberry extract using mesquite gum as wall material. J .Food Compos Anal 2011, 24:889–894.

[32] Lindalva Maria de Meneses Costa Ferreira, Rayanne Rocha Pereira, Fernanda Brito de Carvalho, Alberdan Silva Santos, Roseane Maria Ribeiro-Cost José Otávio Carréra Silva Júnior Green Extraction by Ultrasound, Microencapsulation by Spray Drying and Antioxidant Activity of the Tucuma Coproduct (Astrocaryum vulgare Mart.) 2021, Almonds.

[33] Álvarez-Henao, M.V.; Saavedra, N.; Medina, S.; Jiménez Cartagena, C.; Alzate, L.M.; Londoño-Londoño, J. Microencapsulation of lutein by spray-drying: Characterization and stability analyses to promote its use as a functional ingredient. Food Chem. 2018, 256, 181–187. [CrossRef]

[34] Thamaket P. Raviyan P., Preparation and physical properties of carotenoids encapsulated in chitosan cross-linked tripolyphosphate nanoparticles," Food and Applied Bioscience Journal, 2015, vol. 3, pp. 69−84, 2015.

[35] Liu, N. Park, H.J. Chitosan coated nanoliposome as vitamin E carrier. Journal of Microencapsulation. 2009. 26: 235–242.

[36] Luis Eduardo Ordoñez-Santos, Jader Martínez-Girón, Rafael Humberto Villamizar-Vargas Encapsulation of β-carotene extracted from peach palm residues: a stability study using two spray-dried processes. Ordoñez-Santos et al / Revista DYNA, 2018, 85(206), pp. 128-134.

[37] Guadarrama-Lezama, A.Y., Dorantes-Alvarez, L., Jaramillo-Flores, M.E., Pérez-Alonso, C., Niranjan, K., Gutiérrez-López, G.F. Alamilla-Beltrán, L., Preparation and characterization of non-aqueous extracts from chilli (Capsicum annuum L.) and their microencapsulates obtained by spray-drying. Preparation and characterization of non-aqueous extracts from chilli (Capsicum annuum L.) and their microencapsulates obtained by spray-drying. Food Eng. 2012,112(1), pp. 29-37, DOI: 0.1016/j.jfoodeng.2012.03.032.

[38] Giles, N.M., S. Kumari, R.A. Stamm, S. Patel G.I. Giles, A hydrogen peroxide electrode assay to measure thiol peroxidase activity for organoselenium and organotellurium drugs. Anal. Biochem.2012, 429: 103-107.

[39] Khaled F. Mahmoud, Mona A. Ibrahim, El-Demery Mervat, Hamdy A. Shaaban, 1Mohie M. Kamil and1Nefisa A. Hegazy Nano-encapsulation Efficiency of Lemon and Orange Peels Extracts on Cake Shelf Life.2016, ISSN 1557-4571 DOI: 10.3923/ajft.2016.63.75.

[40] Naomi M. Mulyadi, Tri D. Widyaningsih, Novita Wijayanti, Renny Indrawati, Heriyanto, and Leenawaty Limantara Microencapsulation of Kabocha Pumpkin Carotenoids , International Journal of Chemical Engineering and Applications, 2017, Vol. 8, No. 6.

[41] Goubet, I., Le Quere, J. L., Voiley, A. J. Retention of aroma compounds by carbohydrates: influence of their physicochemical characteristics and of their physical state, (1998). A review. Journal of Agricultural and Food Chemistry, 46, 1981–1990.

[42] Lara Etzbach, Messina Meinert, Thilo Faber, Carolin Klein, Andreas Schieber, Fabian Weber Effects of carrier agents on powder properties, stability of carotenoids, and encapsulation efficiency of goldenberry (*Physalis peruviana* L.) powder produced by

co-current spray drying. Current Research in Food Science 3 2020.73–81.

[43] Gamboa-Santos, J.; Soria, A.C.; Villamiel, M.; Montilla, A. Quality parameters in convective dehydrated carrots blanched by ultrasound and conventional treatment. Food Chem. 2013, 141, 616–624. [CrossRef] [PubMed]

[44] Álvarez-Henao, M.V.; Saavedra, N.; Medina, S.; Jiménez Cartagena, C.; Alzate, L.M.; Londoño-Londoño, J. Microencapsulation of lutein by spray-drying: Characterization and stability analyses to promote its use as a functional ingredient. Food Chem. 2018, 256, 181–187. [CrossRef]

[45] Yamashita, C.; Chung, M.M.S.; dos Santos, C.; Mayer, C.R.M.; Moraes, I.C.F.; Branco, I. Microencapsulation of an anthocyanin-rich blackberry (Rubus spp.) by-product extract by freeze-drying. LWT Food Sci. 2017,Technol. 84, 256–262.

[46]Bustos-Garza, C.;Yáñez-Fernández, J.; Barragán-Huerta, B.E. Thermal and pH stability of spray-dried encapsulated astaxanthin oleoresin from Haematococcus pluvialis using several encapsulation wall materials. Food Res. Int. 2013, 54, 641–649. [CrossRef]

[47] Rachel A. Itle and Eileen A. Kabelka1 Correlation Between L*a*b* Color Space Values and Carotenoid Content in Pumpkins and Squash (Cucurbita spp.) HORTSCIENCE, 2009, 44(3):633–637.

[48] Cecilia Fiorentini, Guillermo Duserm Garrido, Andrea Bassani, Claudia Cortimiglia, Marta Zaccone, Luana Montalbano,Vanesa Martinez-Nogues, Pier Sandro Cocconcelli, Giorgia Spigno. 2022, Foods https://doi.org/10.3390/foods110100302021. 44(3):633–637

[49] Martha G. Ruiz-Gutiérrez, Carlos A. Amaya-Guerra, Armando Quintero-Ramos,Esther Pérez-Carrillo, and Carmen O. Meléndez-Pizarro, Use of Red Cactus Pear (Opuntia ficus-indica) Encapsulated Powder to Pigment Extruded Cereal. Hindawi Journal of Food Quality 2017, Volume 2017, Article ID 7262464, 12 pages.