UTILIZATION OF PHENOLIC COMPOUNDS EXTRACTED FROM DIFFERENT AGRICULTURAL WASTES THROUGH VARIOUS ENCAPSULATION METHODS

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UTILIZATION OF PHENOLIC COMPOUNDS EXTRACTED FROM DIFFERENT AGRICULTURAL WASTES THROUGH VARIOUS ENCAPSULATION METHODS

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ABSTRACT

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The main objective of this dissertation was to encapsulate the phenolic compounds extracted from strawberry pomace and olive leaf waste with different coating materials (maltodextrin, gum Arabic, chickpea flour and lentil flour) by using various techniques namely, high-speed homogenization (HSH), high-pressure homogenization (HPH), primary and double emulsion methods (PE and DE).

This study was mainly divided into three parts. In the first part of the study, phenolic compounds extracted from strawberry pomace were encapsulated by using different concentrations of maltodextrin and gum arabic mixture. HPH caused to have higher efficiencies (85.99-91.32%) than HSH (81.88-88.15%). Besides, addition of Gum Arabic to maltodextrin increased efficiency.

In the second part of the study, another source obtained from olive leaf waste: olive leaf extract (OLE) was encapsulated by using HSH and HPH with lentil and chickpea flours. Efficiencies after HPH treatments (81.87-85.09%) were higher than those after HSH treatment (80.78-80.26%).

In the third part, encapsulation of OLE through emulsification methods showed that DE (95.45%) was a better encapsulation technique than PE (72.05%) and entrapment techniques (80.26-91.32%). DE with chickpea flour was found to have higher storage stability and lower release rate than DE with lentil flour.

Encapsulated OLE was found to be more heat stable than uncoated OLE during baking. Besides, encapsulation prevented the release of the phenolic compounds in simulated gastric fluid. Release of phenolic compounds in intestinal fluid were found to be higher than the gastric fluid.

As a result, DE was found to be the most efficient encapsulation method.

Keywords: Encapsulation, Strawberry Pomace, Olive Leaf Extract, Double Emulsion, Nuclear Magnetic Resonance Relaxometry

ÖZ

FARKLI TARIMSAL ATIKLARDAN ÖZÜTLENEN FENOLİK BİLEŞİKLERİN ÇEŞİTLİ KAPSÜLLEME YÖNTEMLERİ UYGULANARAK DEĞERLENDİRİLMESİ

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Aralık 2018, 269 sayfa

Çalışmanın ana amacı çilek posasından ve zeytin yaprağından özütlenen fenolik bileşenlerin farklı kaplama maddeleri kullanarak (maltodekstrin, Arap zamkı, nohut ve mercimek unları) ve farklı kaplama işlemleri; yüksek hızlı homojenizatör (YHH), yüksek basınçlı homojenizatör (YBH), tek ve çift katmanlı emülsiyon (TE ve ÇE) yöntemleri uygulanarak kaplanmasıdır.

Bu çalışma üç ana parçaya ayrılmıştır. İlk olarak, çilek posasından elde edilen fenolik bileşenler hapsetme tekniğiyle farklı 3maltodekstrin ve arap zamkı karışımları kullanılarak kaplanmıştır. İkinci olarak zeytin kabuğu özütü hapsetme yöntemiyle ve son olarak emülsiyonlaştırma yöntemiyle, nohut ve mercimek unları kullanılarak kaplanmıştır. Çilek posasından özütlenen fenoliklerin kaplanmasında YBH yönteminin (%85.99-91.32) YHH yöntemine göre (%81.88-88.15) daha verimli olduğu bulunmuştur. Maltodekstrine arap zamkı eklenmesi kaplama verimini arttırmıştır.

Zeytin yaprağı özütü, nohut ve mercimek unları ile YBH ve YHH yöntemleri kullanılarak kaplanmıştır. YBH işleminin ardından kaplama verimlerinin (%81.87-85.09) YHH'ye göre daha yüksek olduğu (%80.78-80.26) görülmüştür.

Zeytin yaprağı özütünün farklı emülsifikasyon yöntemleri kullanılarak yapılan analizler sonucunda ÇE (%95.45) yönteminin TE'ye (%72.05) ve hapsetme tekniğinine (%80.26-91.32) göre daha iyi bir kapsülleme tekniği olduğu anlaşılmıştır. Nohut unu kullanılan çift katmanlı emülsiyonların mercimek unu kullanılanlara göre daha yüksek saklama dayanıklılığı ve daha düşük salınım hızına sahip olduğu görülmüştür.

Kapsüllenmiş zeytin yaprağı özütünün pişirme sırasında kapsüllenmemişlere göre daha dayanıklı olduğu görülmüştür. Ek olarak, kapsülleme işleminin mide sıvısında fenolik bileşiklerin salınımını önlemede etkili olduğu belirlenmiştir. Fenolik bileşenlerin bağırsak sıvısında salınımı mide sıvısında salınımından daha yüksek çıkmıştır.

Sonuç olarak, ÇE en etkili kaplama methodu olarak bulunmuştur.

Anahtar Kelimeler: Enkapsülasyon, Çilek Posası, Zeytin Yaprağı Özütü, Çift Emülsiyon, Nükleer Manyetik Rezonans Relaksometri

To all my beloved family members, especially to my spouse and my baby,

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I dedicate this work to each member of my family.

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LIST OF ABBREVIATIONS

ABTS: 2,2'-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid)

BS: Baking stability

CHI_{10}: Microcapsules of olive leaf extract with chickpea flour mixture (10 %, w/w)

CHI_{20}: Chickpea flour mixture with a mass concentration of 20 %

D_{32}: Surface average diameter (μm)

D_{43}: Volume-weighted average diameter (μm)

D_{50}: Median diameter (μm)

DE: Double emulsion

DE-CHI_{20}: Double emulsion with chickpea flour mixture with a mass concentration of 20%

DE-LEN_{20}: Double emulsion with lentil flour mixture with a mass concentration of 20%

$DPPH^{\cdot}$: 1,1-diphenyl-2-picrylhydrazyl GA: Gum Arabic

EE: Encapsulation efficiency

EPC: Encapsulated phenolic content

FID: Free induction decay

GA: Gum Arabic

GAE: Gallic acid equivalent

HHP: High hydrostatic pressure

HPH: High-pressure homogenizer

HSH: High-speed homogenizer

IS: Instant stability

K: Consistency coefficient (Pa.s)

LEN_{10}: Microcapsules of olive leaf extract with lentil flour mixture (10 %, w/w)

LEN_{20}: Lentil flour mixture with a mass concentration of 20 %

MD: Maltodextrin

NMR: Nuclear magnetic resonance

OLE: Olive leaf extract

OLE TPC: TPC result of OLE (mg GAE/g)

O/W: Oil-in-water emulsion

O1/W/O2: Oil-in-water-in-oil double emulsions

PE: Primary emulsion

PFG NMR: Pulsed field gradient nuclear magnetic resonance

PGPR: Polyglycerol polyricinoleate

PP: Phenolic powder

Rpm: Round per minute

RR: Release rate of OLE

RR PE: Release rate of primary emulsion

RR DE: Release rate of double emulsion

SPC: Surface phenolic content

SD: Standard deviation

SS: Storage stability

US: Ultrasonication

T_1: Spin-lattice (longitudinal) relaxation time

T_2: Transverse relaxation time

TAA: Total antioxidant activity

TD-NMR: Time domain nuclear magnetic resonance

TPC: Total phenolic content

TPC_{corr}: TPC corrected

W/O: Water-in-oil emulsion

W1: Inner aqueous phase of emulsion

W2: Outer aqueous phase of emulsion

W1/O/W2: Water-in-oil-in-water double emulsion

LIST OF SYMBOLS

C_a: Loss of TAA of double emulsion

C_{ao}: Loss of TAA at the first of day of storage

C_1: DPPH˙ concentration immediately after the sample and DPPH˙ solution were mixed (ppm)

d: Dilution rate

D_{32}: Surface average diameter (μm)

D_{43}: Volume-weighted average diameter (μm)

D_{50}: Median diameter (μm)

h_c: The height of the upper part of the centrifuged emulsion (cm)

h_o: The height of the emulsion before centrifugation (cm)

k: Release rate constant (day^{-1})

K: Consistency coefficient (Pa.s)

n: Flow behavior index

τ: Shear stress (Pa)

γ̇: Shear rate (1/s)

k: Release rate constant of OLE

MPa: Mega Pascals

OLE TPC: TPC result of OLE (mg GAE/g)

p: Probability

$PC_{coating}$: Phenolic contents of lentil or chickpea flour mixtures

PE TPC: TPC results of PE (mg GAE/g)

TPC_{corr}: TPC corrected

W_{sample}: Amount of dry sample (mg)

$W2_{After}$: TPC result of W2 after DE formation (mg GAE/g)

$W2_{Before}$: TPC results of W2 before DE formation (mg GAE/g)

CHAPTER 1

INTRODUCTION

1.1 Phenolic Compounds

Phenolic compounds are secondary metabolites that are synthesized by plants during normal development and in response to stress conditions as infection, stabbing, UV irradiation, herbivores and reactive oxygen species (Beckman, 2000). They possess high *in vitro* antioxidant capacity (Gardner, White, McPhail, & Duthie, 2000). In this respect, they defend other compounds or tissues from damages caused by free radicals. In addition with their antioxidant property, phenolic compounds offer a wide range of physiological properties like anti-microbial, anti-inflammatory, anti-allergenic, anti-thrombotic effects (Manach et al., 2004). Moreover, the intake of phenolic compounds decrease the proclivity to several chronic diseases such as several types of cancer (Eberhardt, Lee, & Liu, 2000; Zu et al., 2014), coronary, artery and cardiovascular diseases (Hercberg et al., 1999; Yardim-Akaydin, Özkan, Özkan, Torun, & Şimşek, 2003).

In their general structure, phenolic compounds have an aromatic ring with a hydroxyl substituent and a functional residue. These compounds are classified into different groups according to the number of phenol rings that they contain and the structural elements that bind these rings to another as phenolic acids, flavonoids, lignans and stilbenes (Figure 1). Besides this diversity, most of the naturally occurring phenolic compounds are associated with various carbohydrates and organic acids and with each other (Manach et al., 2004).

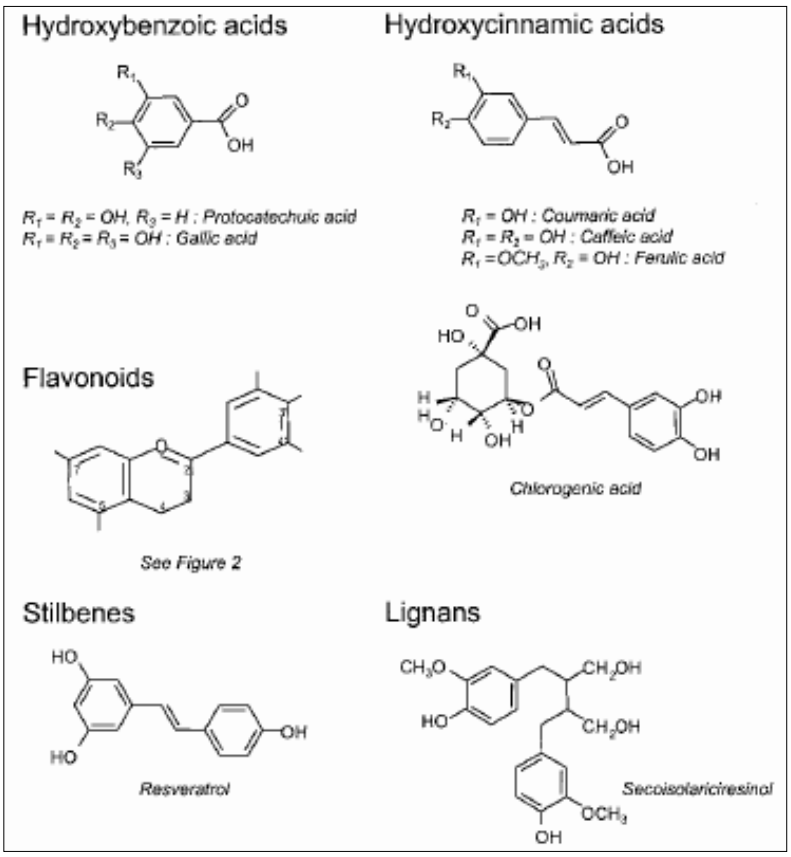

Figure 1 Chemical structure of phenolic compounds (Manach et al., 2004)

Phenolic compounds are responsible for the red, blue or violet color of edible fruits or vegetables such as plums, many berries, apples and eggplant. The color of anthocyanidins changes when pH is altered. They are usually red at low pH, become colorless (pH 4.5) with sulfur compound and then shift to blue when pH increases. As the fruit approaches its ripeness, anthocyanin content usually increases (Peterson & Dwyer, 1998) and therefore color changes.

1.2 Antioxidant Properties of Phenolic Compounds

Normal metabolic processes or exogenous factors such as cigarette smoke, car exhaust fumes and oxidant gases, such as ozone, nitrogen dioxide and sulphur dioxide cause formation of free radicals such as superoxide ($O2^{\cdot}$), hydroxyl ($OH^{\cdot}$) and peroxyl ($RO2^{\cdot}$). They are known to attack and damage body cells because of their missing electron. Various chronic diseases, such as coronary heart diseases, cancer, atherosclerosis and aging are enhanced by such oxidative damages on nucleic acids, lipids and proteins. Antioxidants protect compounds or tissues from damage caused by oxygen or free radicals and prevent or decelerate those oxidation reactions. Therefore, they are health supporting substances (Kirkham & Rahman, 2006). Lipid oxidative rancidity in food is also retarded by antioxidants, so they are deliberately added as food additives to enhance the quality of foods. Butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT), which are synthetic phenolic antioxidants, are added to foods to prevent free radical damage to lipids. However, BHA and BHT are possible carcinogens and there is an increasing demand by the consumers to not use synthetic antioxidants (Duh, Tu, & Yen, 1999; Velioglu, Mazza, Gao, & Oomah, 1998).

There are two major types of antioxidants. Firstly, “primary” (chain breaking, free radical scavengers) and “secondary” (deactivation of metals, inhibition of breakdown of lipid hydroperoxides, singlet oxygen quenching and regeneration of “primary” antioxidants) (Gordon, 1990). Natural and synthetic tocopherols, BHA, BHT, tertiary butyl hydroquinone (TBHQ), alkyl gallates are some examples of primary antioxidants. Secondary antioxidants include ethylenediaminetetraacetic acid (EDTA), citric acid, phosphoric acid, ascorbic acid (useful in canned or bottled products), ascorbyl palmitate (useful in fatty foods because of high solubility in fat phase) and erythrobic acid (isoascorbic acid) (Madhavi, Umamaheswari, & Venkateswarlu, 1995). Antioxidants are generally present as mixtures in plants and in various foods, so there is currently a great activation due to the synergistic interaction between antioxidants. Mixtures of antioxidants produce a more pronounced activity

than the sum of the activities of the individual antioxidants when they are used separately (Duh et al., 1999). Primary antioxidants are often used in combination with other phenolic antioxidants, or with various metal chelating agents in order to have maximum efficiency like the usage of ascorbic acid with tocopherols (Doba, Burton, & Ingold, 1985).

1.3 Extraction of Phenolic Compounds

It is important to extract phenolic compounds from plants or agricultural wastes for the fabrication of phenol-rich products, which are highly used in the functional food production, pharmaceutical, nutraceutical and cosmetic industry.

Maceration, pressurized liquid extraction, microwave and ultrasound assisted extractions are some examples of different extraction types. Ethanol, methanol, acetone, water or their mixtures can be used as solvent; nevertheless, special care is needed when foods are processed. It must not have any toxicity or should not leave any dregs after utilization with respect to health and safety concerns (Adil, Yener, & Bayindirli, 2008).

Maceration is the classical method for the extraction of phenolic compounds. This technique is simply associated with the dissociation of soluble phenolic compounds by diffusion from the solid matrix using a solvent. The mechanism has two stages: initial and diffusion stage. In the initial stage, solid particles are swollen due to absorption of the solvent through the solid phase caused by osmotic forces and capillarity. In the second stage, phenolics diffuse from the solid phase into the medium (Ćujić et al., 2016).

In the application of pressurized liquid extraction, generally a solid sample is put into a stainless steel extraction cell with a solvent under high pressure (500-3000 psi) and above its boiling point (40 - 200 ºC) for a short time (5 - 15 min) (Garcia-Salas, Morales-Soto, Segura-Carretero, & Fernández-Gutiérrez, 2010).

Microwave assisted extraction gives a chance to perform extractions in a small time and without light, which are effective factors for phenolic compounds to be destroyed. Recently, microwave assisted extraction, also called microwave-assisted process has been used for extraction of organic compounds from food matrices and edible plants. It is a method in which the microwave radiation energy is used to heat solvents rapidly and effectively. Desorption of chemicals from the matrix is enhanced because cell disruption is promoted by internal superheating owing to water within the plant absorbs microwave energy. A study shows the usage of microwave assisted extraction in determination of the total phenolic contents of eight bean types (Sutivisedsak et al., 2011). Microwave assisted extraction is commonly applied for its time saving effect; however, special care must be taken when the usage of flammable solvents or with samples that contain constituents which couple strongly with microwave radiation. A rapid rise in temperature may occur and thereby result in potentially hazardous situations (Jáuregui & Galceran, 2001).

In ultrasound assisted extraction, the surface area between the solid and liquid phases is greater owing to the disruption of particles (Filgueiras, Capelo, Lavilla, & Bendicho, 2000). There are two types of application; namely, using probe system and bath set up. Both methods are suitable for extraction purposes (Rostagno, Palma, & Barroso, 2003). The phenolic compounds in strawberries was extracted previously by using ultrasound (Herrera & Luque De Castro, 2005), but the researchers did not utilize strawberry pomace.

1.4 Microencapsulation

It is very healthy to consume natural phenolic compounds; however, they are very vulnerable to oxidants, light, heat (Lou, Lin, Hsu, Chiu, & Ho, 2014), and enzymatic activities (Bakowska, Kucharska, & Oszmiański, 2003). Easy degradation during storage, unpleasant taste, and off-color are the disadvantages of consuming natural

phenolic compounds. The stability and shelf life of sensitive food ingredients can be enhanced by several techniques such as drying, irradiating, and microencapsulation.

Microencapsulation is one of the solutions to overcome these problems (Cilek, Luca, Hasirci, Sahin, & Sumnu, 2012; Laine, Kylli, Heinonen, & Jouppila, 2008; Riaz & Masud, 2013). It can be achieved by using ultrasonic homogenization, high-pressure homogenization, and high-speed homogenization. Additionally, the advantages of using microcapsules are the prolongment of shelf life, ease of handling, prevention of off-flavor, the improvement of stability, and bioavailability (Neethirajan & Jayas, 2011). In microencapsulation, core material (active agent to be encapsulated) is embedded inside a homogeneous or heterogeneous matrix, which is named as the coating material. Microencapsulation is one of the techniques used for enhancing the shelf life and stability of food ingredients for over 60 years. It is described as a technique in which a bioactive compound is encapsulated by a biopolymer, so that it is protected from oxygen, light, water or other environmental conditions (Desai & Jin Park, 2005). Natural phenolic compounds can be more stable with the help of encapsulation. It can be performed to store phenolic compounds extracted from fruits or vegetables from which compounds can be released under specific conditions (Bakowska-Barczak & Kolodziejczyk, 2011; Laine et al., 2008; Luca, Cilek, Hasirci, Sahin, & Sumnu, 2014; Saenz, Tapia, Chavez, & Robert, 2009).

1.4.1 Entrapment

For the encapsulation of bioactives in aqueous solutions, entrapment is usually implemented. As a matter of fact, it is a part of encapsulation process. Before microcapsules are obtained, a suspension is prepared. It is composed of core material, the substance that is wanted to be capsulated, and coating material dissolved in suitable solvent. After this technique is applied, it can be dried (by spray or freeze drying) in order to obtain microcapsules in powder form (Bakry et al., 2016).

Suspensions are generally obtained by two types of emulsification techniques such as low energy emulsification and high energy/pressure emulsification. Phase inversion temperature or phase inversion composition is low energy emulsification methods. Ultrasonication and microfluidization are used as high energy emulsification methods (Jafari, He, & Bhandari, 2007b).

Several morphologies can be developed for encapsulation, but two of them are commonly seen. The first type is in the shape of mononuclear capsules, having a single core restricted by a shell, while the second is aggregates, in which the core material embedded in a matrix (Figure 2). Applied process technologies and coating materials (as well as the core material) affect the specific shape and efficiency of capsules.

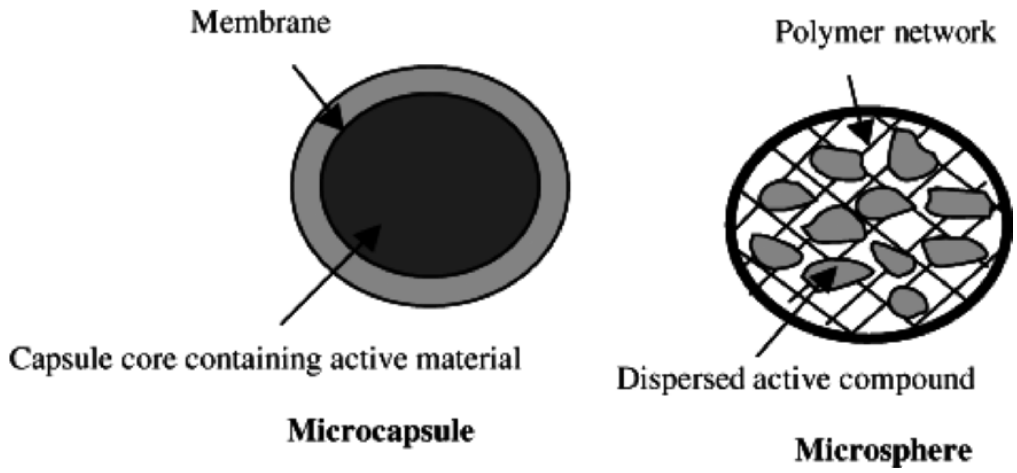

Figure 2 A schematic representation of encapsulated bioactives. (Cilek, 2012)

1.4.2 Single and Double Emulsion

Two immiscible liquids, usually oil and water, compose an emulsion with a continuous phase and a dispersed phase. Non-soluble small droplets are dispersed in the continuous phase. Emulsions are grouped according to the distribution of the oil and aqueous phases. An emulsion type can be oil-in-water (O/W) emulsion in which oil droplets are dispersed in an aqueous phase, for instance, mayonnaise, milk, cream, soups, and sauces. In the other type, water droplets are dispersed in an oil phase which is called water-in-oil (W/O) emulsion such as margarine, butter, and spreads. W/O type of emulsion can be seen in Figure 3 and Figure 4.

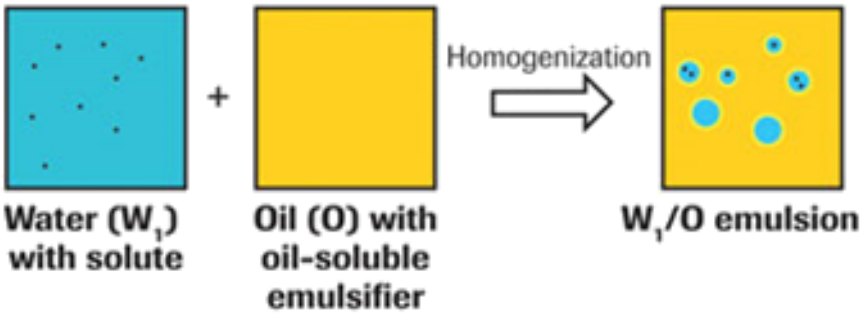

Figure 3 Water-in-oil type of an emulsion with hydrophilic solute.

The latest trend in food studies is to prepare multiple emulsions. Double emulsions are complex systems and can be described as being emulsions within emulsions. Multiple emulsions are multi-sectioned systems where the globules of the dispersed phase contain smaller dispersed droplets (Cilek Tatar, Sumnu, & Sahin, 2017). The two major types of multiple emulsions are water-in-oil-in-water (W1/O/W2) and oil-in-

water-in-oil (O1/W/O2) double emulsions (Figure 4 and Figure 5). Even a triple emulsion study was conducted in 2011 by using double emulsions (Hasinovica and Friberg, 2011). These emulsions have many valuable properties like the ability to entrap and protect sensitive substances as well as control their release from the inner to outer phase (Bou et al., 2014) and to reduce the total fat content of emulsion-based food products (Lobato-Calleros et al., 2006).

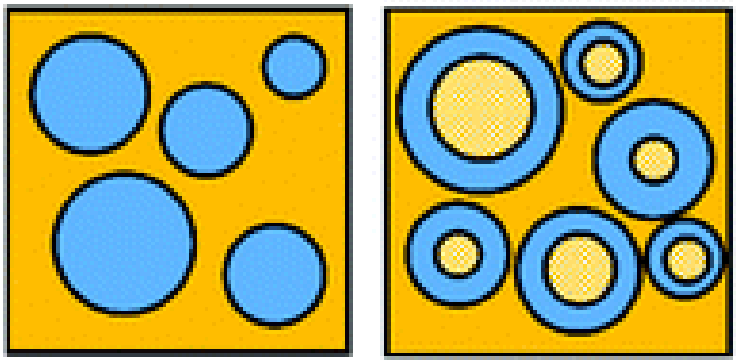

Figure 4 Schematic representative of water-in-oil (a) and oil-in-water-in-oil (O/W/O) type of emulsions.

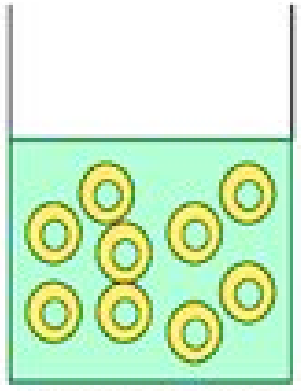

Figure 5 Schematic representative of water-in-oil-in-water (W/O/W) type of double emulsion.

Emulsions can easily be formed from food-oriented ingredients by simple processing techniques; therefore, food engineers, food scientists, and producers are highly interested in emulsions. In recent years, different systems have been proposed for the delivery of hydrophobic and hydrophilic bioactive compounds in foods. In this context, O/W emulsions have been widely used for the delivery of hydrophobic compounds, and W/O emulsions were chosen for the delivery of hydrophilic ones.

Double or multiple emulsions offers a variety of promising opportunities for the food industry, e.g. the delivery of hydrophobic compounds in aqueous systems, it allows for the encapsulation of hydrophilic bioactive compounds and can help improve the fat content of foods (by reducing fat content and providing a healthier fatty acid profile) (Yildirim, Sumnu, & Sahin, 2017).

Although the double emulsions constitutionally have a major problem of thermodynamic instability, they offer wide range of possible advantages over single emulsions. Many food items are emulsion systems; hence, the double emulsions

become a valuable technology to improve food emulsions. Multiple emulsions have many advantages, such as the ability to entrap and protect sensitive substances as well as control their release from the inner to the outer phase, more than single emulsions. It has been found that double emulsions are more suitable than the single ones for encapsulating the valuable compounds in terms of encapsulation efficiency (Vasiljevic, Parojcic, Primorac, & Vuleta, 2009).

1.4.3 Ultrasonication

Different types of ultrasound devices are used to produce microcapsules. The main phenomenon responsible for ultrasonication is acoustic cavitations. Turbulence was generated by the occurrence of micro-size bubbles, and with pressure difference explosion of bubbles makes high levels of turbulence. Turbulence causes smaller particles to be obtained (Li & Fogler, 1978). Increasing sonication time caused formation of smaller emulsion droplets by increasing energy input. This can be explained by the increasing disruption with the droplet deformation (Jafari, He, & Bhandari, 2007a).

Zambaux et al. (1999) investigated the influence of the sonication time on the activity of protein C. They used sonication technique for emulsification in order to obtain the protein C nanoparticles. It was indicated that when the sonication time decreased or when a vortex was used, the residual activity of protein C increased. It was stated that the emulsification operated with a vortex is less stable than by sonication, probably due to increase in contact area between protein and organic solvent.

Yeo and Park (2004) proposed a microencapsulation method by using an ultrasonic atomizer (Yeo & Park, 2004). In this method, lysozyme was succesfully encapsulated with no loss of functional integrity and releasing was found to fit zero-order kinetics for over 50 days. Yeo and Park (2004) stated that the proteins (core material) were not subjected to detrimentally strong mechanial stresses when the microcapsules formed by the collision between different species of liquid droplets.

1.4.4 Microfluidization

A high-pressure homogenizer (HPH) can be used to obtain emulsions (Figure 6). A microfluidizer, a type of HPH, is working on the principle of a pressure stream which is divided into two parts; each part of stream passes through an orifice and so transports the fluid to the crucial part of microfluidizer that is the interaction chamber (Mahdi Jafari, He, & Bhandari, 2006). In the interaction chamber of the microfluidizer, two channels coming from opposite sides collide with another. The stream is carried by a pump which is able to pressurize the internal compressed air up to approximately 300 MPa.

Figure 6 A batch type high-pressure homogenizer.

In a recent study, ultrasonication and HPH was compared and results showed that ultrasonication provided to the apple juice less polyphenoloxidase inactivation than HPH. Also, ultrasonication showed lower energy consumption than HPH (Bot et al., 2018).

The researchers recently described the principles and equipment for HPH and high hydrostatic pressure (HHP) technologies, as well as their effects on microorganisms, enzymes, food constituents, juice structure, physical and technological properties. Finally, they also described the industrial perspectives, commercial use and future challenges of the techniques. Although similar in name, the technologies are completely different in relation to principles and processes of the HHP. The HHP process is based on the isostatic principle and the principle of Le Chatelier; where the pressure is equally applied in all directions of the foods, being responsible for the observed reaction. It can be used for solid, fluid, or particulate food, being conducted in-package and discontinuously. On the other hand, different from the HHP process, in the HPH process, there is shear stress distribution across the product, which is the responsible for the obtained results. The HPH process is continuous and available only for fluid products (Augusto, Tribst, & Cristianini, 2017).

In microfluidization, there was a critical level above which the emulsions became over-processed (Jafari et al., 2007b). Therefore, it is important to find the optimum process time of microfluidization. In the same research, encapsulation efficiency of oils and microemulsions was obtained by two different methods; microfluidization and ultrasonication. It was found that emulsion size was increased whereas powder size was decreased when ultrasonication was used to prepare capsules (Jafari et al., 2007b). The results also showed that the emulsion temperature increased by increasing pressure or time. Although a cooling jacket was used at the exit of the valve, significant temperature rise was observed in the emulsion (Bouaouina, Desrumaux, Loisel, & Legrand, 2006). Another problem can be caused from the high-energy densities used during microfluidization because volume of emulsion passing through chamber is smaller than the emulsion volume in the emulsification zone (Jafari et al., 2007a).

The effects of high-pressure homogenization and thermal treatment on the phenolic bioaccessibility and the ABTS scavenging activity of apple, grape and orange juice were also investigated by researchers. high pressure homogenization diminished apple juices's total phenolic bioaccessibility 29.3 %, whereas thermal treatment hardly affected it (He et al., 2016).

1.4.5 Coating Material Types

As coating materials, different kinds of natural or modified polysaccharides, proteins, and lipids can be used (Desai & Jin Park, 2005; Sanchez, Baeza, Galmarini, Zamora, & Chirife, 2013). Maltodextrin (MD) is preferred commonly as coating material in the food industry because of its soft flavor, high solubility in water, and transparent color (Saenz et al., 2009). Gum arabic (GA) is also widely used as a coating material due to its high emulsifying ability and low viscosity (Gabas, Telis, Sobral, & Telis-Romero, 2007).

Flours are also widely used in the food industry for baking however they were not used as a coating material for encapsulation. Legume-flour usage is getting importance in baking because the number of people with celiac disease is getting higher. Legume flours do not contain gluten, therefore people with celiac disease can consume legume flours. Consuming legumes is very healthy because of their high protein and high fiber contents. There are studies about using protein and polysaccharides mixture in the coating of microcapsules, and the results showed that encapsulation efficiency became higher when coating material is mixed with a protein. Besides, a study has shown that phenolic content is influenced by a high pressure homogenizer (He et al., 2016). Although increasing pressure or number of cycles may aid in a more homogenous dispersion and higher encapsulation efficiency, HPH may cause a temperature rise in the product (Bot et al., 2018; Håkansson, 2018) resulting in the loss of phenolic content. Therefore, it is beneficial to find an optimum condition for performing microencapsulation of natural phenolic compounds.

Researhers investigated the effects of different coating materials on the encapsulation efficiency and particle size of seed oil capsules (Lim, Tan, Bakar, & Ng, 2012). It was concluded that using GA and MD (DE 10) mixture as coating material had the smallest particle size among all other mixtures except sodium caseinate and maltodextrin mixture. In general, GA is known to have stabilizing and emulsifying effects on encapsulation (Krishnan, Bhosale, & Singhal, 2005).

Similarly, researchers encapsulated cumin oleoresin by spray drying using GA, MD and modified starch and their blends as wall materials (Kanakdande, Bhosale, & Singhal, 2007). According to their results usage of gum Arabic / MD / modified starch blend of 4/6:1/6:1/6 resulted in better protection when compared to protection provided by GA alone. Besides, in order to avoid the degradation of vitamin over the storage time, the bioactives compounds were encapsulated by spray drying with GA as the wall material (Romo-Hualde, Yetano-Cunchillos, González-Ferrero, Sáiz-Abajo, & González-Navarro, 2012).

1.4.6 The Type of Emulsifier

The selection of surfactant when preparing an emulsion is very important because surfactant type and its concentration affect the emulsion type and stability. In a recent research, polyglycerol polyricinoleate (PGPR) and lecithin found to be the best combination to prepare DE (Altuntas, Sumnu, & Sahin, 2017), therefore PGPR and lecithin were used as surfactants. It is also necessary to add an oil-insoluble solute to W1 and W2 phases to stabilize emulsions (Kanouni, Rosano, & Naouli, 2002). In order to keep the osmotic pressure between the phases, NaCl was added to aqueous phases (Hemar, Cheng, Oliver, Sanguansri, & Augustin, 2010). It was found that NaCl modulated the release rate of core material (Sapei, Naqvi, & Rousseau, 2012). The added salt led to a stronger reduction in interfacial tension by lecithin (Scherze, Knoth, & Muschiolik, 2006) and it was shown that the surface potential is reduced by the addition of electrolytes causing instability of the emulsions (De Vleeschauwer, D. &

van der Meeren, P., Vanderdeelen, 1996). The NaCl effect on lecithin stabilized W/O emulsions can also be from electrostatic effects. Since the polar heads of phospholipids repel each other by electrostatic repulsion, their close association is reduced (Walstra, 2003). Thus, the possible interfacial film breakdown by the presence of salt causing coalescence was reduced by addition of salt (Scherze et al., 2006).

A stable emulsion is one with no discernible change in the size distribution of the droplets, or their state of aggregation, or their spatial arrangement within the sample vessel, over the time-scale of observation. This time-scale may vary from hours to months depending on the situation. The dominant mechanisms of instability are gravity creaming, Ostwald ripening, flocculation and droplet coalescence (Cilek Tatar et al., 2017). Table 1 gives the main factors affecting the stability of an emulsion. It is seen that emulsifier type is effective on the stability. The state of flocculation of the droplets is dependent on the interactions between stabilising layers, which in turn depends on factors such as the biopolymer surface coverage, the layer thickness, the surface charge density, and the aqueous solution conditions (especially pH, ionic strength, and divalent ion content). For a freshly prepared fine triglyceride oil-in-water emulsion, the most obvious initial manifestation of instability is creaming, which typically leads on to macroscopic phase separation into separate discernible regions of cream and serum. This may then be followed by droplet coalescence within the cream and “oiling off” at the top of the sample. All these factors affect the stability of emulsions.

Table 1 Principal factors affecting oil-in-water emulsion stability (Dickinson, 2003).

Droplet-size distribution
Initially determined by
Emulsification equipment
Concentration of emulsifier
Type of emulsifier
Oil/water ratio
Other factors (temperature, pH, viscosity)
Nature of interfacial adsorbed layer
Determined by
Concentration and type of emulsifier
Interactions of adsorbed species
Competition between adsorbed species
Nature of continuous aqueous phase
Rheology, solvent quality, ionic environment, unadsorbed polymers and amphiphiles
Nature of dispersed oil phase
Solid/liquid content
Solubility in continuous phase

Food systems generally contain proteins and polysaccharides which can be used as gelling agents. Table 2 shows the common general characteristics and the differences between proteins and polysaccharides as functional biopolymers in foods. They have both complicated structure and complex aggregation behavior. Whereas proteins have emulsifying activity, polysaccharides have thickening activity (Dickinson, 2003). Flours also contain considerable amount of proteins and polysaccharides; therefore, they can be used as emulsifying agent.

Table 2 The common general characteristics and the differences between proteins and polysaccharides as functional biopolymers in food systems (Dickinson, 2003).

Similarities	
Natural polymers	
Widespread in food colloids	
Used in pharmaceuticals, cosmetics, personal products	
Environmentally friendly polymers	
Complicated structure	
Complex aggregation behavior	
Gelling / stabilizing agents	
Differences	
Proteins	*Polysaccharides*
Wide-ranging structures	Similar structures
Reactive	Unreactive
Monodisperse	Polydisperse
Many segment types	Few segment types
Linear chain	Linear or branched
Flexible chain	Stiff chain
Medium molecular weight	High molecular weight
Small molecular volume	Large molecular volume
Amphiphilie	Hydrophilic
Surface-active	Not surface-active
Polyelectrolyte	Non-ionic or charged
Emulsifying / foaming	Thickening / waterholding
Temperature sensitive*	Temperature insensitive
Strong surfactant binding	Weak surfactant binding

*That is the structure and properties of most proteins can change drastically when heated above a characteristic ‘denaturation temperature’.

1.5 Agricultural Wastes

1.5.1 Strawberry Pomace

Strawberries (*Fragaria x ananassa*) contain high amount of phenolic compounds (Jaroslawska et al., 2011a). Consuming strawberries is healthy; however, as mentioned in the *Section 1.1*, phenol-rich fruits have short shelf lives. There are several ways to keep phenol-rich fruits fresh longer. Juice production, one of the major utilization of fruits, is a process through which pomace is obtained as a by-product. Similar to fruits, their pomace is also rich in phenolic compounds and antioxidants (Khoo, Clausen, Pedersen, & Larsen, 2011). In order to benefit from phenolic compounds, it is important to extract natural phenolic compounds from fruits or plants. One way to extract them is maceration, which is simply associated with the dissociation of soluble natural phenolic compounds from a solid matrix (Rahmanian, Jafari, & Wani, 2015).

Strawberry production in Turkey was 415 000 tons in 2016, whereas it was 211 000 tons in 2006 (TÜİK, 2016). Production of strawberry is high and growing. Since cultivation of strawberry is substantially high, different kinds of strawberries have been studied by several researchers in Turkey (Kafkas, Koşar, Paydaş, Kafkas, & Başer, 2007; Karacam, Sahin, & Oztop, 2015; Pirlak & Köse, 2009; Vulić et al., 2011). Production of strawberry juice, its incorporation inside mixed fruit juices and production of concentrated strawberry juice cause accumulation of strawberry pomace in Turkey (Ministry of Trade, 2016). Strawberry pomace (Figure 7) was investigated by researchers in order to more benefit from this valuable by-product (Jaroslawska et al., 2011b; Kumar, Chanalia, Gandhi, & Dhanda, 2016; Pieszka, Gogol, Pietras, & Pieszka, 2015).

Figure 7 Strawberry pomace obtained after strawberry juice production.

1.5.2 Olive Leaf Extract

Olive leaf extract (OLE) is regarded as one of the resources with the richest polyphenolic compounds among the plants (Ganje et al., 2016; Rahmanian, Jafari, & Galanakis, 2014). OLE has also high amount of phenolic compounds and high antioxidant capacity (Moudache, Colon, Ner?n, & Zaidi, 2016). Besides, researches have shown that phenolic compounds available in olive leaf display antimicrobial and antifungal effects (Pereira et al., 2007; Rahmanian et al., 2015) . Phenolic compounds as well as natural antioxidants, are secondary metabolites and found in olive leaves (Moudache et al., 2016). Unfortunately, they are very sensitive; therefore, increasing their stability and shelf life from chemical and physical damage will be valuable for food and health sciences. OLE, which is shown in Figure 8, can be more stable with the help of encapsulation.

Figure 8 Olive leaf extract.

A recent research was about olive leaf encapsulation within DE prepared by sonication. Nano-emulsions were loaded with oleuropein and it was found that after 28 days storage, emulsions released approximately 40% of their total content (Gharehbeglou, Jafari, Homayouni, Hamishekar, & Mirzaei, 2019). Although these researchers compared the results related to the concentration change of surfactants or core material, there is lack of study about characterization of OLE containing emulsions. There are researches about phenolic content analysis of DE (Akhtar, Murray, Afeisume, & Khew, 2014; Barroso et al., 2014), but the purposes of these studies were to decrease lipid oxidation rate (Flaiz et al., 2016) and to investigate the *in vitro* digestion of DEs (Ydjedd et al., 2017). Accelerated shelf life test of microencapsulated olive leaf extract in tomato paste has been conducted (Ganje et al., 2016). In another study, phenolic content and antioxidant activity of olive by-products and olive leaf extract were also performed and it was found that at low concentrations olive leaf extract showed an unusual combined antibacterial and antifungal action, which suggest their great potential as nutraceuticals, particulalry as a source of phenolic compounds (Moudache et al., 2016).

Olive production in Turkey is very high, more than 2 000 000 tons in 2017 (TÜİK, 2017). The Mediterranean olive tree (*Olea europaea*) is one of the first trees to be domesticated and is currently of major agricultural importance in the Mediterranean region as the source of olive oil (Cruz et al., 2016). There are various amounts of olive tree in Turkey (TÜİK, 2017) and therefore its leaves. The olive leaf has a length about 5 - 6 cm with the width of the middle part is 1 - 1.5 cm (Figure 9).

Figure 9 A branch of an olive tree, with its leaves and fruit.

Olive trees can live up to 400 years and are always green. They have leaves on branches during all the seasons; however, their leaves can live only 18-30 months (Baytop, 1999; Cruz et al., 2016). They shed leaves, but leaf production always occurs

during life span of a tree. Therefore, utilization of this valuable waste, olive leaf, will be beneficial to gain more natural phenolic compounds and anti-microbial agents from this natural waste source.

1.6 Objective of the Study

Utilization of industrial by-products for phenolic compound extraction and encapsulation have gained importance in recent years. In Turkey, production of strawberry is considerably high (TÜİK, 2016). After consuming strawberry as juice, huge amount of waste is gathered as the pomace (TÜİK, 2017). Extraction of phenolic compounds from this pomace will be very beneficial due to its considerably high phenolic content. Polyphenols extracted from the pomace can further be used in the production of natural food additives.

Another waste that is very highly produced in Turkey is the olive leaf extract (TÜİK, 2017). OLE has very valuable phenolic compounds and has an antimicrobial effect. However, it has an undesired taste and is vulnerable to environmental conditions. The encapsulated OLE could be efficiently used in food industry and could replace synthetic antioxidants used in the production of functional foods, cosmetics and pharmaceuticals.

Encapsulation is an important method that is used to enhance the shelf life and stability of compounds and to prevent unwanted taste and odor of food ingredients like phenolics.

Strawberry pomace was investigated by researchers (Jaroslawska et al., 2011b; Kumar et al., 2016; Pieszka et al., 2015); however, the researches were not focused on the encapsulation of phenolic compounds obtained from the pomace. Altough a recent research was about olive leaf encapsulation within DE prepared by sonication (Gharehbeglou et al., 2019), there is lack of study about characterization of OLE containing emulsions because they investigated the concentration change of

surfactants and core material. In literature, there are researches about phenolic content analysis of DE (Akhtar et al., 2014; Barroso et al., 2014); however, the purposes of these studies were to decrease lipid oxidation rate (Flaiz et al., 2016). Legume flours have been increasingly using in the food industry for baking to produce gluten-free goods and there are studies about using protein and polysaccharides mixture in the coating of microcapsules, but flours were not used as a coating material for encapsulation. There is no study about comparision of microencapsulation of agricultural wastes, strawberry pomace and olive leaf extract, by using different coating materials including different flour types with different encapsulation techniques.

The objective of this study was to encapsulate the phenolic compounds extracted from strawberry pomace and olive leaf extract by using different encapsulation methods. In addition, the effects of coating material formulation, encapsulation technique and microfluidization parameters on the encapsulation efficiency, antioxidant activity, surface morphology, release rate, storage stability, particle size, color, kinetic modelling and NMR results were investigated. Baking stability and *in vitro* release tests were performed to cakes with encapsulated OLE to analyze the bioavailability of phenolic compounds.

CHAPTER 2

MATERIALS AND METHODS

2.1 Materials

Strawberry pomace was supplied from KARMEY Fruit Juice Factory located in Karaman, Turkey. Stems and other foreign particles were removed by screening. Remaining pomace was stored in a deep-freezer (-80 ^{o}C, D 8340 SM; Beko, Istanbul, Turkey) for 2 months. Maltodextrin (MD) and gum arabic (GA) were supplied by Sigma-Aldrich Chemical Co. (St. Louis, MO, USA), and they were used as coating materials. All other reagents used in this study (Folin-Ciocalteu's phenol reagent, sodium carbonate, ethanol (absolute), methanol G CHROMASOLV®, gallic acid, and acetic acid (≥ 99.7 %) were analytical grade and purchased from Sigma-Aldrich Chemical Co. (St. Louis, MO, USA).

OLE was purchased in powder form from FURLAB (İstanbul, Turkey) and stored at -20 °C in a deep freezer (D 8340 SM; Beko, Istanbul, Turkey). Polyglycerol polyricinoleate (PGPR) and lecithin were supplied from ETI Food Corp. (Eskisehir, Turkey). Lentil flour and chickpea flour were obtained from Smart Chemical Trading Co. Inc. (Turkey) and Molar Chemical Materials Trading Co. Inc. (Turkey) respectively. Mean values for analysis of lentil flour (as analyzed by AOAC method, 1996) are moisture, 7.47, protein 24.75, fat 1.61 and starch 52 g per 100 g lentil flour. The chemical composition of chickpea flour (as analyzed by AOAC method, 1996) are moisture, 6.13, protein 22.15, fat 7.53 and starch 46.3 g per 100 g lentil flour.

Folin-Ciocalteau's phenol reagent, sodium carbonate, 1,1-diphenyl-2-picrylhydrazyl (DPPH), methanol G CHROMASOLV®, gallic acid, ethanol (absolute), acetic acid (100%), sodium chloride, pancreatin from porcine pancreas, pepsin from porcine

gastric mucosa, and sodium hydroxide (NaOH, ACS reagent, ≥ 97.0 %, pellets) were purchased from Sigma-Aldrich Chemical Co. (St. Louis, MO, USA). Sunflower oil and other ingredients used in the production of cake were purchased from the local markets.

2.2 Methods

2.2.1 Microcapsules Obtained from Strawberry Pomace and Their Characterization

2.2.1.1 Extraction of Phenolics from the Strawberry Pomace

Previously frozen strawberry pomace was freeze dried (Alpha 1-2 LD plus; Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany) at -52 ^{o}C below 7.5 Pa for 48 h. Prior to encapsulation, maceration was performed to obtain phenolic powder (PP) (Figure 10). As in the study of Cilek et al. (2012), extraction was performed in 500 mL sealed glass flasks containing 10 g pomace and 200 ml ethanol-water solvent (1:1, in volume fraction). Flasks were placed in a shaking water bath at 70 rpm for 24 h (GFL Gesellschaft für Labortechnik mbH, Burgwedel, Germany), at 30 ^{o}C. Then, solid particles were removed by a cloth filter, and the liquid part was vacuum filtered. After that, it was concentrated by a rotary evaporator (Laborota 4000 efficient; Heidolph Instruments GmbH & Co, Schwabach, Germany) at 40 ^{o}C under vacuum. The concentrate was frozen at - 20 ^{o}C in a deep freezer. Frozen concentrate was freeze dried at -52 °C for 72 h to obtain dry phenolic powder (PP).

Figure 10 Phenolic powder obtained from strawberry pomace.

2.2.1.2 Preparation of microcapsules of phenolic powder

Maltodextrin (MD) was dissolved in distilled water (to have mass fractions of 20.0, 16.0 and 12.0 %). Gum arabic (GA) solutions having mass fractions of 4.0 and 8.0 % were prepared by using distilled water. After 2 hours, MD and GA solutions were mixed using a magnetic stirrer (MR 3001K, Heidolph Instruments GmbH & Co, Schwabach, Germany) at 1 250 rpm to obtain a total mass fraction of 10% with MD:GA ratios of 10:0, 8:2 and 6:4.

PP and coating solution were mixed to obtain the core to coating ratio of 1:20 (in mass fraction). The suspensions were homogenized using a high-speed homogenizer (HSH), and then, selected suspensions were treated by a high-pressure homogenizer (HPH) to analyze the effect of high-pressure homogenization on encapsulation efficiency. HSH was performed at 15 000 rpm for 30 s (IKA T25 digital Ultra-Turrax, Selangor, Malaysia) (Figure 11). A batch type HPH was used at different pressures (50 and 70 MPa) and different process cycles (3, 5, and 7 cycles) (Nano Disperser, NLM 100, South Korea). Then, suspensions were freeze dried under the same conditions. Finally,

dry samples were manually ground between 2 pieces of aluminum foils by using a roller to obtain microcapsules in homogenous size. Each experiment was triplicated.

Figure 11 High-speed homogenizer (IKA T25 digital Ultra-Turrax, Selangor, Malaysia).

2.2.1.3 Total Phenolic Content Analysis

Total phenolic content (TPC) of PP and dry microcapsules were determined by the modified version of Folin-Ciocalteu method (Beretta, Granata, Ferrero, Orioli, & Facino, 2005). In this study, the same method described in our previous study (Cilek, Luca, Hasirci, Sahin, & Sumnu, 2012) was used. Samples were accurately weighed as 100 mg and dissolved in 3 ml ethanol:acetic acid:water mixture (50:8:42, in volume fraction). Then, the mixture was agitated using Vortex (ZX3, VELP Scientifica, Usmate, MB, Italy) for 1 min, and filtered through a micro filter (0.45 µm, Gema Medical Filter, Spain). Then, 500 µl diluted sample and 2.5 ml 0.2 N Folin-Ciocalteau (2N, Sigma-Aldrich F9252) reagent were put into a tube, and vortexed. After being kept in a dark place for 5 minutes, 2 ml of 75 g/L sodium carbonate (Sigma-Aldrich

S7795) solution was added to tubes and mixed again. Samples were kept at room temperature (25°C) in the dark for 1 h, and then absorption at 760 nm was measured by using UV/VIS spectrometer T 70, (PG Instruments LTD, UK).

Calibration curve was prepared with different gallic acid concentrations (15, 30, 60, 90, and 120 mg/l) in ethanol:acetic acid:water mixture (50:8:42, in volume fraction), so TPC results were expressed as gallic acid equivalents in mg per gr dry weight (mg GAE/g). Calibration curve was given in Appendix A.

2.2.1.4 Surface Phenolic Content Analysis of Microcapsules

For the determination of surface phenolic content (SPC), the same method described in the section of *2.2.1.3 Total phenolic content analysis* was applied by using a different solvent. Microcapsules (100 mg) were dissolved for 1 min in a 3 ml of ethanol:methanol mixture (1:1, in volume fraction) by using a vortex. A calibration curve was prepared in different concentrations of gallic acid (10, 20, 30, 40, and 50 mg/L) using ethanol:methanol mixture 1:1 (in volume fraction). Calibration curve was given in Appendix A.

2.2.1.5 Encapsulation Efficiency

The difference between TPC and SPC was defined as the encapsulated phenolic content (EPC), which was used to calculate the encapsulation efficiency (EE) of microcapsules according to *Equation 1:*

$$EE\ (\%) = EPC \times 100 / TPC = (TPC\text{-}SPC) \times 100 / TPC \qquad (Equation\ 1)$$

2.2.1.6 Total Antioxidant Activity

Total antioxidant activity of PP and its capsules was measured by modified DPPH method. Substances which have antioxidant characteristics cause a color change of DPPH˙ solution, which is determined spectrophotometrically. Since DPPH˙ solution is very sensitive and degradation of DPPH˙ occurs very quickly in the presence of light, it is always stored at dark. The samples were prepared as the same method described in the section of *2.2.1.3 Total phenolic content analysis* and TAA was evaluated by following the steps in our previous study (Cilek et al., 2012).

The sample was accurately weighed as 100 mg and dissolved in 1 ml ethanol:acetic acid:water mixture (50:8:42 v/v). This mixture was agitated using a Vortex (ZX3, VELP Scientifica, Usmate, MB, Italy) for 1 min, and then filtered by using a syringe filter having pore size of 0.45 µm (Gema Medical Filter, Spain). Then samples were diluted. 3.9 ml of 25 ppm DPPH˙ radical solution (2.5 mg DPPH˙/ 100 ml MetOH) and 100 µl of methanol were mixed, and absorption at 517 nm was measured (A_1) by using UV/VIS spectrofotometer T 70 (PG Instruments LTD, UK) using methanol as blank. Diluted samples of 100 µl were mixed with 3.9 ml DPPH˙ radical solution and allowed to wait in the dark at room temperature. After 1 hour, the absorptions of samples and their parallels were detected spectrometrically (A_2).

Different concentrations of DPPH˙ in methanol with the highest concentration of 25 ppm DPPH˙/L of methanol were used to prepare a calibration curve. By using calibration curve, concentrations (C_1 and C_2) were found for A_1 and A_2, and then the results were calculated according to *Equation 2*:

$$AA(mg\ DPPH/g) = \frac{(C_1 - C_2)}{W_{sample}} \times V \times d \qquad \textit{(Equation 2)}$$

In *Equation 2*, C_1 is the concentration of DPPH˙ immediately after the sample and DPPH˙ solution were mixed (ppm), C_2 is the concentration of DPPH˙ 1 h after mixing (ppm), *d* is the dilution rate, *V* is the volume of extract (ml), W_{sample} is the amount of dry sample (mg).

2.2.1.7 Color analysis

Surface color measurements of phenolic powder and capsules were performed by CIE L*, a*, b* color space by using a UV-2450 visible spectrophotometer (Shimadzu Co, Kyoto, Japan) with illuminant type C (2° standard observer) (Luca, Cilek, Hasirci, Sahin, & Sumnu, 2013). The experiments were triplicated. Total color change (ΔE^*) was calculated from *Equation 3* by using barium sulfate ($BaSO_4$) as a standard (L_o*, a_o*, b_o*).

$$\Delta E^* = \sqrt{(L^* - L_o^*)^2 + (a^* - a_o^*)^2 + (b^* - b_o^*)^2} \qquad \textit{(Equation 3)}$$

In *Equation 3*, *L**, *a**, *b** are the color measurements results of sample, and L_o*, a_o*, b_o* are the results of barium sulfate ($BaSO_4$) used as a standard.

2.2.1.8 Optical microscopy analysis

The phenolic powder and capsules were observed by optical microscopy (Zeiss, Primo Vert, Jena, Germany) at different magnifications with a CCD Camera (Microscope Eyepiece Camera, 10MP, Aptina Color). TopView software program (SPECwise, Inc., West Palm Beach, FL) was used to analyze the images. Emulsions were observed upon five-fold dilution with distilled water in order to increase the visuality (Vermeir, Sabatino, Balcaen, Van Ranst, & Van der Meeren, 2014a)

2.2.1.9 Particle size distribution

The particle size distributions of microcapsules were measured in dry form by using a particle size analyzer (Malvern Instruments, Mastersizer 3000 hydro, Worcestershire, UK) as a type of laser diffraction method. Hydro EV flexible volume wet dispersion unit was filled with distilled water as dispersant for microcapsule particle size measurements (Figure 12).

Figure 12 Particle size analyzer (Malvern Instruments, Mastersizer 3000 hydro, Worcestershire, UK).

The size distributions of emulsion droplets were also analyzed by the same particle size analyzer. A few drops of PE were brought into the Hydro EV flexible volume wet

dispersion unit filled with sunflower oil as dispersant and it was filled with distilled water for DE measurements.

The experiments were triplicated. Sauter mean diameter, representing a surface average diameter, namely D_{32} ($\sum n_i d_i^3 / \sum n_i d_i^2$) and volume-weighted mean diameter, namely D_{43} ($\sum n_i d_i^4 / \sum n_i d_i^3$), representing a volume average diameter, were used to compare the mean diameters of the particles or droplets. Span and median diameter (D_{50}) of samples were also measured by the instrument. The 'span' or width of the size distribution can also be calculated with *Equation 4* (Elversson, Millqvist-Fureby, Alderborn, & Elofsson, 2003):

$$Span = (D_{90} - D_{10}) / D_{50} \qquad \textit{(Equation 4)}$$

In *Equation 4*, D_{90}, D_{10}, and D_{50} represent the diameters at 90, 10, and 50% of the total volume, respectively.

2.2.2 Microcapsules Obtained from Olive Leaf Extract and Their Characterization

Since olive leaf extract was used an extract itself, no extraction as in the case of strawberry pomace was conducted.

2.2.2.1 Preparation of microcapsules of OLE

Flour mixtures were used as coating materials. Chickpea or lentil flour of 10 g was weighed and mixed with distilled water (90 g) for 3 min at 9 500 rpm using a high-speed homogenizer (IKA T25 digital Ultra-Turrax, IKA Works Co, Malaysia). If

chickpea flour was used, coating was named as CHI_{10}, and if lentil flour was used, coating was named as LEN_{10}.

OLE and coating solution were mixed with each other to obtain the core to coating ratio of 1:20 in mass fraction (1 g OLE : 20 g coating solution). The suspensions were homogenized using a HSH, and then, selected suspensions were treated by a HPH to analyze the effect of high-speed and high-pressure homogenization on encapsulation efficiency. HSH was performed at 15 000 rpm for 30 s (IKA T25 digital Ultra-Turrax, Selangor, Malaysia). A batch type HPH was used at 70 MPa with 5 process cycles (Nano Disperser, NLM 100, South Korea). Then, suspensions were freeze-dried at -52 ^{o}C for 48 h below 7.5 Pa (Alpha 1-2 LD plus; Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany). Finally, dry samples were manually ground between 2 pieces of aluminum foils by using a roller to obtain microcapsules in homogenous size. Each experiment was duplicated.

2.2.2.2 Total Phenolic Content Analysis of Microcapsules of OLE

TPC of OLE, microcapsules and coating materials were determined by the same method described in the section of *2.2.1.3 Total phenolic content analysis*.

2.2.2.3 Surface Phenolic Content Analysis of Microcapsules of OLE

The same method described in the section of *2.1.1.4* was applied to microcapsules of OLE to determine SPC results.

2.2.2.4 Encapsulation Efficiency of Microcapsules of OLE

The phenolic contents of lentil flour and chickpea flour mixtures were determined as "$PC_{coating}$". The difference between "$TPC_{capsule}$" and "$PC_{coating}$" gave the corrected TPC result (TPC_{corr}). The difference between corrected *TPC* and *SPC* was defined as

the encapsulated phenolic content (*EPC*) inside microcapsules, which was used to calculate the encapsulation efficiency (*EE*) of microcapsules according to *Equation 5:*

$$EE\ (\%) = \left(\frac{TPCcorr - SPC}{TPCcorr}\right) \times 100\ = \left(\frac{TPCcapsule - PCcoating - SPC}{TPCcapsule - PCcoating}\right) \times 100$$

(Equation 5)

2.2.2.5 Total Antioxidant Activity

TAA of OLE and its capsules were performed as described in the section of *2.2.1.6*.

2.2.2.6 Color Analysis

Surface color measurements of OLE and its capsules were performed as described in the section of *2.2.1.7*.

2.2.2.7 Optical Microscopy Analysis

Optical microscopy analysis of OLE and its capsules were performed as described in the section of *2.2.1.8*.

2.2.2.8 Particle Size Analysis

Particle size measurements of OLE and its capsules were performed as described in the section of *2.2.1.9*.

2.2.2.9 Preparation of Cakes for Utilization of Encapsulated Products

Cake batter formulation contained 100 % wheat flour, 25 % margarine, 12 % milk powder, 9 % egg powder, 3 % salt, 5 %baking powder, and 90 % water (Sakiyan et al., 2011). Cake batters with no sugar were prepared for the determination of the retention of total phenolic content and antioxidant activity of capsules and powders after baking, since sugar has a negative effect on the analysis of phenolic content (Waterhouse, 2002). To prepare cake batters, dry ingredients were mixed with by a mixer at 85 rpm for 1 min (Kitchen Aid, 5K45SS; Benton Harbor, MI, USA). Then, molten margarine and distilled water were added, and mixing continued at 140 rpm for 2 min followed by mixing at 85 rpm for 1 min to obtain a smooth mix. Control cake was obtained from this batter. 800 mg of OLE or encapsulated OLE was added as dry ingredients. One hundred gram of batter was then weighed in 250 ml beakers. Four samples were placed in the middle of a preheated conventional oven (Arçelik 9411 FT; Arçelik, Istanbul, Turkey) adjusted to 175 °C for 21 min (Sakiyan et al., 2011). At the end of baking, cake samples were removed from the pans and cooled for 1 h at room temperature.

Baking Stability:

Baking stability of capsules were investigated during baking. In order to extract phenolic compounds from the crumb of cake, 20 ml of solvent, ethanol:acetic acid:water (50:8:42), was added to 10 g of crumb. Then, the sample was crushed manually using a glass rod. Crushed crumb dispersed in the solvent was then ultrasonicated (160 W, 50 % pulse) for 2 min. After the first cycle, sample was manually agitated. Then, 7.5 ml of dispersion was centrifuged at 10 000 rpm for 2 min. Liquid part was collected and filtered through 0.45 μm filter (Luca et al., 2014). Procedure described in the section of *2.2.1.3* was used to determine TPC of each cake. Retention of phenolic content remained after baking, which shows the baking stability, was expressed as the percent of phenolic content remained in the cake after baking.

Digestions inside simulated body fluids were also investigated:

In vitro release tests of phenolic compounds from uncoated and encapsulated OLE were performed by using simulated gastric fluid (SGF) and simulated intestinal fluid (SIF) prepared according to US Pharmacopoeia (2000). Release of phenolic compounds in SGF or SIF was carried out in 15 ml centrifuge tubes. Each tube contained 500 mg of sample to which 7.0 ml of SGF or 12.0 ml SIF was added. Tubes containing SGF were incubated in a shaking water bath for 2 h at 37 °C with continuous shaking at 80 rpm. At the end of incubation, samples were immediately filtered through 0.45 μm filter and neutralized by the addition of 2 M NaOH solution. Samples dispersed in SIF were incubated for 2 h at 37 °C in water bath, then filtered through 0.45 μm filter and mixed with 3 M HCl solution in order to stop the (Zheng et al. 2010). After 15 min, the samples were neutralized by the addition of 2 M NaOH. Phenolic compounds released in the simulated fluids were determined by Folin-Ciocalteu spectrophotometric method. Results of the tests were expressed as the percent phenolic content remained after baking.

2.2.3 Encapsulation of Olive Leaf Extract Through Double Emulsions and Their Characterization

For olive leaf extract, double emulsions were used as an alternative encapsulation method to powder microcapsules.

2.2.3.1 Preparation of Primary Emulsion

In this study, water-in-oil (W1/O) type of emulsion was prepared for primary emulsion (PE) because OLE is also soluble in water.

In order to produce W1, OLE (5 g / 100 g W1) and NaCl (0.6 g / 100 g W1) were mixed with distilled water by using a high-speed homogenizer at 10 000 rpm for 1 min (IKA T25 Digital Ultra-Turrax, Selangor, Malaysia). The oil phase (O), consisted

PGPR (2.5 g / 100 g O), lecithin (2.5 g / 100 g O) and sunflower oil (95 g / 100 g O), were mixed at 10 000 rpm for 1 min (IKA T25 Digital Ultra-Turrax, Selangor, Malaysia). Then, the oil phase was kept in water bath (GFL 1086, Burgwedel, Germany) at 50 °C for 15 min. To obtain PE, 60 g oil phase and 40 g W1 were mixed by using a high-speed homogenizer at 15 000 rpm for 5 min.

2.2.3.2 Preparation of Double Emulsions

Flour mixtures were used in outer aqueous phase of double emulsions (W2). After weighing chickpea or lentil flour (20 g flour / 100 g W2), NaCl (0.6 g / 100 g W2) was added and they were mixed with distilled water (79.94 g / 100 g W2) for 30 sec at 9 500 rpm using a high-speed homogenizer (IKA T25 digital Ultra-Turrax, IKA Works Co, Malaysia). Flour mixtures were carefully prepared in order to denature the protein found in flour. The pH of the mixture was adjusted to 12 by adding 2M NaOH solution (Badifu & Akubor, 2001; Emami, Tabil, Tyler, & Crerar, 2007). Flour containing coating materials were obtained after heating the mixtures to 75 ^{o}C while being mixed with magnetic stirrer (MR 3001K, Heidolph Instruments GmbH & Co, Schwabach, Germany) (Giancone, Torrieri, Masi, & Michon, 2009). If chickpea flour was used, coating was named as CHI_{20}, and if lentil flour was used, coating was named as LEN_{20}.

W1/O/W2 type DEs were prepared according to the two-step emulsification method (Figure 13) (Altuntas et al., 2017; Fechner, Knoth, Scherze, & Muschiolik, 2007). The first step was to produce PE. The second step was to mix the PE with W2. In order to produce DE, W2 (60 g / 100 g DE) and PE (40 g / 100 g DE) were mixed gently with magnetic stirrer (MR 3001 K, Heidolph Instruments GmbH & Co, Schwabach, Germany) at 1 000 rpm for 15 min. If chickpea flour was used in W2, DE was named as DE-CHI_{20}. If lentil flour was used, DE was named as DE-LEN_{20}.

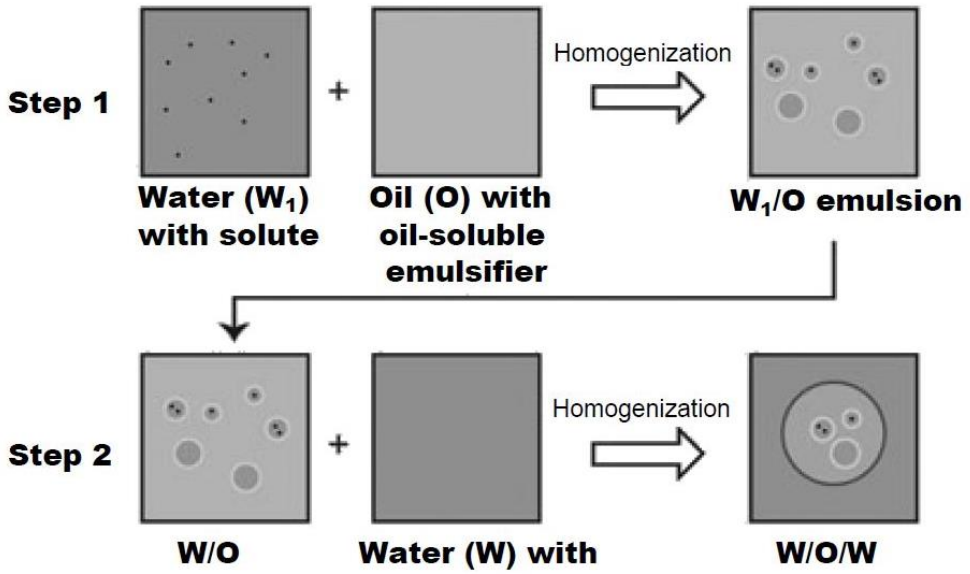

Figure 13 Two-step emulsification technique to prepare double emulsion.

2.2.3.3 Determination of the Type of Emulsions

Dilution technique was applied to find if the emulsion is oil-in-water (O/W) or water-in-oil (W/O) (Vermeir, Sabatino, Balcaen, Van Ranst, & Van der Meeren, 2014b). This technique is a quick method that depends on the miscibility of the outer phase in oil or in water. A few drops of the emulsion were spattered into two beakers, one containing sunflower oil and the other containing distilled water. If the emulsion was miscible in oil, this showed that the emulsion was W/O because easy dissolution in oil could be performed when continuous phase was oil.

2.2.3.4 Stability of double emulsions

Stability of emulsions was evaluated by two different methods; namely, instant stability (sedimentation rate) and storage stability (phase separation rate). Two independent runs were carried out for each emulsion.

2.2.3.4.1 Instant Stability

The emulsions were centrifuged at 3 000 rpm for 30 min immediately after preparation, and the heights of the separated phases were determined (Altuntas et al., 2017). Instant stability (IS) was calculated according to *Equation 6:*

$$IS\ (\%) = (h_c / h_o) \times 100 \qquad \textit{(Equation 6)}$$

In *Equation 6,* h_c is the height of the upper part of the centrifuged emulsion (cm), and h_o is the height of the sample before centrifugation (cm).

2.2.3.4.2 Storage Stability

Emulsions were stored in cylindrical glass tubes of 15 ml at room temperature (25 °C). The separated phase of emulsions was analyzed at various intervals over 120 days. Storage stability (SS) was calculated as can be seen in *Equation 7:*

$$SS\ (\%) = (h_c/h_o) \times 100 \qquad \textit{(Equation 7)}$$

In *Equation 7,* h_c is the height of the upper (opaque) part in cm, and h_o is the initial height of the emulsion in cm.

2.2.3.4.3 Utilization of Double Emulsions in Cakes

Cake batter formulation changed while using PE or DE as an ingredient because they contain water and oil components initially.

Preparation of cake with PE:

100 % wheat flour, 12 % milk powder, 9 % egg powder, 2.9 % salt, 5 % baking powder were mixed with by a mixer at 85 rpm for 1 min (Kitchen Aid,5K45SS; Benton Harbor, MI, USA). Then, 75 g distilled water was added, and mixing continued at 140 rpm for 2 min. Later, 40 g PE was added. Finally, the batter was mixed for 1 min at 85 rpm to obtain a smooth mix. Cake batters with no sugar were prepared for the determination of the retention of total phenolic content and antioxidant activity of capsules and powders after baking, since sugar has a negative effect on the analysis of phenolic content (Waterhouse, 2002).

Preparation of cake with DE:

To prepare cake batters with DE, 88 % wheat flour, 12 % milk powder, 9 % egg powder, 2.5 % salt, 5 % baking powder were mixed with by a mixer at 85 rpm for 1 min (Kitchen Aid,5K45SS; Benton Harbor, MI, USA). Then, 28 g distilled water was added, and mixing continued at 140 rpm for 2 min. Later, 100 g DE was added. Finally, the batter was mixed for 1 min at 85 rpm to obtain a smooth mix.

After obtaining cake batters, baking and related analysis were performed as described in the section of *2.2.2.9.*

2.2.3.5 Optical Microscopy Analysis

Optical microscopy analysis of emulsions was performed as described in the section of *2.2.1.8.*

2.2.3.6 Rheological Measurements

The rheological behavior of emulsions was characterized using a dynamic shear rheometer (Kinexus-Lab+, Malvern Instruments, Worcestershire, UK) fitted with a cup and bob measurement cell (Cup ID 37.0 mm; Bob OD 33.64 mm) held at 25 ± 0.1 °C. Shear rate varied from 0.1 to 100 s^{-1}. The apparent viscosities were calculated by using Kinexus rSpace software with shear stress and shear rate data.

Flow behavior of emulsions was also evaluated by fitting shear stress (τ) and shear rate ($\dot{\gamma}$) data to Power law model (*Equation 8*):

$$\tau = K \dot{\gamma}^{n} \qquad \textit{(Equation 8)}$$

In *Equation 8*, τ is the shear stress (Pa), K is the consistency coefficient (Pa.s), $\dot{\gamma}$ is the shear rate (1/s) and n is the flow behavior index (dimensionless).

2.2.3.7 Particle Size Distribution

The size distributions of emulsion droplets were also analyzed by the particle size analyzer. A few drops of PE were brought into the Hydro EV flexible volume wet dispersion unit filled with sunflower oil as dispersant and it was filled with distilled water for DE measurements.

The experiments were performed as described previously in the section of *2.2.1.9*.

2.2.3.8 OLE Release and Kinetics in Emulsions

OLE is soluble inside aqueous phase of PE. In order to calculate total phenolic content (TPC) of PE and DE, emulsions must be totally disrupted by centrifugation at 4 000 rpm for 20 min to reach aqueous phase. The supernatant was removed because it was

oil phase. The bottom liquid was analyzed to calculate TPC by Folin-Ciocalteau method (Cilek et al., 2012). The release rate of OLE was calculated by analyzing TPC of PE and DE. The results were expressed in terms of gallic acid equivalents in milligrams per gram (mg GAE/g).

Release rate of PE was calculated according to *Equation 9:*

$$RR\ PE\ (\%) = \left(1 - \frac{PE\ TPC}{OLE\ TPC}\right) \times 100 \qquad \textit{(Equation 9)}$$

In *Equation* 9, *PE TPC* and *OLE TPC* are the TPC results of PE and OLE in mg GAE/g respectively.

To calculate release rate of DE, TPC of W2 must be calculated before and after DE formation because lentil and chickpea flours have their own initial phenolic contents. Before DE formation, immediately after W2 preparation (Section *2.2.3.2 Preparation of double emulsions*), it was diluted, filtered by using syringe filter (0.45µm) and TPC of W2 result was recorded as '$W2_{Before}$'. After DE formation, DE was diluted by dropwise addition of distilled water while gently mixing with magnetic stirrer at 200 rpm. Centrifugation at 2 000 rpm for 2 min was applied to separate W2 without disrupting PE. The supernatant was removed, and the remaining emulsion was centrifuged again at 2 000 rpm for 2 min. The supernatant was removed again, and the bottom liquid was analyzed to calculate TPC. The flow chart of the procedure can be seen in Figure 14. The result was recorded as '$W2_{After}$'. Corrected TPC of W2 was calculated according to the *Equation 10:*

$$W2_{TPC} = W2_{After} - W2_{Before} \qquad \textit{(Equation 10)}$$

In *Equation 10, $W2_{TPC}$* is the corrected phenolic content of W2 phase. Besides, $W2_{After}$ and $W2_{Before}$ are the TPC results of W2, after and before DE formation in mg GAE/g respectively.

TPC was determined over 30 days of storage at given times (1, 2, 3, 4, 7, 11, 15 and 30 days) as explained above. Release rate of DE was calculated according to the *Equation 11:*

$$RR\ DE\ (\%) = \frac{W2\ TPC}{DE\ TPC} \times 100 \qquad \textit{(Equation 11)}$$

In *Equation 11,* $W2_{TPC}$ and DE_{TPC} are the TPC results of W2 and DE in mg GAE/g respectively.

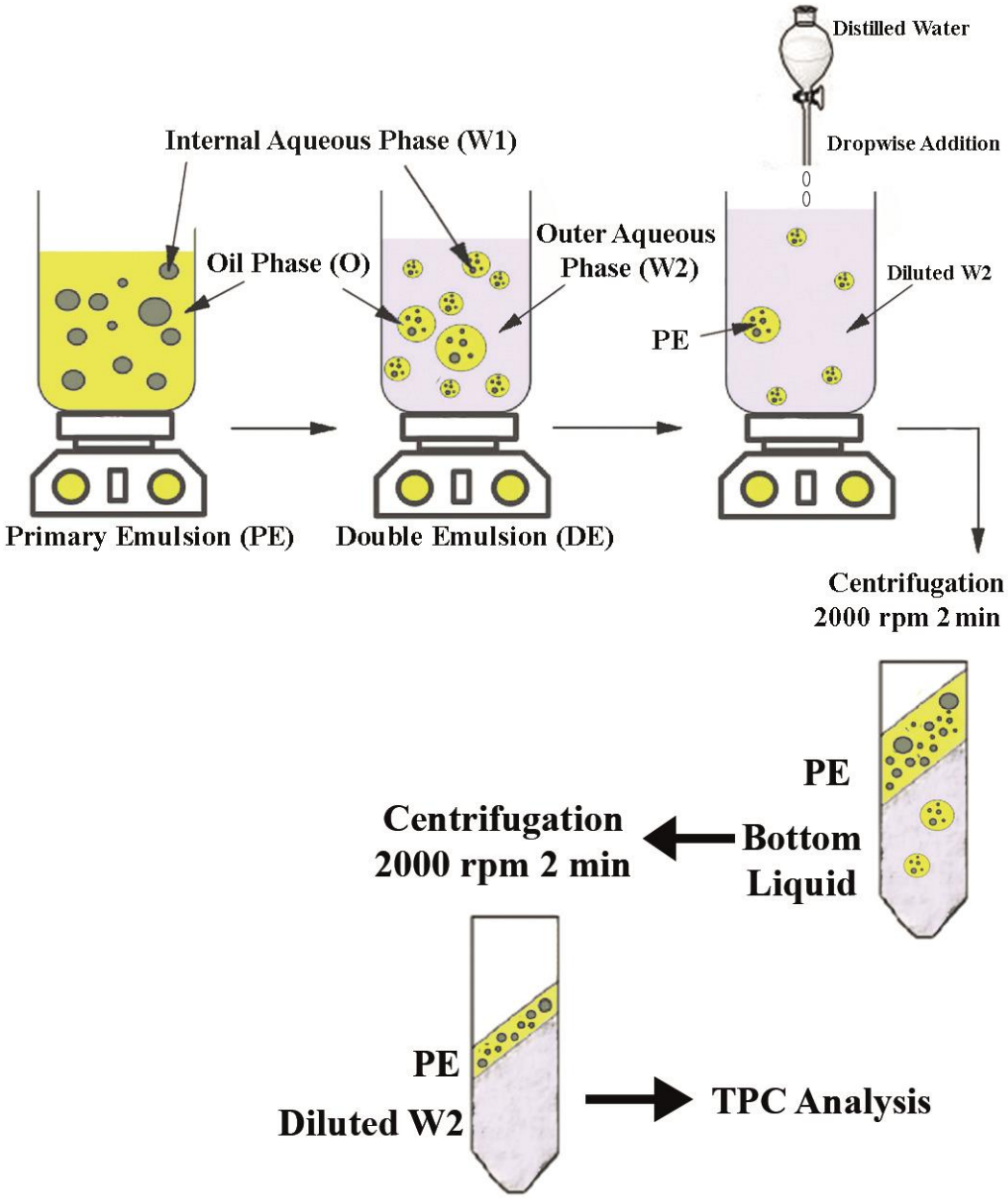

Figure 14 Flow chart of double emulsion preparation.

2.2.3.9 Encapsulation Efficiency

Encapsulation efficiency (EE) was defined as the percentage of remaining OLE still entrapped within W1 *(Equation 12).*

$$EE\ (\%) = 100 - RR\ (\%) \qquad (Equation\ 12)$$

In *Equation 12*, *EE* is encapsulation efficiency (%) and *RR* is release rate (%).

2.2.3.10 Total Antioxidant Activity

Total antioxidant activity (TAA) of OLE and emulsions at any specific period of storage was measured by modified DPPH method. The samples were prepared as the same method described in the section of *2.2.3.8 OLE release and kinetics in emulsions* and TAA was evaluated by following the steps in our previous study (Cilek et al., 2012) as described in the section of *2.2.1.6*.

Loss of TAA (%) during storage was expressed as the fraction of TAA lost relative to TAA of OLE or emulsions.

2.2.3.11 Modelling OLE Release Kinetics

The release characteristics of OLE in DEs during 30-day storage was described by the first-order kinetics *(Equation 13).*

$$C_a = C_{ao}\, e^{-kt} \qquad (Equation\ 13)$$

In *Equation 13*, C_a is the TPC of DEs, C_{ao} is the TPC at the first of day of storage, k is the release rate constant (day^{-1}) and t is the storage time (day). Linear fitting was applied to $\log_e$ of the data to find the release rate constants of DE-CHI_{20} and DE-LEN_{20}. The results were also given in terms of $W2_{TPC}$ (corrected W2) and loss of total antioxidant activity (loss of TAA) results.

2.2.3.12 *In Vitro* Release Tests

The same method was applied to cakes with PE or DE in order to measure the released amount of phenolic content in simulated gastric fluid and simulated intestinal fluid (section *2.2.2.11*).

2.2.3.13 NMR relaxometry experiments

Time domain nuclear magnetic resonance (TD-NMR) relaxometry experiments were carried out in a 0.5 T Benchtop NMR low resolution system (Spin Track, Russia) operating at a frequency of 20.34 MHz. Relaxation period (TR) and observation time were chosen as 1 500 ms and 3 s respectively in saturation recovery sequence for T_1 measurements. T_2 measurements were conducted using Carr-Purcell-Meiboom-Gill (CPMG) sequence with 200 ms echo time, 3 000 echoes for DEs and 6 000 echoes for PEs. 4 scans were applied for both T_1 and T_2 measurements. The results were given as the means of five replicates. Mono-exponential fitting was conducted for T_1 relaxation curves by MATLAB. T_2 is characterized by a time constant of an exponentially decaying curve which is obtained after CPMG sequence. T_2 results were evaluated both by mono-exponential and bi-exponential fittings (T_{2a} and T_{2b}). Representative graphs of T_1 and T_2 signals are given in Figure 15 and Figure 16 respectively.

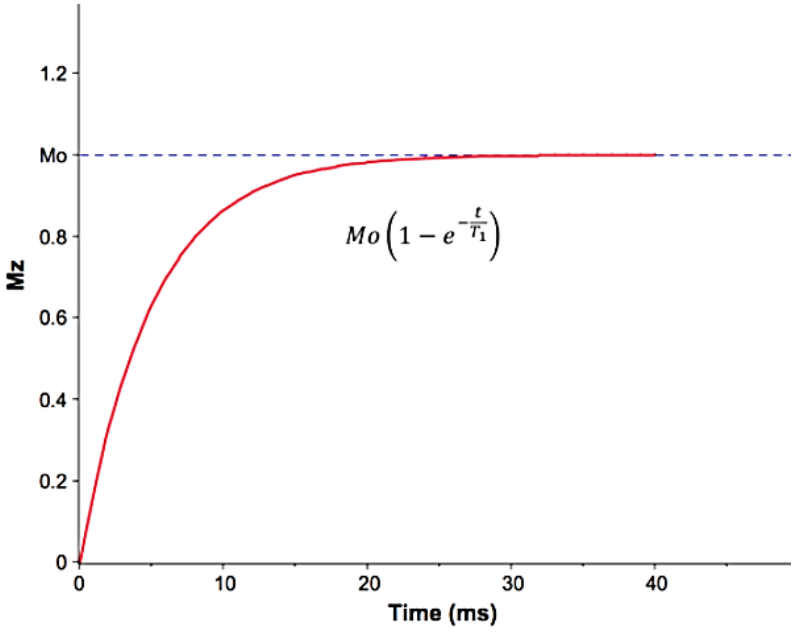

Figure 15 A representative T_1 curve obtained through Saturation Recovery Sequence.

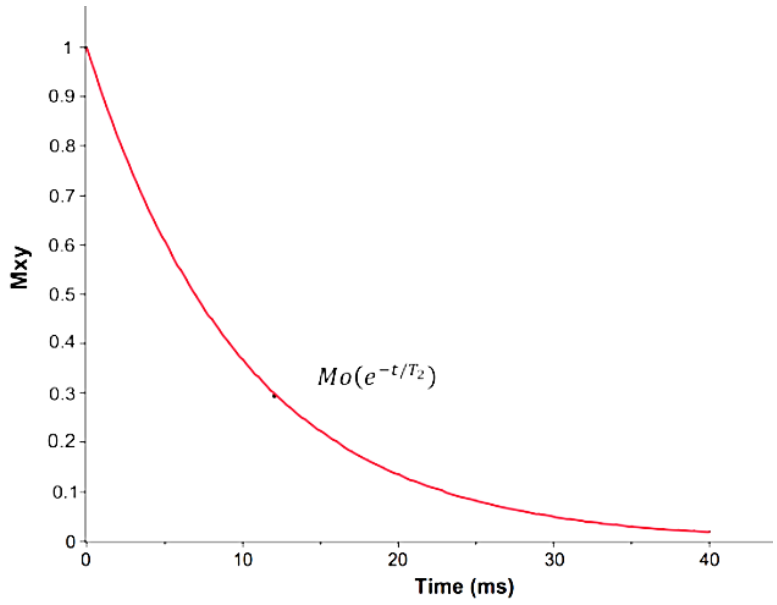

Figure 16 A representative T_2 curve obtained through CPMG sequence.

2.3 Overall Experimental Design

To summarize the parameters used and levels studied, and responses measured, an overall experimental design table is given in Table 3.

Table 3 Overall experimental design table with variables.

Microcapsules obtained from strawberry pomace	Encapsulation Technique • HSH • HPH Coating Material • MD:GA 10:0 • MD:GA 8:2 • MD:GA 6:4 HPH Pressure • 70 MPa • 50 MPa HPH Process Cycle • 3 passes • 5 passes • 7 passes	• Total phenolic content analysis • Surface phenolic content analysis • Encapsulation efficiency • Total antioxidant activity • Color analysis • Optical microscopy analysis • Particle size distribution
Microcapsules obtained from olive leaf extract	Encapsulation Technique • HSH • HPH Coating Material • Lentil flour (10%) • Chickpea flour (10%)	• Total phenolic content analysis • Surface phenolic content analysis • Encapsulation efficiency • Total antioxidant activity • Color analysis • Optical microscopy analysis • Particle size distribution • Baking stability • *In vitro* release tests

Table 3 Continued

Encapsulation of olive leaf extract through emulsification methods	Encapsulation Technique • Primary Emulsion • Double Emulsion Coating Material • Lentil flour (20%) • Chickpea flour (20%)	• Total phenolic content analysis • Surface phenolic content analysis • Encapsulation efficiency • Total antioxidant activity • Color analysis • Optical microscopy analysis • Particle size distribution • Baking stability • *In vitro* release tests • Instant Stability • Storage Stability • Release rate during storage • Modelling of kinetics • Rheological Measurements • NMR Relaxometry

2.4 Statistical Analysis

The measured data were expressed as means ± SD. Statistical analysis of data was performed to determine the differences between treatments through analysis of variance (ANOVA) using SAS software (SAS software version 9.1, SAS Institute Inc., NC, USA). If a significant difference was found, Duncan's Multiple Comparison Test

was used to compare the results at $p \leq 0.05$ (Appendix B). All the results represented the means of at least two replications. Correlations between results were determined by using Minitab software 16.1.1 (Minitab Inc., PA, USA).

CHAPTER 3

RESULTS AND DISCUSSION

3.1 Microcapsules Obtained from Strawberry Pomace and Their Characterization

In this study, extracted strawberry phenolic powder (PP) was encapsulated with different combinations of MD and GA. MD was used in all coating types because of its ability to increase encapsulation stability (Sanchez et al., 2013). GA is also known as a good encapsulating agent; therefore, it increased EE by increasing interconnected structure of the coating material matrix. This structure resulted in a better environment for encapsulation of phenolic compounds. A recent research showed that using GA caused higher efficiencies (Elez Garofulić, Zorić, Pedisić, & Dragović-Uzelac, 2017).

3.1.1 TPC, SPC and Encapsulation Efficiency of Microcapsules of PP

To calculate encapsulation efficiency (EE) of microcapsules, it was necessary to determine TPC and SPC. Deterioration of polyphenols during processing or presence of high amount of phenolic content on the surface of the capsules can cause a reduction in EE. TPC of PP was found as 225.12 mg GAE/g, and TPC of capsules were found between 68.15 and 79.29 mg GAE/g (Table 4).

In this study, all the samples were treated by high-speed homogenizer (HSH). After using HSH, high-pressure homogenizer (HPH) was used for the selected samples to see the effect of encapsulation method on encapsulation, because phenolic contents of fruit juices were shown to be influenced by HPH (He et al., 2016). Determining the required level of pressure, the duration of applied pressure (process cycle), and the

temperature inside the chamber are three important issues when running a HPH. Increasing pressure or number of cycles may aid to obtain a more homogenous dispersion and higher encapsulation efficiency. However, HPH may cause a high consumption of energy, and a temperature increase in the product (Bot et al., 2018; Håkansson, 2018). High temperatures may damage the phenolic compounds found in the dispersion. Therefore, it is necessary to find an optimum condition to perform a good encapsulation for the phenolic compounds.

In order to determine the number of process cycles during HPH treatment, recent studies were carefully analyzed. In a recent study, encapsulation was performed by HPH at 101 MPa for 1 pass (Altuntas, Sumnu, & Sahin, 2016), and the data showed that HPH was not an effective way of encapsulation due to low pressure and insufficient number of cycles. Therefore, 3, 5 and 7 cycles were chosen for HPH.

Table 4 shows the TPC and SPC results of capsules prepared by different methods. Statistical results are given in Appendix B. TPC results showed that applying 3 or 5 cycles did not damage the phenolic compounds. However, TPC decreased sharply ($p \leq 0.05$) at 7 cycles for all the capsules listed in Table 4 (from 74.95 to 69.13, from 75.63 to 70.71, from 76.38 to 70.64; from 74.34 to 68.15; from 76.12 to 70.07; and from 75.59 to 69.21 mg GAE/g). The loss of TPC showed the deterioration of phenolic compounds, which might be due to high temperatures. It was observed that the higher the cycle number, the higher temperature of the outer wall of the chamber. Phenolic compounds deteriorate as temperature increases, since they are very sensitive to heat and high temperatures (Lou et al., 2014). Therefore, application of 7 process cycles was found to be less efficient than 3 and 5 cycles. In order to determine the optimum process cycle, EE results should also be investigated.

Table 4 Total phenolic content (TPC) and surface phenolic content (SPC) results (mg GAE/g) of microcapsules prepared by different methods (HSH: high-speed homogenizer, HPH: high-pressure homogenizer). *Different letters within rows represent significant difference at $p < 0.05$.

MD:GA	HSH		HPH 50 MPa						HPH 70 MPa					
			3 cycles		5 cycles		7 cycles		3 cycles		5 cycles		7 cycles	
	TPC	SPC	TPC	SPC	TPC	SPC	TPC	SPC	TPC	SPC	TPC	SPC	TPC	SPC
10:0	75.79^{a*}	13.94^{c}	76.20^{a}	15.91^{c}	74.95^{a}	10.50^{d}	69.13^{b}	9.60^{e}	75.72^{a}	13.72^{c}	74.34^{a}	9.43^{e}	68.15^{b}	7.84^{f}
8:2	75.66^{a}	12.25^{d}	77.63^{a}	11.51^{d}	75.63^{a}	7.34^{ef}	70.71^{b}	8.38^{e}	79.29^{a}	14.16^{c}	76.12^{a}	7.26^{ef}	70.07^{b}	7.12^{f}
6:4	74.82^{a}	8.86^{dc}	76.42^{a}	9.64^{c}	76.38^{a}	6.40^{d}	70.64^{b}	9.02^{c}	77.41^{a}	8.52^{dc}	75.59^{a}	5.80^{d}	69.21^{b}	7.95^{dc}

EE results showing the effects of different methods, different coating material types, different pressures, and different process cycles were given in Figure 17. There was significant ($p \leq 0.05$) difference between EE of samples with MD:GA 10:0 and 6:4 after HSH treatment (Appendix B, Table B.2). It was seen that increasing gum concentration resulted in higher efficiencies except the treatment of HPH 7 cycles (Figure 17). This can be due to the higher emulsifying capacity of GA and its synergistic effect with MD at higher concentrations. MD:GA 6:4 gave the highest EE when 70 MPa 5 cycles treatment was applied. Therefore, MD:GA ratio of 6:4 was chosen as the best coating material for the encapsulation of PP.

There was no difference between EE results of capsules with the same coating material type between HSH and HPH 70 MPa / 3 cycles. Besides, when HSH was compared to HPH 50 MPa / 3 cycles, EE results of MD:GA 10:0 and 6:4 were not significantly different (Figure 17) ($p > 0.05$). Therefore, there was no need to apply HPH 3 cycles treatment because it was not an efficient way when compared to HSH. However, the same situation was not valid for HPH 5 and 7 cycles (Figure 17). Increasing HPH cycle from 3 to 5 resulted in a significant positive effect on EE. The results of EE of capsules with MD:GA 10:0 increased from 79.12 to 85.99 % at 50 MPa, and from 81.88 to 87.32 % at 70 MPa. EE of capsules with MD:GA 6:4 increased from 87.39 to 91.62 % at 50 MPa, and from 88.99 to 92.33 % at 70 MPa. When the number of process cycles was increased to 7, there was no significant difference between EE results of capsules with MD:GA 10:0 ($p > 0.05$). The results of EE of capsules with MD:GA 10:0 prepared by HPH 50 MPa were 85.99 and 86.11 % with 5 and 7 cycles, respectively. In the same manner, EE results of capsules with MD:GA 10:0 prepared by HPH 70 MPa were 87.32 and 88.50 % with 5 and 7 cycles, respectively. The same trend can be seen in Figure 17 on the capsules with MD:GA 8:2 prepared by HPH 70 MPa with 5 and 7 cycles (90.47 - 89.99 %). On the contrary, for capsules with MD:GA 6:4, increasing number of process cycles to 7 had significantly negative effect on EE ($p \leq 0.05$). This may be due to the significant decrease in TPC when process cycle number was increased to 7 (Table 4) from 76.38 to 70.64 mg GAE/g, and from 75.59 to 69.21

mg GAE/g. Therefore, selection of 5 cycles was found to be the optimum process number. The data also showed that there was no difference between 50 and 70 MPa treatments on the capsules with the same coating materials. Under these circumstances, particle size results were needed to determine the optimum HPH pressure.

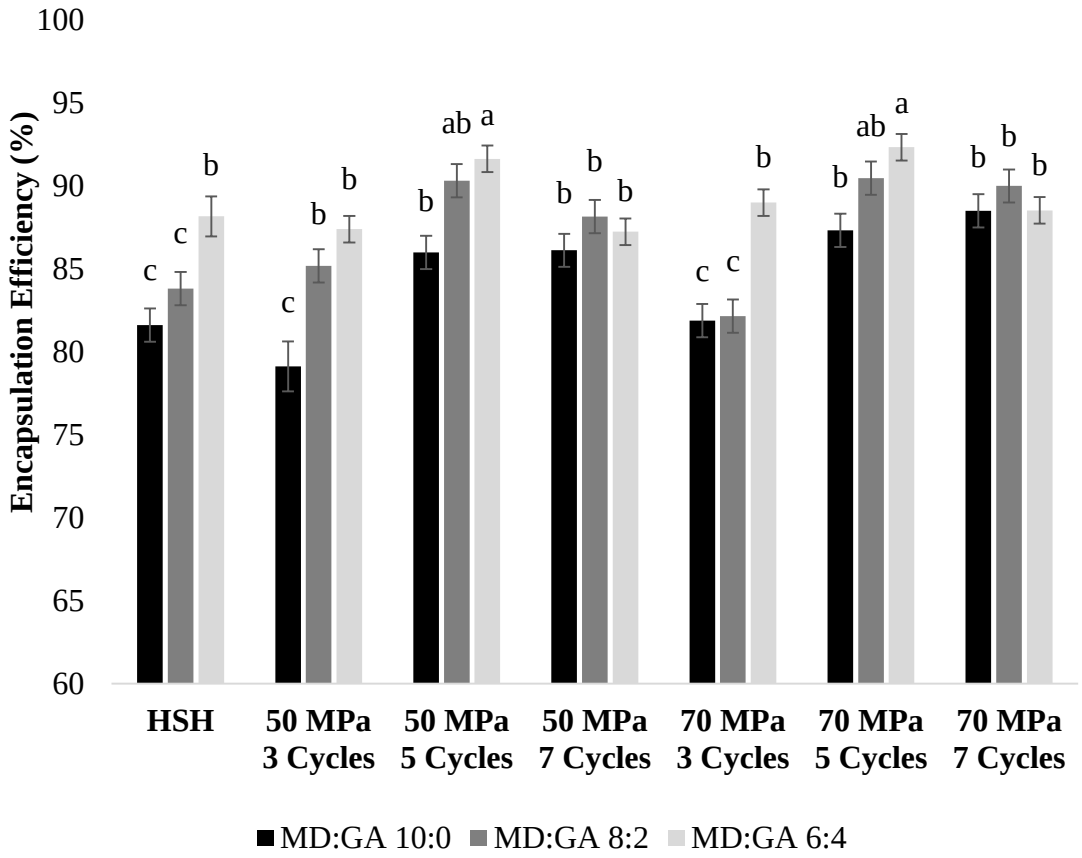

Figure 17 Encapsulation efficiency (EE %) results of microcapsules prepared by high-speed homogenizer (HSH) and high-pressure homogenizer (HPH). *Means followed by the different (a, b, and c) letters are significantly different at $p \leq 0.05$.

3.1.2 Color Measurements of Microcapsules of PP

The color results of PP and microcapsules with different MD:GA ratios prepared by HPH 70 MPa are given in Table 5. When color of PP was compared to color of microcapsules, it was found that HPH had significant effect on total color difference (ΔE*). The effect of different coating materials could also be analyzed for capsules treated with 3 cycles. It was seen that addition of GA caused no significant difference in color ($p > 0.05$). When the capsules treated with 3, 5 and 7 cycles were analyzed, it was seen that L* value decreased significantly when process cycle of HPH was increased from 3 to 5 ($p \leq 0.05$). However, when process cycle was further increased to 7, no significant color change (L*, b*, ΔE*) was observed (Table 5) ($p > 0.05$). This could be explained by perfect mixing of the core and coating materials after 5 process cycles.

Additionally, no significant change was observed between a* values of 3 and 5 cycles ($p > 0.05$) (Table 5). On the other hand, a significant decrease was found between process cycles of 5 and 7 (Table 5). This result was in correlation with TPC results which were given in Table 4. Redness value, a*, is coming from the color of PP. Any decrease in TPC could be seen by color measurements.

Table 5 Effect of high-pressure homogenizer (HPH) process cycle and coating materials on color results of phenolic powder (PP) and microcapsules.

	Pressure	Process Cycle	Coating MD:GA	L*	a*	b*	ΔE*
PP	-	-	-	43.86±0.26^{a**}	28.21±0.31^{a}	11.95±0.32^{b}	63.89±1.21^{b}
HPH	70 MPa	3	10:0	42.17±0.42^{b}	27.96±0.53^{a}	12.66±0.41^{ab}	65.34±0.91^{a}
HPH	70 MPa	3	6:4	42.25±0.21^{ab}	28.05±0.42^{a}	14.29±0.41^{a}	65.71±1.25^{a}
HPH	70 MPa	3	8:2	41.75±0.29^{b}	28.50±0.25^{a}	13.92±0.42^{a}	66.41±1.22^{a}
HPH	70 MPa	5	8:2	39.02±0.42^{c}	27.93±0.61^{a}	12.86±0.36^{ab}	67.93±1.13^{a}
HPH	70 MPa	7	8:2	39.95±0.47^{c}	25.12±0.42^{b}	13.95±0.42^{a}	66.82±1.02^{a}

**Means followed by the different (a, b and c) letters within columns are significantly different at $p \leq 0.05$. Results are given as "means ± SD", n = 3.

3.1.3 Optical Microscopy Analysis of Microcapsules of PP

Microcapsules of PP (70 MPa with 5 cycles) were analyzed by optical microscopy to detect the outer shape of the capsules. Visual observation of capsules showed that microcapsules were in irregular shape and they had rough surfaces (Figure 18). It was also seen that increasing gum concentration resulted in smaller sizes of microcapsules with more uniform distribution (Figure 18).

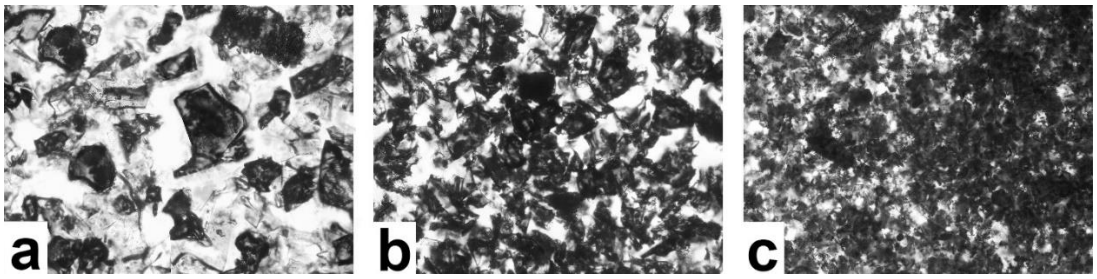

Figure 18 Optical micrographs of microcapsules of phenolic powder with (a) MD:GA 10:0, (b) MD:GA 8:2 and (c) MD:GA 6:4.

3.1.4 Particle Size Analysis of Microcapsules of PP

The results of median diameter of the volume distribution (D_{50}), Sauter mean diameter (D_{32}), and span values were determined to compare the particle size distribution of the samples. Table 6 shows the effects of different parameters of encapsulation techniques on particle size analysis of PP and capsules with MD:GA ratio of 8:2. D_{50}, D_{32}, and span values of PP are 62.67 μm, 16.77 μm and 2.94 respectively (Table 6). It can be seen in Table 6 that application of HPH resulted in significantly smaller capsules when compared to PP ($p \leq 0.05$). The reduction of droplet size of a sample obtained after

application of HPH was also found in other researches (Bouaouina et al., 2006; Tsai, Tseng, & Chen, 2009). In this research, D_{32} and D_{50} of microcapsules with 50 and 70 MPa treatments were analyzed. The results were significantly smaller when 70 MPa pressure was applied ($p \leq 0.05$). Therefore, 70 MPa was chosen as the optimum pressure. This can be explained by the fact that the increase in given energy leads to an extra disruption and formation of smaller particles. Besides, there was no significant difference between 5 and 7 cycles ($p > 0.05$) (Table 6). Therefore, 5 cycles were chosen to obtain low-processed capsules.

Table 6 The effects of different treatments on particle size analysis of phenolic powder (PP) and capsules with MD:GA ratio 8:2 (HPH: high-pressure homogenizer).

Treatment	Pressure	Process Cycle	D_{50} (μm)	D_{32} (μm)	Span
none (PP)	-	-	62.67±2.50^{a*}	16.77±1.29^{a}	2.94±0.09^{c}
HPH	50 MPa	5	37.65±2.28^{b}	4.75±0.15^{b}	1.78±0.02^{d}
HPH	70 MPa	3	4.34±0.32^{c}	2.54±0.36^{c}	8.67±0.36^{b}
HPH	70 MPa	5	3.92±0.14^{c}	2.31±0.18^{c}	11.69±0.35^{a}
HPH	70 MPa	7	4.21±0.67^{c}	2.36±0.17^{c}	11.23± 0.42^{a}

*Means followed by the different (a, b, c, and d) letters within columns are significantly different at $p \leq 0.05$. Results are given as "means ± SD", n = 3.

In Table 6, it can be observed that there was an inverse relation between particle sizes and span values (except PP results). The relation of larger particle sizes with small span values are explained by the formation of uniform droplets in suspension. Besides, the large particles prevent the detection of smaller particles during laser diffraction measurements and cause small span values. This case shifts the particle size distribution curve to the left (to smaller sizes) and can cause higher span values (Table 6). Inverse relation of D_{50} and D_{32} with span values were also shown in a recent study (Lavelli, Sri Harsha, Mariotti, Marinoni, & Cabassi, 2015).

3.2 Microcapsules Obtained from Olive Leaf Extract and Their Characterization

3.2.1 TPC, SPC and Encapsulation Efficiency of Microcapsules of OLE

In this study, OLE was encapsulated with different type of coatings. Chickpea and lentil flours were chosen because of their high nutritional and functional value (Sanchez et al., 2013). It was found that flours can be used as encapsulating agents, because EEs of microcapsules were found between 80.26 and 85.09 % (Table 7). Flours contain protein and carbohydrates, which provide proper materials for encapsulation by forming an interconnected matrix. Polysaccharides and proteins are generally used for their ability to establish polymer interactions and create a continuous network responsible for the encapsulation property and flours contain both polysaccharides and proteins together. This composite structure resulted in a good environment for encapsulation of OLE.

Before calculation of EE of microcapsules, it was necessary to determine the phenolic content of chickpea and lentil flour mixtures, because they inherenetly contain phenolic compounds. The phenolic content of chickpea and lentil flour mixtures ($PC_{coating}$) were found as 2.70 and 4.01 mg GAE/g respectively. These values were used to calculate the encapsulted phenolic contents correctly. They were used as correction factors and the difference between $TPC_{capsule}$ and $PC_{coating}$ was denoted as

TPC_{corr}. The phenolic content of the encapsulated OLE was used in the calculation of EE, which was the difference between TPC_{corr} and SPC. Deterioration of polyphenols during processing or presence of high amount of phenolic content on the surface of the capsules can cause a reduction in EE.

Table 7 shows the $TPC_{capsule}$, $PC_{coating,}$ TPC_{corr}, SPC and EE results of capsules prepared by different methods with different coatings. TPC of OLE was found as 120.67 mg GAE/g, and corrected TPC of capsules (TPC_{corr}) were found between 39.80 and 40.00 mg GAE/g (Table 7). As expected, TPC_{corr} was nearly one third of the TPC result of OLE because the results were reported in 'per g dry weight' and microcapsules contained 1 g of OLE with 2 g of flour in their coatings.

The effects of different methods on EE can also be seen in Table 7. It is found that there was significant ($p \leq 0.05$) difference between EE of capsules prepared by HPH treatment (81.87 and 85.09 %) and there was no difference between EE results of capsules prepared by HSH treatment (80.78 and 80.26 %) (Table 7) (Appendix B). The results of EE of capsules with lentil flour increased from 80.26 to 85.09 % when HPH was applied. This might be due to the lower viscosity of lentil flour mixture. However, EE of capsules with chickpea flour showed no difference when HPH was applied. The effect of high-pressure became significant when a lower viscous coating material was used because inter-molecular bonds were weaker in less viscous samples and therefore applied pressure became more effective. Lentil flour was found to be more efficient coating material as compared to chickpea flour while obtaining dry microcapsules of OLE.

Table 7 Total phenolic content (TPC, mg GAE/g), surface phenolic content (SPC, mg GAE/g), and encapsulation efficiency (EE, %) results of microcapsules prepared by different methods (HSH: high-speed homogenizer, HPH: high-pressure homogenizer) with different coating materials (CHI10: chickpea flour, LEN10: lentil flour).

Coating Material	Method		$TPC_{capsule}$	$PC_{coating}$	TPC_{corr}	SPC	EE (%)
	HSH	HPH					
CHI_{10}	+	-	42.50 ± 0.20	2.70 ± 0.14	39.80 ± 0.06	7.65 ± 0.45	80.78 ± 1.17 **b***
CHI_{10}	+	+	42.41 ± 0.68	2.70 ± 0.14	39.71 ± 0.54	7.20 ± 0.35	81.87 ± 0.64 **b**
LEN_{10}	+	-	44.01 ± 0.13	4.01 ± 0.01	40.00 ± 0.15	7.90 ± 0.13	80.26 ± 0.41 **b**
LEN_{10}	+	+	43.97 ± 1.15	4.01 ± 0.01	39.96 ± 1.16	5.95 ± 0.36	85.09 ± 1.32 **a**

* Values (mean ± SD, n = 3) followed by the different (a and b) letters are significantly different at $p \leq 0.05$.

3.2.2 Color Measurements of Microcapsules of OLE

The color results of OLE and its microcapsules prepared by different methods with different flour types are given in Table 8. Color measurements of OLE and microcapsules showed that OLE had significantly higher a* (redness/greenness) and b* (yellowness/blueness) values. Besides, OLE had the lowest L* value. The significant difference between ΔE* results of samples verified that they had different colors and even could be differentiated by naked eye (Knösel et al., 2009). The color difference was related to the color of coating materials. When color of OLE was compared to the color of capsules, it was found that encapsulation increased lightness (L*) and decreased redness (a*) significantly ($p \leq 0.05$). This could be explained by the light colors of chickpea and lentil flours that were used as coating materials in capsules. The flour mixtures obtained after mixing with distilled water, were not transparent, therefore their usage in coating contributed to important changes in the color, especially significant difference was observed for ΔE* value (Table 8). When Table 8 was analyzed, it was clearly seen that HSH and HPH were significantly effective on L*, b* and ΔE* values. Usage of different flour types had significant effect on color results espacially on L* and ΔE* values. L* values of capsules with lentil flour were significantly higher than capsules with chickpea flour ($p \leq 0.05$). Additionally, different encapsulation techniques had also significant effect on color of microcapsules. This can be explained by mixing of core and coating materials. When encapsulation technique changed, power of mixing force changed, which affected both color and EE results. It was previously shown that EE results were also affected by different encapsulation techniques.

Table 8 Effect of high-speed homogenization (HSH), high-pressure homogenization (HPH) and coating materials on color results of olive leaf extract (OLE) and its capsules.

Sample	Method		L*	a*	b*	ΔE*
	HSH	HPH				
OLE	-	-	45.65 ± 0.34^{d}	8.59 ± 0.11^{a}	29.38 ± 0.33^{a}	62.35 ± 1.34^{a}
LEN_{10}**	+	-	58.77 ± 0.73^{a}	4.25 ± 0.23^{b}	24.04 ± 0.21^{c}	47.88 ± 1.23^{d}
CHI_{10}**	+	-	54.19 ± 0.64^{b}	4.83 ± 0.38^{b}	26.65 ± 0.37^{b}	53.19 ± 1.02^{b}
LEN_{10}	+	+	55.27 ± 0.45^{b}	4.53 ± 0.12^{b}	22.20 ± 0.45^{d}	50.11 ± 0.63^{c}
CHI_{10}	+	+	52.71 ± 0.87^{c}	4.71 ± 0.37^{b}	25.01 ± 0.91^{bc}	53.67 ± 1.17^{b}

*Values (mean ± SD, n = 3) followed by the different letters within columns are significantly different at $p \leq 0.05$. **LEN_{10} represents the microcapsules of OLE with lentil flour (10 %, w/w) and CHI_{10} represents the microcapsules of OLE with chickpea flour (10 %, w/w).

3.2.3 Optical Microscopy Analysis of Microcapsules of OLE

Microcapsules of OLE in dry form were analyzed by optical microscopy to detect the outer shape of the capsules. The results showed that microcapsules were in irregular shape, and they had rough surfaces whereas OLE particles were in spherical-like shape (Figure 19). The capsules obtained after HPH treatment had smaller particle sizes than HSH treatment (Figure 20). In HSH application, a mixer working at high speed is using to homogenize the sample by decreasing the particle sizes. The aim of utilization of HPH is also the same, which is to homogenize the sample and to decrease the particle size. However, in the HPH process, there is high-pressure application, which is more effective than high-speed mixing. HPH causes a shear stress distribution across the

product, which is the responsible for the obtained results. A more energy can be given by high pressure application than by high speed application.

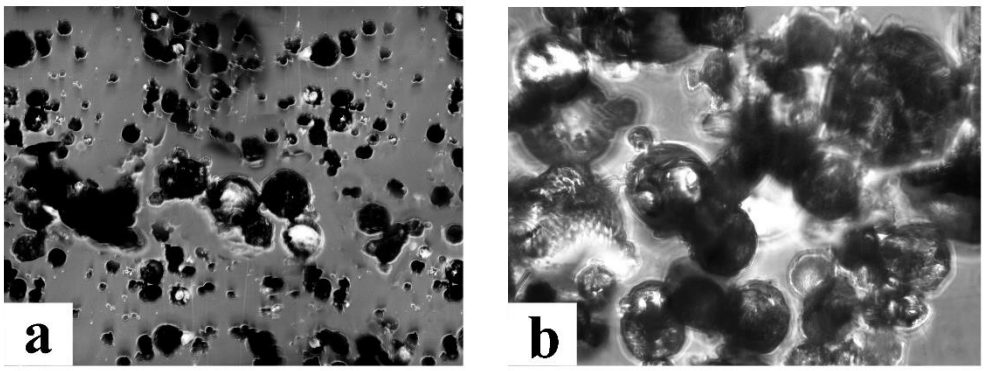

Figure 19 Optical microscopy analysis of OLE in dry form at 20X (a) and 40X (b) magnifications.

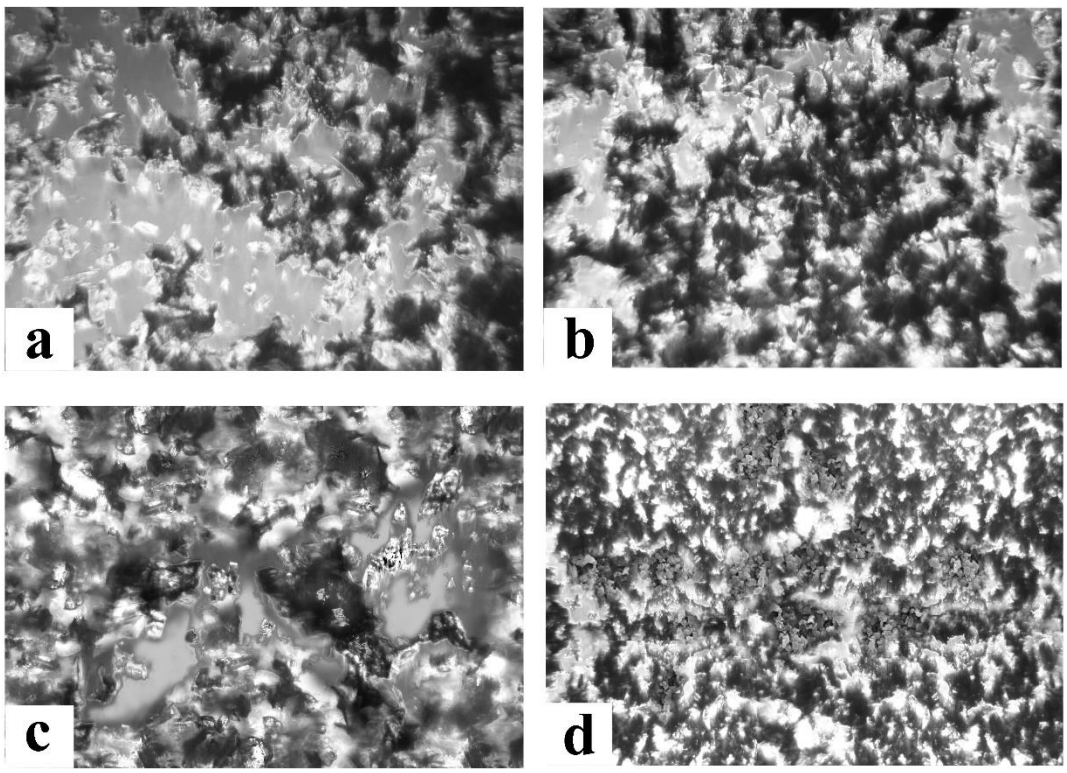

Figure 20 Optical microscopy analysis of microcapsules of OLE in dry form a) high-speed homogenizer, with chickpea flour, b) high-pressure homogenizer, with chickpea flour, c) high-speed homogenizer, with lentil flour, d) high-pressure homogenizer, with lentil flour at 10X magnification.

3.2.4 Particle Size Analysis of Microcapsules of OLE

The results of median diameter of the volume distribution (D_{50}), volume-weighted mean diameter (D_{43}), Sauter mean diameter (D_{32}) and span values were determined to compare the particle size distribution of the samples. Table 9 shows the effects of different encapsulation techniques on particle size analysis of OLE and capsules of OLE with different coatings. D_{50}, D_{32}, and span values of OLE are 18.26 ± 0.42 μm,

5.71 ± 0.10 μm and 5.63 ± 0.16 μm respectively (Table 9). Application of HPH resulted in significantly smaller capsules than OLE ($p \leq 0.05$). The reduction of particle size of a sample obtained after application of HPH was also found in other researches (Bouaouina et al., 2006; Tsai et al., 2009). When different treatments were analyzed, HPH resulted in significant decrease in particle size when compared to HSH. This result can also be seen in optical micrographs (Figure 20).

The results of microcapsules of OLE with chickpea flour were significantly smaller than the microcapsules with lentil flour when HSH was applied, however the results became significantly bigger when HPH was applied. This may be explained by the different protein and starch contents of flours and their interactions. HPH might be more effective on the protein structure because lentil flour had higher protein content than chickpea flour. Decrease in particle size of lentil when HPH was used could be explained by pressure effect on protein size. A study provided information of pressure-induced structural changes on both the molecular and the sub-molecular level of lentil protein (Ahmed, Varshney, & Ramaswamy, 2009). Besides, particles found in lentil flour might have swollen more than chickpea flour during HSH treatment. It was shown that chickpea had lower swelling power than lentil (Chung, Liu, Hoover, Warkentin, & Vandenberg, 2008). Application time of HSH might be insufficient for swelling of chickpea flour. On the contrary, lentil was known to have higher swelling power (Chung et al., 2008) which led a more effective application of HPH on decreasing the sizes of swollen particles.

Additionally, it was seen that span values significantly decreased after application of HPH ($p \leq 0.05$). Microcapsules became more homogenous after HPH treatment (Table 9).

Table 9 The effects of different treatments on particle size analysis of olive leaf extract (OLE) and capsules with different coatings (LEN_{10} represents the microcapsules of OLE with lentil flour (10 %, w/w) and CHI_{10} represents the microcapsules of OLE with chickpea flour (10 %, w/w) prepared by different methods (HSH: high-speed homogenizer, HPH: high-pressure homogenizer). D_{50}, D_{43} and D_{32} are given in μm.

Coating	HSH	HPH	D_{50}	D_{32}	D_{43}	Span
CHI_{10}	+	-	$28.67 \pm 1.48^{b*}$	7.70 ± 0.20^{b}	34.7 ± 0.30^{b}	3.57 ± 0.23^{b}
CHI_{10}	+	+	16.50 ± 0.26^{d}	4.99 ± 0.11^{d}	19.53 ± 0.59^{d}	1.55 ± 0.25^{d}
LEN_{10}	+	-	37.43 ± 0.06^{a}	9.58 ± 0.33^{a}	50.9 ± 0.66^{a}	2.78 ± 0.01^{c}
LEN_{10}	-	+	12.17 ± 1.06^{e}	2.63 ± 0.21^{e}	11.75 ± 0.65^{e}	1.15 ± 0.15^{de}
OLE	-	-	18.26 ± 0.42^{c}	5.71 ± 0.10^{c}	25.32 ± 0.38^{c}	5.63 ± 0.16^{a}

*Values (mean ± SD, n = 3) followed by the different letters within columns are significantly different at $p \leq 0.05$.

3.3 Encapsulation of Olive Leaf Extract through Double Emulsions and Their Characterization

3.3.1 Instant and Storage Stability Analysis

One of the most important physicochemical characteristics of the DEs is their stability, which is the resistance capability to the change of physical properties over the time. The application of DEs can be restricted by the thermodynamic instability in the food industry because instability can lead to leakage of inner aqueous phase (W1) and therefore, core material can go through outer phases. Besides, it can lead to separation of phases and flocculation of the droplets inside the emulsions during process and storage (Benichou, Aserin, & Garti, 2004). From a thermodynamics perspective, DEs are unstable and they can turn into simple emulsions after a certain time. Coagulation, flocculation, or breaking down of the droplets should be considered while designing emulsion systems (Matsumoto, 1986).

Since stability is the main criteria of emulsions, this study evaluated the instant and storage stability of prepared emulsions. Figure 21 demonstrates the instant stability of the emulsions following their storage stability at room temperature (Figure 22). In order to analyze the storage stability, the emulsions were monitored over a period of 120 days, with measurements taken at different days. There was no mold or bacterial growth in DEs; due to presence of OLE. Stability of the emulsions was calculated by measurement of the height of the separated layer. Instant stability results showed that instant stability of PE was 45.06 %. As in literature, DEs protect the valuable core material better than the single ones because of the second layer (Cilek Tatar et al., 2017). According to instant stability (IS) results, the most stable emulsion was found as DE-CHI_{20} with the result of 48.24 % (Figure 21). It might be related to the high viscosity of CHI_{20} emulsions. IS analysis was performed by applying centrifugal forces. When a material had higher viscosity, separation of layers became more difficult as compared to a less viscous material. Thus, emulsions with higher viscosity were more stable.

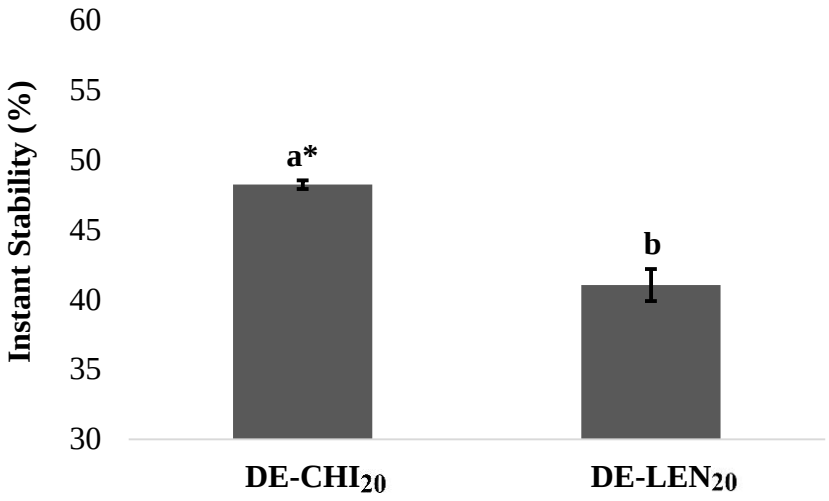

Figure 21 Instant stability results of double emulsions. *Bars with different letters represent the significant difference at $p \leq 0.05$.

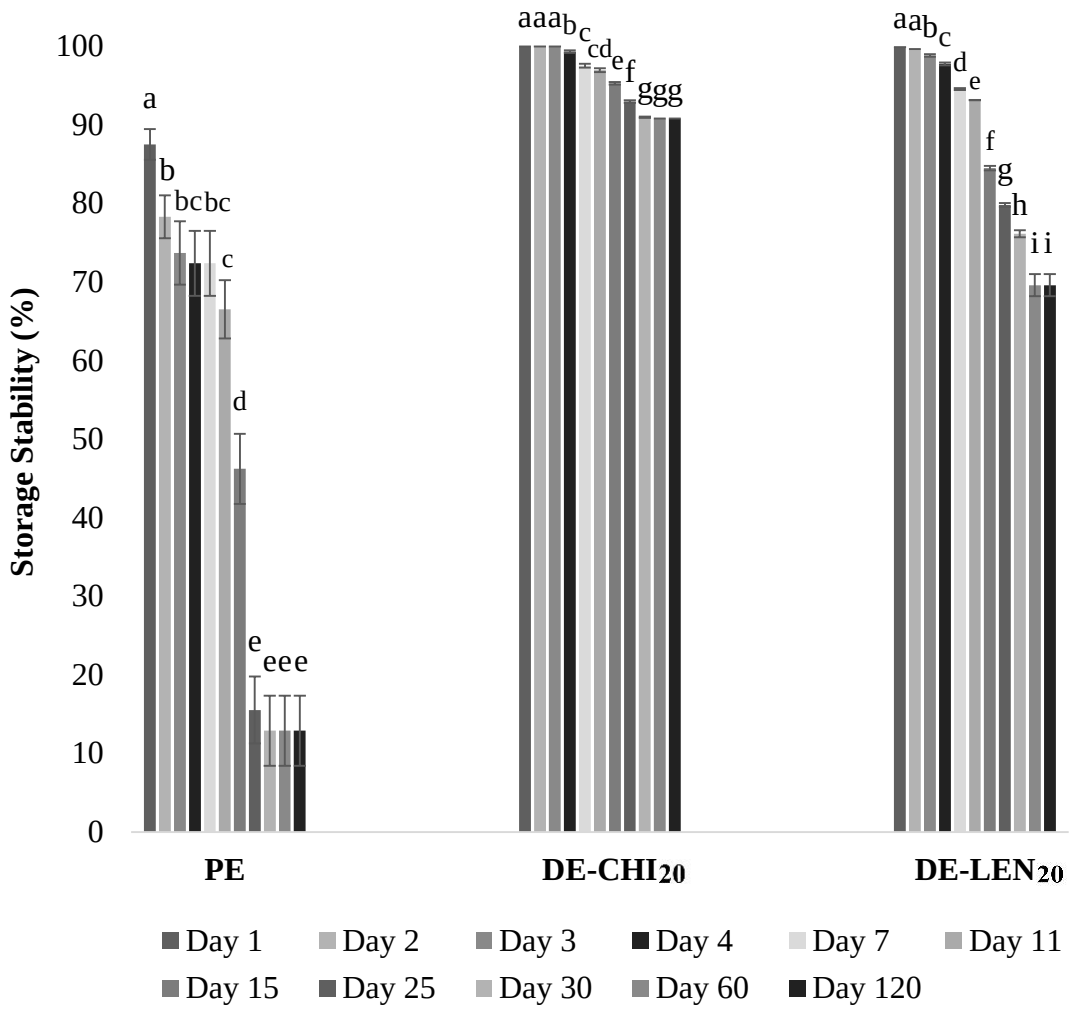

Figure 22 Storage stability (%) results of primary emulsion (PE), double emulsion with chickpea flour (DE-CHI_{20}) and double emulsion with lentil flour (DE-LEN_{20}) during 120 days.

PE showed distinct layers of oil and aqueous parts after 24 h of preparation, on the contrary, there was no difference on DE-CHI_{20} systems even after 3 days (Figure 22). The rapid phase separation in PE could be associated with the higher energy level of

interfacial layer because droplets thermodynamically tend to go to a less energic state. The walls of oil particles coated by outer layer in DE can prevent more aggregations than single-layers; consequently, the stability increases for double-layer emulsions. It can be clearly seen that DEs were more stable than PE (Figure 22). The storage stability of DE-CHI_{20} after 120 days was found as 90.80 % whereas the result of DE-LEN_{20} was 69.60 %. Therefore, a more stable emulsion was obtained when chickpea flour was used. It was concluded that chickpea flour was a better encapsulating agent than lentil flour in double emulsions when the stability values are considered. This can be explained by higher oil holding capacity and higher emulsifying activity of chickpea as compared to lentil (Ladjal Ettoumi & Chibane, 2015).

3.3.2 Optical Microscopy Analysis

Prepared single and double emulsions were analyzed by optical microscopy in order to see the continuous phases with oil and water droplets. The visual observation of droplet sizes and the distribution of the droplets inside the emulsions can be seen in Figure 23. Micrographs showed that droplets were in spherical shape. In the case of PE, the dispersion of water droplets in the continuous oil phase can be clearly observed (Figure 23 a). It is seen that DE technique changes the phase structure with the addition of flour mixture and DEs have their characteristic structures (Figure 23 b and c). In the larger droplets of DEs, smaller water droplets (W1 phase) can be observed inside the oil phase. When Figure 23 (b) and (c) parts were analyzed, the micrographs were similar regardless of their flour type, because flour addition enabled to form a matrix around dispersed droplets by the entrapment of oil phase inside the W2 phase. DEs had larger droplets compared to PE (Figure 23). Besides, DE-CHI_{20} had smaller droplets as compared to DE-LEN_{20}.

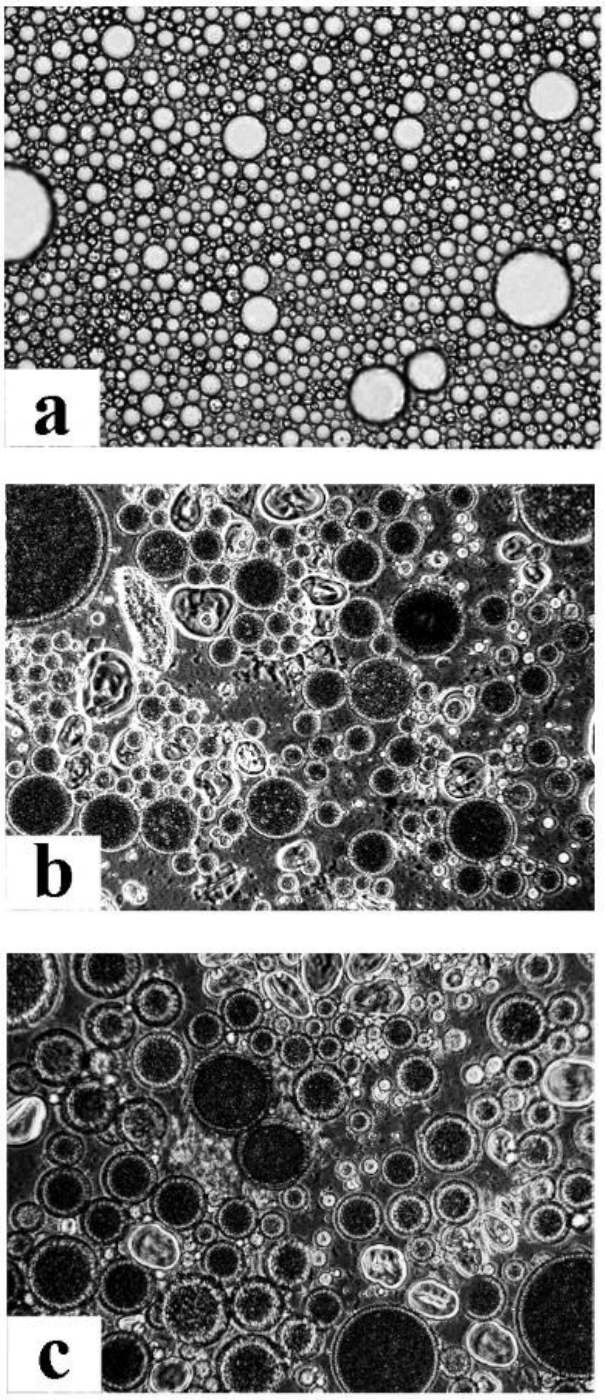

Figure 23 Optical microscopy analysis of (a) primary emulsion; (b) double emulsion with chickpea flour DE-CHI_{20}; and (c) double emulsion with lentil flour DE-LEN_{20} at 10X magnification.

When double emulsions were further analyzed at 40X magnification, the characteristic structure of double emulsion can be easily seen on Figure 24. The structure of the outer aqueous phase, LEN_{20}, can also be seen in Figure 24.

Figure 24 Optical micrograph of double emulsion with lentil flour (20 %) at 40X magnification.

3.3.3 Rheological Measurements

Apparent viscosity vs shear rate plots of emulsions are given in Figure 25. Measurement of viscosity is important for emulsions because it has a profound effect on the diameter of the dispersed phase of the emulsions due to its effect on the movement of surfactants and emulsifiers in the aqueous phase to reach the oil interface. Moreover, viscosity has an important role on the stability improvement by reducing the aggregation of droplets (van Aken, Vingerhoeds, & de Wijk, 2011). Results showed that PE had lower consistency coefficient than DEs (Figure 25, Table 10).

Besides, it was shown that different formulations of DE caused significantly different consistency coefficients (Figure 25, Table 10). DE-CHI_{20} had higher consistency coefficient than that of DE-LEN_{20}. PE had been found to have the lowest consistency coefficient, which could be related to the lowest storage stability (Figure 22). When internal friction of a matrix becomes weak, colloidal interaction can increase easily and therefore interfacial tension decreases, which results in bigger droplets and lower stability.

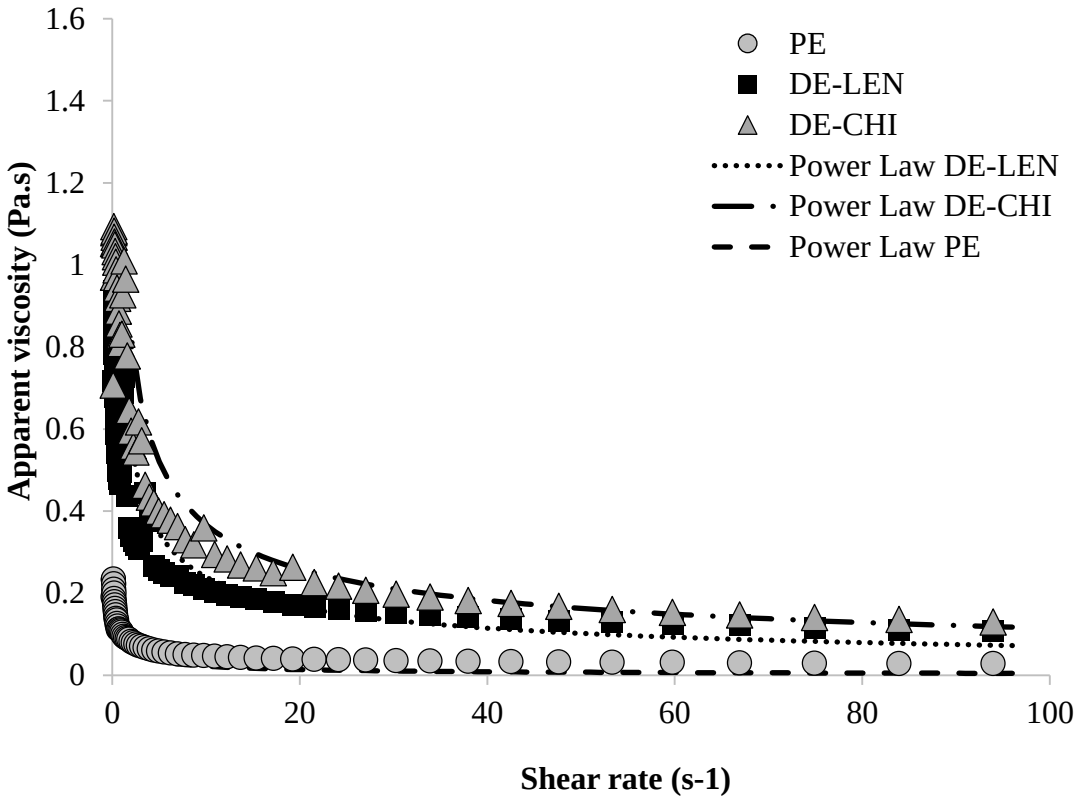

Figure 25 Apparent viscosity (Pa.s) vs shear rate (s^{-1}) data of primary emulsion (PE), double emulsion with chickpea flour (DE-CHI_{20}) and lentil flour (DE-LEN_{20}) at room temperature with Power Law fitting.

The most commonly observed flow behavior in emulsions is probably the shear thinning behavior, characterized by a decreasing apparent viscosity with an increasing shear rate (Bouyer, Mekhloufi, Rosilio, Grossiord, & Agnely, 2012). The rheological data (Figure 25) were analyzed to fit to a Power Law model. Table 10 shows the consistency coefficient (*K*) and flow behavior index (*n*) with coefficient of determination (R^2) values. These values were calculated from the data found in Figure 25. In a recent research, the flow properties of the double emulsion also fit to Power Law model with high coefficient of determination (Yildirim et al., 2017).

Table 10 Power law model fit for the primary emulsion (PE), double emulsion with chickpea flour (DE-CHI_{20}) and lentil flour (DE-LEN_{20}) (*K:* consistency coefficient, *n:* flow behavior index, R^2: coefficient of determination).

	K (Pa.s)	*n*	R^2
PE	0.106^{c}	0.681^{a}	0.999
DE-CHI_{20}	1.171^{a}	0.504^{c}	0.991
DE-LEN_{20}	0.809^{b}	0.529^{b}	0.995

*Values followed by the different letters within columns are significantly different at $p \leq 0.05$.

The consistency coefficients (*K* values) were found to be statistically different for all the emulsions; 1.171 Pa.s for DE-CHI_{20}, 0.809 Pa.s for DE-LEN_{20} and 0.106 for PE. Storage stability results were found to be directly correlated with the consistency

coefficients results (Figure 22). High consistency coefficient, as in the case of high viscosity, resulted in a great decrease in the possibility of phase separation through creaming and sedimentation. In this regard, chickpea flour provided significant increase in apparent viscosity and stability.

The flow behavior index (*n*) indicated the degree of the pseudoplasticity. Thus, lower *n* value means higher pseudoplasticity. In the table of rheological measurements results, *n* values were found to be lower than 1.0, which was an indicative of a non-Newtonian and shear-thinning flow characteristics. In terms of rheological behavior, all emulsions showed shear thinning properties (Table 10).

3.3.4 Particle Size Distribution

Particle size distributions were analyzed to investigate the effect of encapsulation technique. Table 11 shows the results of OLE and OLE containing emulsions. D_{50}, D_{32} and D_{43} values of OLE was found as 18.26 μm, 5.71 μm and 25.32 μm respectively. The results of PE were smaller than OLE, as expected. A size reduction was observed as a consequence of high-speed homogenization during emulsion formation. Additionally, the results indicated that particle sizes were significantly affected by the different techniques ($p \leq 0.05$). D_{43} results of PE, DE-CHI_{20} and DE-LEN_{20} were found as 19.22 μm, 52.27 μm and 74.07 μm respectively. PE had significantly smaller particle sizes in terms of D_{50}, D_{32} and D_{43} results than DEs ($p \leq 0.05$). Similarly, in another research, it was shown that the homogenization technique was effective on the droplet sizes of the emulsions (Moreau, Kim, Decker, & McClements, 2003).

Besides, a significant difference ($p \leq 0.05$) was observed between the different coating types on particle size results (Table 11). It was seen that the mean diameters of droplets found in DE-CHI_{20} were significantly smaller when compared to DE-LEN_{20} ($p \leq 0.05$). Therefore, it could be easily stated that usage of chickpea flour led smaller particle sizes. It was also found that DE-CHI_{20} had significantly higher instant and storage stabilities ($p \leq 0.05$) (Figure 21 and Figure 22), which could be explained by smaller

particle sizes of DE-CHI_{20}. DEs with smaller droplets have less tendency to aggregation, which makes them more stable during storage. The presence of biopolymers in the emulsion hinders the coalescence by reducing the capillary pressure between the phases and by increasing the viscoelastic properties of the interfacial layer. Therefore, the layer becomes more flexible and stable against disruption (Wilde, Mackie, Husband, Gunning, & Morris, 2004). Consistency coefficient of DE-CHI_{20} was found to be higher than that of DE-LEN_{20} (Table 10) therefore DE-CHI_{20} was expected to be more stable than DE-LEN_{20}. In the case where the particles do not have a yield stress that prevents free movement, the droplets having bigger sizes will merge faster than small ones; as indicated by the modified Stokes equation (Gouldby, Gunning, Hibberd, & Robins, 1991). This aggregation process increases the phase separation rate and decreases storage stability of emulsions. On the other hand, the reason of instability of PE can be explained by lack of protein at the outer phase.

Table 11 Particle size distribution results of samples (OLE: olive leaf extract, PE: primary emulsion, DE-CHI_{20}: double emulsion with chichkpea flour 20 %, DE-LEN_{20}: double emulsion with lentil flour 20 %).

Sample name	D_{50} (μm)	D_{32} (μm)	D_{43} (μm)	Span
OLE	$18.26 \pm 0.42^{c*}$	5.71 ± 0.10^{c}	25.32 ± 0.38^{c}	5.63 ± 0.16^{b}
PE	9.69 ± 0.15^{d}	4.06 ± 0.07^{d}	19.22 ± 1.08^{d}	7.97 ± 0.30^{a}
DE-CHI_{20}	31.40 ± 1.43^{b}	12.93 ± 0.35^{b}	52.27 ± 0.74^{b}	5.97 ± 0.34^{b}
DE-LEN_{20}	37.70 ± 0.20^{a}	15.43 ± 0.06^{a}	74.07 ± 0.90^{a}	2.74 ± 0.05^{c}

*Results (mean ± SD, n = 3) followed by the different letters within columns are significantly different at $p \leq 0.05$.

Additionally, an inverse relationship between D_{43} and span values can be observed in Table 11. The same relation can be seen between D_{50} and D_{32} results with span values. The relation of larger particles with small span values could be explained by the formation of uniform droplets in emulsion. Inverse relationship between mean diameters and span was also shown in a recent study (Lavelli et al., 2015). Smaller span values were an indicative of homogeneity of a distribution and can be seen in optical micrographs (Figure 23). DE-LEN_{20} had the smallest span value (Table 11) and contained more homogenous droplets than other emulsions (Figure 23).

3.3.5 OLE Release and Kinetics in Emulsions

In order to calculate release rate (RR), determination of TPC of emulsions are needed (Figure 26). Presence of a high amount of phenolic content in W2 or deterioration of polyphenols during processing increases RR. Therefore, choosing the right coating material becomes very important to decrease RR. The effect of different coating material types on RR during storage is shown in Figure 27. It is seen in Figure 27 that the RR of DE-CHI_{20} and DE-LEN_{20} after 30 days are 9.47 and 20.30 % respectively. Using LEN_{20} in W2 caused a fast increase in RR, whereas using CHI_{20} in W2 slowly increased release rate for 30 days. It was stated in a study that the interaction between polysaccharides and proteins can be modified by changing the microstructure and configuration of the environment, which resulted in a more regulated release character (Picone & da Cunha, 2010). This interconnected structure might prevent core material to go through W2. After 30 days, RR of DE-CHI_{20} reached to 9.47 % while it increased to 20.30 % for DE-LEN_{20}. It was seen that the RR was lower in DE-CHI_{20} than DE-LEN_{20}.

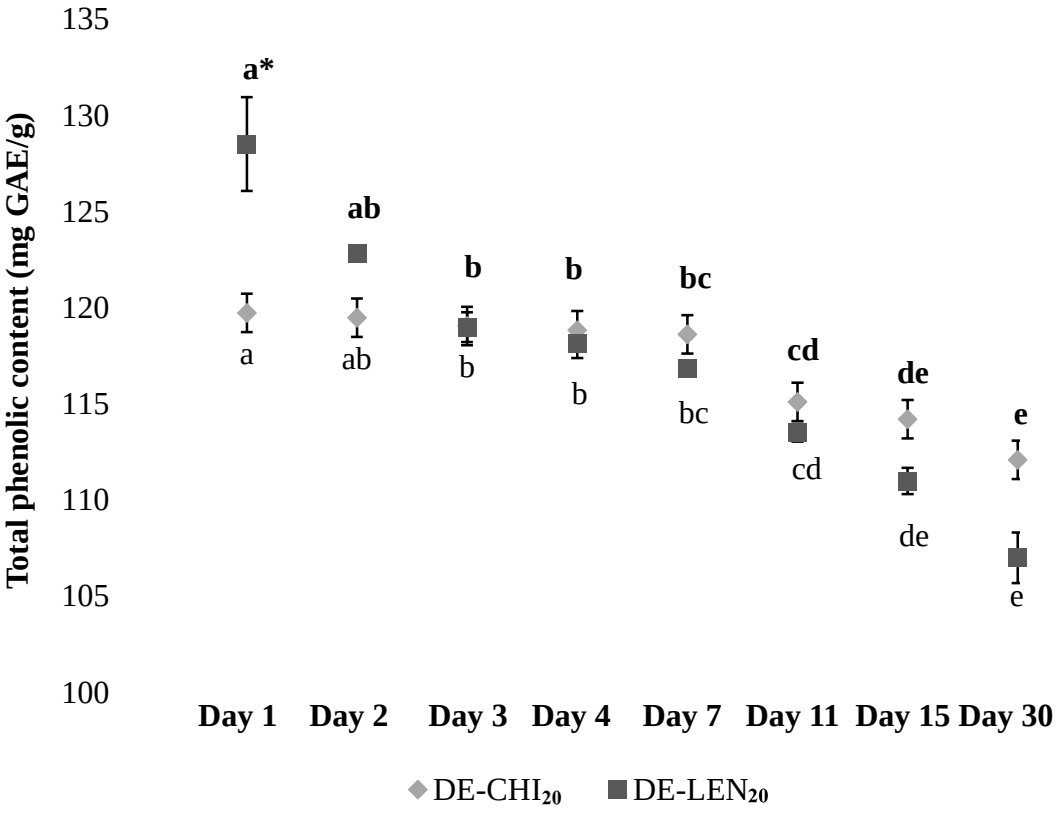

Figure 26 Total phenolic content (mg GAE/g) results of DE-CHI20 and DE-LEN20 during storage. * Different letters within same sample represent significant different values ($p \leq 0.05$).

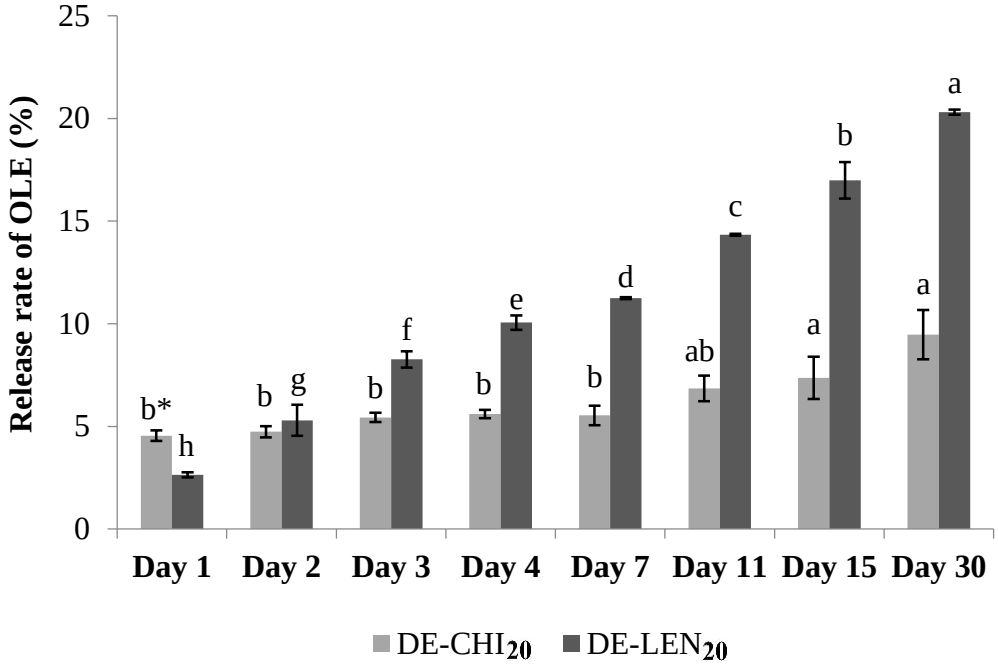

Figure 27 Release rate of OLE (%) during storage. *Different letters within same sample represent significant different values ($p \leq 0.05$).

It was also seen that the mean diameters of droplets found in DE-CHI_{20} were significantly smaller when compared to DE-LEN_{20} ($p \leq 0.05$) (Table 11). Therefore, it could easily be stated that usage of chickpea flour led to smaller particle sizes. It was also found that DE-CHI_{20} had significantly higher instant and storage stabilities and lower release rate than DE-LEN_{20} ($p \leq 0.05$) (Figure 21, Figure 22 and Figure 27). Both smaller particle size and higher stability of DE-CHI_{20} might be the reasons for lower release rate. Therefore, it was found that CHI_{20} was a better encapsulating agent than LEN_{20}.

TPC results of DEs for 30 days can be seen in Figure 26. A decreasing trend of TPC was due to the deterioration of phenolic compounds during storage at room

temperature. The reason of this decrease in TPC was the releasing of OLE through outer phases (W2). As released amount of OLE increased, phenolic compounds were exposed to environmental conditions like temperature and light, therefore they were degraded. TPC decreased sharply in DE-LEN_{20} for 30 days as compared to DE-CHI_{20} (Figure 26). The higher TPC result of DE-LEN_{20} at the first day was because of the fact that lentil inherently contained higher phenolic content than chickpea (Chung et al., 2008).

An increasing trend in RR results was observed in Figure 27, meaning that the released amount was rising during storage. After storage of 30 days, it was found that RR results of DE-CHI_{20} and DE-LEN_{20} were significantly different; 9.47 and 20.30 % respectively ($p \leq 0.05$). Higher RR was expected for DE-LEN_{20}, because better encapsulation was obtained by using chickpea flour because consistency coefficients of DE-CHI_{20} and DE-LEN_{20} were found as 1.171 and 0.809 Pa.s respectively (Table 10).

Encapsulation efficiency of DE-CHI_{20} after 24 h of preparation was 95.45 ± 0.17 %, whereas PE had an EE of 72.05 ± 2.97 %. The reason of PE having the least EE might be due to lacking a protein matrix around droplets. Additionally, it could be seen in optical micrographs and particle size results that DE-CHI_{20} had small particle sizes which led to higher storage stability and lower release rates (Figure 23, Table 11).

3.3.6 Total Antioxidant Activity

The same trend in TPC results was also seen for the TAA results. As can be seen in Figure 28, loss of TAA is increased during storage. That was related to the decreasing amount of phenolic content. When total phenolic content was degraded, the activity of antioxidants also decreased. The higher loss of TAA was found for DE-LEN_{20}, meaning that DE-CHI_{20} protected phenolic contents better than DE-LEN_{20}.

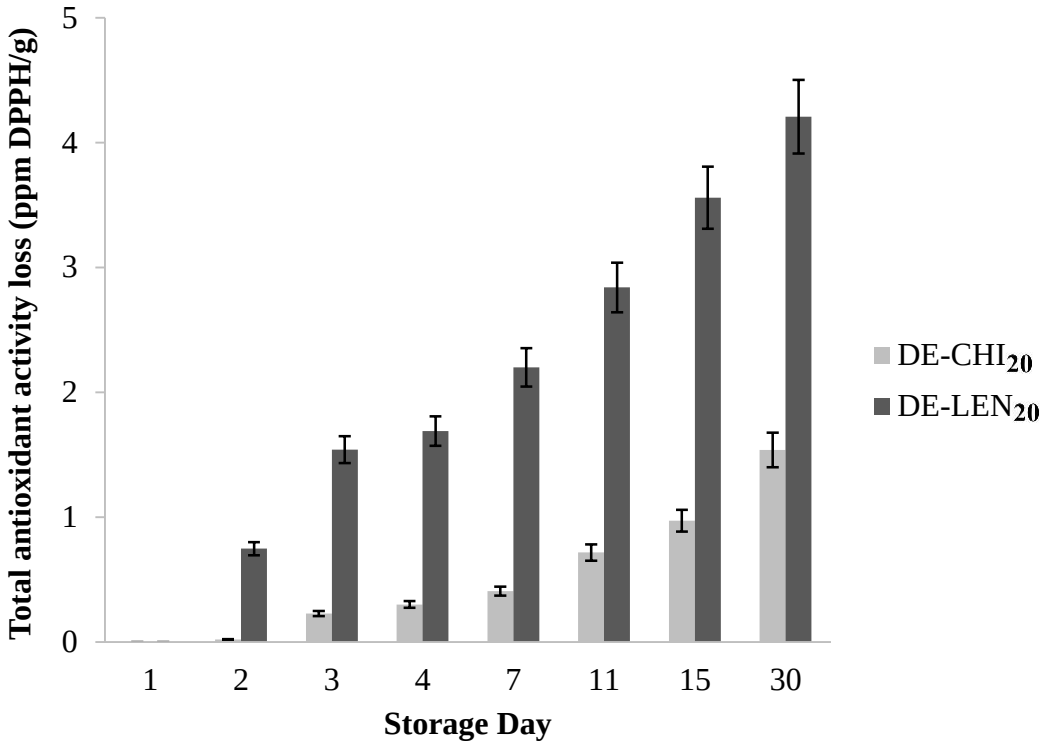

Figure 28 Total antioxidant activity loss (mg DPPH/g) of double emulsions with chickpea flour (DE-CHI_{20}) and lentil flour (DE-LEN_{20}) during storage.

3.3.7 Modelling OLE Release Kinetics

Release kinetics of OLE were calculated by using three different data. TPC of double emulsions (Figure 29), phenolic content of outer aqueous phase (corrected W2) (Figure 30), and loss of TAA (Figure 31) of double emulsions during storage were considered to find the model of release kinetics of OLE. The DE system can be simplified mathematically into a two-phase system consisting of OLE in the inner phase of W1/O emulsion, and the outer continuous phase (W2). The release kinetic of OLE was explained by using a first-order kinetics (Figure 29, Figure 30, Figure 31). Fitting of

first-order kinetics model yielded high coefficient of determination values ($R^2 > 0.99$) for the curves except the fitting curve of corrected W2 in DE-LEN_{20} (Figure 30).

When total phenolic contents of double emulsions were considered (the data in Figure 26), the kinetic constants of TPC of DE during storage were found as -0.003 for DE-CHI_{20} and -0.006 for DE-LEN_{20} (Figure 29). The minus sign showed a decreasing trend with increasing storage days. The higher decreasing rate belonged to DE-LEN_{20}, which showed a faster release of OLE through W2 in DE-LEN_{20} (Figure 30), as found in also RR results (Figure 27).

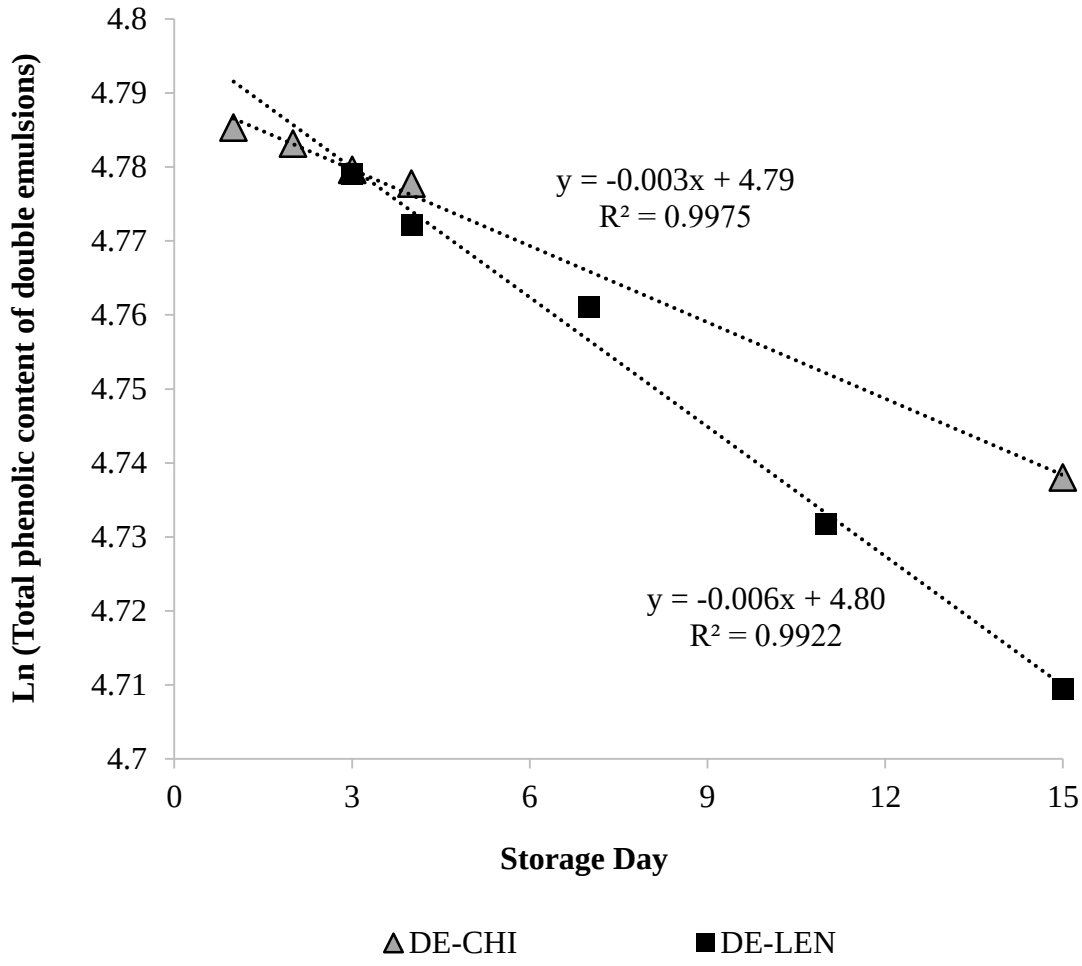

Figure 29 Fittings of the first-order kinetics of the total phenolic content of double emulsions with chickpea flour (DE-CHI_{20}) and lentil flour (DE-LEN_{20}) during storage.

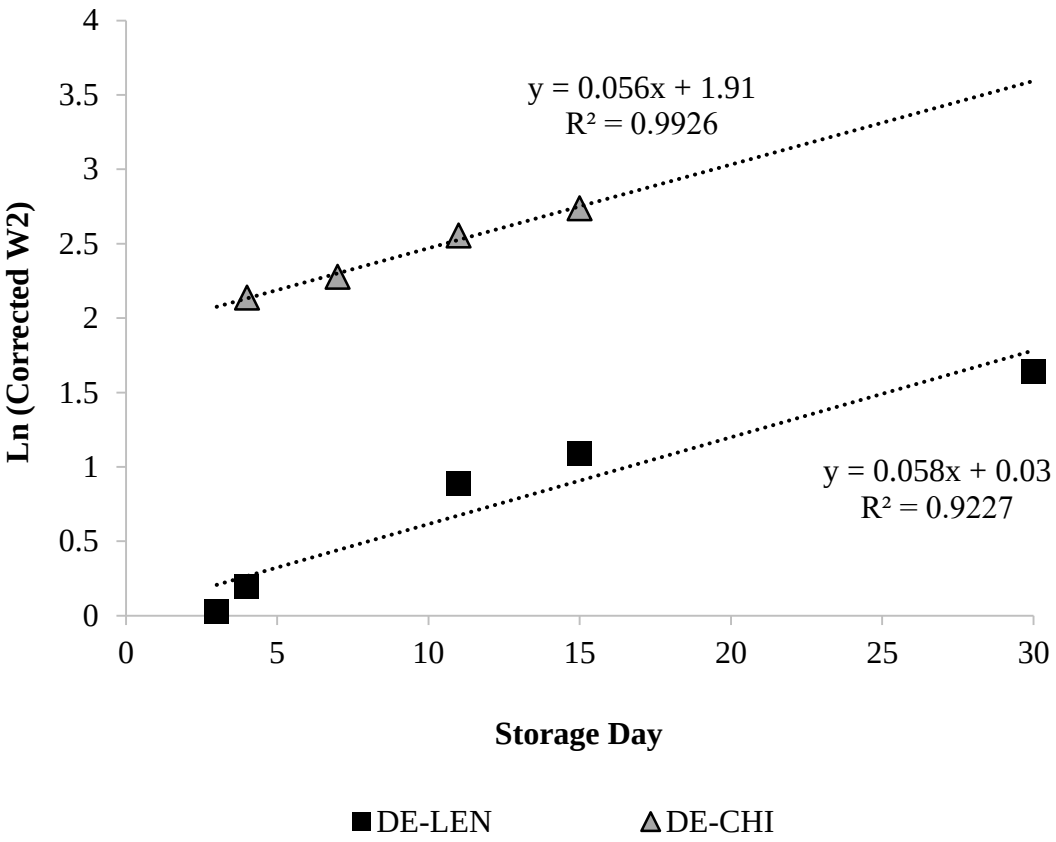

Figure 30 Fittings of the first-order kinetics of the corrected W2 results of double emulsions with chickpea flour (DE-CHI_{20}) and lentil flour (DE-LEN_{20}) during storage.

The kinetic constants of DE-CHI_{20} and DE-LEN_{20} in terms of corrected W2 results were given in Figure 30. The constants were found as 0.056 and 0.058 for DE-CHI_{20} and DE-LEN_{20} respectively. The positive value of kinetic constants showed that releasing of OLE through W2 was increasing during storage. DE-CHI_{20} had the slower increasing rate as compred to DE-LEN_{20}, which showed a slower release of OLE

through W2 in DE-CHI$_{20}$ (Figure 30), as can be found in also TPC (Figure 29) and RR results (Figure 27).

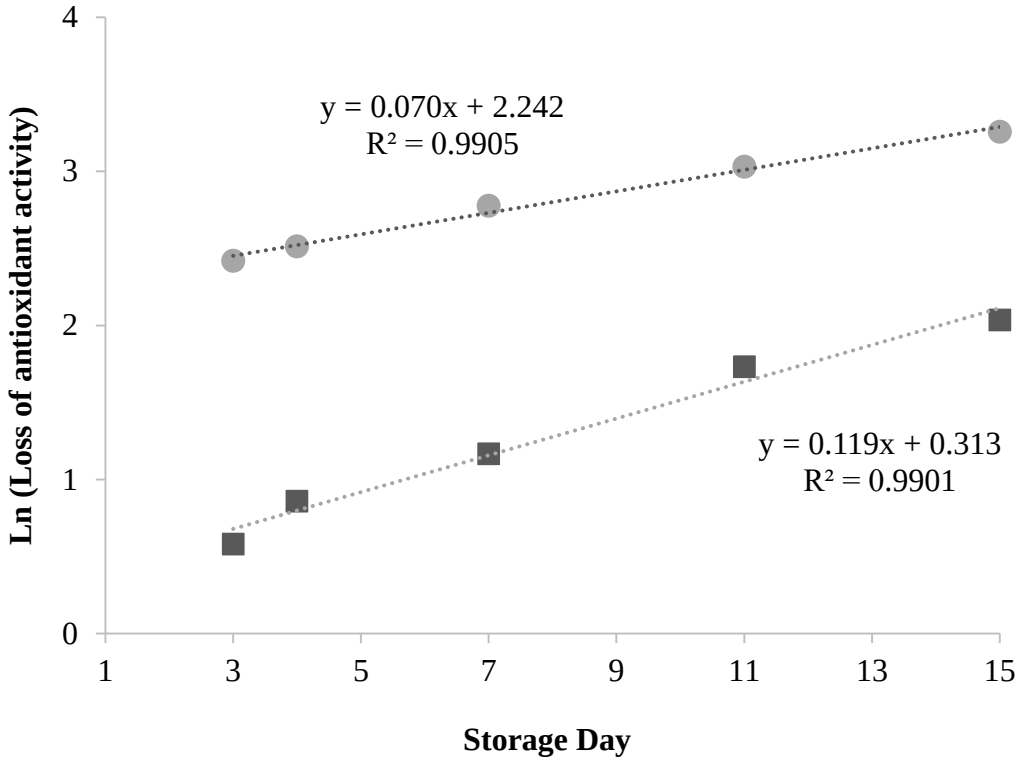

Figure 31 Release kinetics of OLE in terms of loss of antioxidant activity (%) during storage of double emulsion with chickpea flour (gray colored-circle: DE-CHI$_{20}$) and double emulsion with lentil flour (dark colored-square: DE-LEN$_{20}$).

When the loss of total antioxidant activity (TAA) of emulsions were analyzed, the OLE release rate constants (*k*) were found as 0.070 for DE-CHI$_{20}$ and 0.119 for DE-

LEN_{20} (Figure 31). It can be understood that loss of TAA is higher in DE-LEN_{20} than in DE-CHI_{20}.

3.3.8 NMR Relaxometry Analysis

Time domain nuclear magnetic resonance (TD-NMR) relaxometry is an analytical tool that utilizes radio frequency (RF) pulses to analyze materials. TD NMR methods are mainly based on acquiring signals to measure longitudinal (T_1) and transverse relaxation times (T_2). T1 is called the spin-lattice relaxation time, because it refers to the time it takes for the spins to give the energy they obtained from the radio frequency pulse back to the surrounding lattice, in order to go back to their equilibrium state (Hashemi, Bradley, & Lisanti, 2010). An example relaxation curve for T_1 is given in Figurre 15 in the section of *2.2.3.13*. T_2 is known as the transverse relaxation time and obtained by a signal curve decreasing exponentially. An example relaxation curve for T_2 is given in Figure 16 in the section of *2.2.3.13*.

TD - NMR provides a non-destructive, non-invasive method of analysis for the internal contents of food products (Mecit Halil Oztop, Bansal, Takhar, McCarthy, & McCarthy, 2014) since each organic material possesses a distinct relaxation time characteristic, T_2 measurement is a good way to reveal the internal compositions of foods, in this case emulsions (Barrabino, Keleşoğlu, Sørland, Simon, & Sjöblom, 2014). It is hypothesized that T_2 of specified proton pools can also be used to measure the efficiency of DEs. In literature, there are some studies investigating double emulsions by NMR measurements that focused on NMR spectroscopy experiments, free induction decay (FID) and transverse relaxation of different components (Vermeir, Balcaen, Sabatino, Dewettinck, & Van der Meeren, 2014; Vermeir, Sabatino, et al., 2014a). However, NMR relaxometry can also provide relaxation profiles for the whole emulsion system and supply information on the overall processes and mechanisms taking place within the system. T_2 values of emulsions was also used

to follow the polymorphic changes of lipid crystals (Sevdin, Ozel, Yucel, Oztop, & Alpas, 2017).

The other signal obtained through NMR is T_1 which is highly dependent on the mobility of water protons (Zhang, 2007). For instance, during gelatinization, starch granules start to absorb water and upon cooling this process leads to formation of a gel network. The formed gel network reduced the mobility of water protons since water molecules formed hydrogen bonds with the surrounding starch molecules as gelatinization took place. Lower movements of water molecules reduced the T1 value thus induced faster longitudinal relaxation (Ozel, Dag, Kilercioglu, Sumnu, & Oztop, 2017).

In recent years, with the popularity of mobile, bench type, low-frequency NMR systems, applications in various areas of this technique has increased. NMR relaxation spectrum gives information about the proton pools a the sample (Luyts et al., 2013; Mecit H. Oztop, Rosenberg, Rosenberg, McCarthy, & McCarthy, 2010) which could be associated with water distribution in food samples. This technique has also been commonly used by food scientists in the analysis of physiological and biochemical changes of fruits, vegetables and protein-rich samples (Mariette, Collewet, Davenel, Lucas, & Musse, 2007; Peters, Vergeldt, Boom, & van der Goot, 2017; Van As & Van Duynhoven, 2013). Analyzing emulsions by TD - NMR can give information about emulsion characteristics by exploring the proton populations found in the emulsions. In addition to relaxation profile analysis, self diffusion coefficients can be used for characterizing the mobility of water molecules within food materials with the help of pulsed field gradient NMR (PFG – NMR) (Salami, Rondeau-Mouro, van Duynhoven, & Mariette, 2013).

NMR results of PE, DE-CHI_{20} and DE-LEN_{20} can be seen in Table 12. It was known that T_1 results are highly related to mobility of water protons (Zhang, 2007). PE has the highest T_1 and T_2 times, and it was found that PE had the smallest consistency coefficient and stability, which could be a result of higher mobility of water protons.

In general, DEs had significantly lower relaxation times when compared to PE. Different encapsulation techniques significantly affected the T_1 and T_2 times ($p \leq 0.05$). Moreover, changing W2 of DE also significantly affected the relaxation times ($p \leq 0.05$). Relaxation times of DE-CHI_{20} were significantly shorter than DE-LEN_{20} (Table 12).

Table 12 Mean NMR relaxation times (T1, T2, T2a and T2b) of samples. (PE: primary emulsion, DE-CHI20: double emulsion with chickpea flour, DE-LEN20: double emulsion with lentil flour, CHI20: chickpea flour mixture, LEN20: lentil flour mixture)

	T_1	T_2	T_{2a}	T_{2b}
PE	296.32 ± 6.090^{a}*	205.48 ± 9.990^{a}	362.40 ± 12.580^{a}	65.40 ± 3.210^{a}
DE-CHI_{20}	210.24 ± 4.497^{c}	56.10 ± 1.195^{c}	110.71 ± 4.111^{c}	21.00 ± 0.816^{c}
DE-LEN_{20}	230.61 ± 1.253^{b}	67.03 ± 1.388^{b}	130.40 ± 6.656^{b}	26.00 ± 2.00^{b}
CHI_{20}	179.97 ± 3.859^{d}	24.76 ± 1.424^{e}	31.43 ± 5.412^{e}	12.75 ± 0.957^{e}
LEN_{20}	232.51 ± 3.259^{b}	34.08 ± 0.529^{d}	46.00 ± 0.816^{d}	18.50 ± 0.577^{d}

*Results (mean ± SD, n = 5) followed by the different letters within columns are significantly different at $p \leq 0.05$.

When W2 types of DEs were compared, it was seen in Table 12 that DE-CHI_{20} had significantly longer T_1 time than CHI_{20} ($p \leq 0.05$). It was previously found that DE-CHI_{20} was less viscous than CHI_{20}, meaning that it had more mobile protons, which led to significantly longer T_1 value. However, T_1 times of DE-LEN_{20} and LEN_{20} were not significantly different ($p > 0.05$). It was concluded that the addition of lentil flour was not as effective as in addition of chickpea flour, meaning that chickpea flour had higher emulsifying capacity than lentil flour. As known from the studies in the literature, chickpea had higher oil holding capacity and higher emulsifying activity than lentil (Ladjal Ettoumi & Chibane, 2015). Besides, it was found that consistency coefficient of DE-CHI_{20} was higher than that of DE-LEN_{20} (Table 10). High consistency coefficient might prevent oil droplets to agglomerate easily. It was also found that the release rate of OLE in DE-CHI_{20} was lower than DE-LEN_{20} (Figure 27).

T_2 relaxation was the same or faster than T_1, since return of magnetization in the z-direction basically causes loss of magnetization in the x–y plane (i.e. spin–spin relaxation). CHI_{20} was more viscous than LEN_{20}, therefore CHI_{20} had shorter T_2 time (24.76 ms) whereas LEN_{20} had longer T_2 time (34.08 ms).

For T_2 measurements, biexponential fitting was also performed because emulsions have basically two compartments; oil and water phases. T_{2a} and T_{2b} results were obtained after biexponential fitting (Table 12). The results were found to be in accordance with the T_1 and T_2 results. It was found that with increasing mobility of protons, T_1 and T_2 increased and stability decreased.

3.4 Baking Stability and *In Vitro* Release Tests

3.4.1 Baking Stability

Phenolic contents of cakes, which were retained after baking at 175 ^{o}C for 21 min, were determined to calculate the retention of phenolic contents of the cakes. The retention of phenolic contents of the cakes with encapsulated OLE were given with

respect to initial value before baking. The results can be seen in Figure 32. After baking, TPC of OLE was lost due to thermal processing although they were encapsulated. However, encapsulation had a significant effect ($p \leq 0.05$) on the retention of phenolic compounds as compared to uncoated OLE. Uncoated OLE has lost its phenolic content more than 90 % (Figure 32). It can be seen in Figure 32 that cakes with double emulsions retained phenolic content approximately 55%. Besides, cakes with LEN_{10}, CHI_{10}, and PE had approximately 25% retention after baking. This can be explained by the fact that phenolics entrapped in the coating material had an extra protecting barrier against thermal degradation. Likewise, loss of hydroxycitric acid of the encapsulated *Garcinia Cowa* extract was found lower as compared to loss of uncoated extract when samples were incorporated into a pasta (Pillai, Prabhasankar, Jena, & Anandharamakrishnan, 2012).

No significant difference on TPC was found between cakes with powdered microcapsules of OLE with LEN_{10} and CHI_{10} and with PE ($p > 0.05$). There was a significant positive difference in retention of phenolic content when DE technique was used (Figure 32). The most durable encapsulation technique was found as DE. Cakes with DEs had approximately 2 fold higher retention as compared to cakes with LEN10, CHI10, and PE. This can be explained by having an extra outer layer in double emulsions. A better encapsulation medium was formed with protein and starch present in the outer layer of DEs.

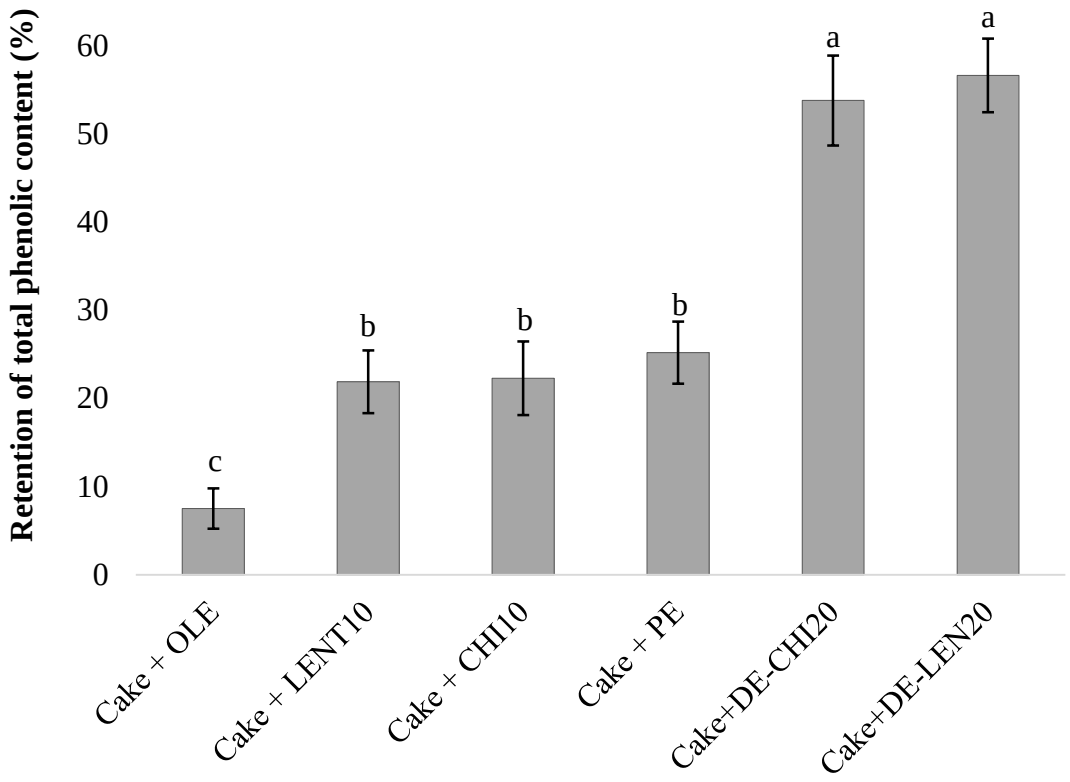

Figure 32 Retention of phenolic content (%) remained after baking at 175 ^{o}C for 21 min with respect to initial value before baking.

3.4.2 *In Vitro* Release Tests

In vitro release tests of cakes with uncoated OLE and encapsulated OLE were performed in SGF and SIF and the results can be seen in Figure 33 and Figure 34 respectively. These results showed that the release of phenolic compounds in SGF (pH 1.2) was less as compared to data obtained for digestion in SIF (pH 6.8). The results were in agreement with another research which was about dissolution / release of

microencapsulated flavonoids in simulated gastric and intestinal fluids (Sansone et al., 2011). Significantly less ($p \leq 0.05$) phenolic compounds were released from the encapsulated powders in SGF as compared to uncoated OLE. This can be related to the low solubility of coating materials at low pH. This is in fact the desired result, because phenolic compounds are vulnerable to acidic conditions. Their bioaccessibility increased if they were released in intestinal flora which had neutral pH. The difference between the amounts of released phenolic compounds from encapsulated OLE can be due to the difference in the particle size of the capsules. However, it must be noticed that during release experiments in simulated juices, several structural changes in emulsions may occur, including flocculation and coalescence of droplets and modification of interfacial layers. Digestion of phenolic compounds are not desired in gastric environment due to its low pH. Uncoated OLE was digested nearly 40% in simulated gastric fluid; and this caused to degradation of vulnerable and valuable phenolic compounds (Figure 33). On the other hand, both DE types were less digested in simulated gastric juice (Figure 33) because of having an extra layer.

In SIF, no significant difference ($p > 0.05$) between PE and DEs was observed (Figure 34). Amount of phenolic content released in SIF was higher for uncoated OLE. However, the difference between the releases of phenolic content from the encapsulated and uncoated OLE was not as significant as in the SGF test. Nevertheless, the release of DEs in SIF increased as compared to results of DEs in SGF (Figure 34), which was a desirable result in terms of increasing the intake of phenolic compounds to body because phenolic compounds were easily degraded in acidic medium of gastric juice (pH = 1.2).

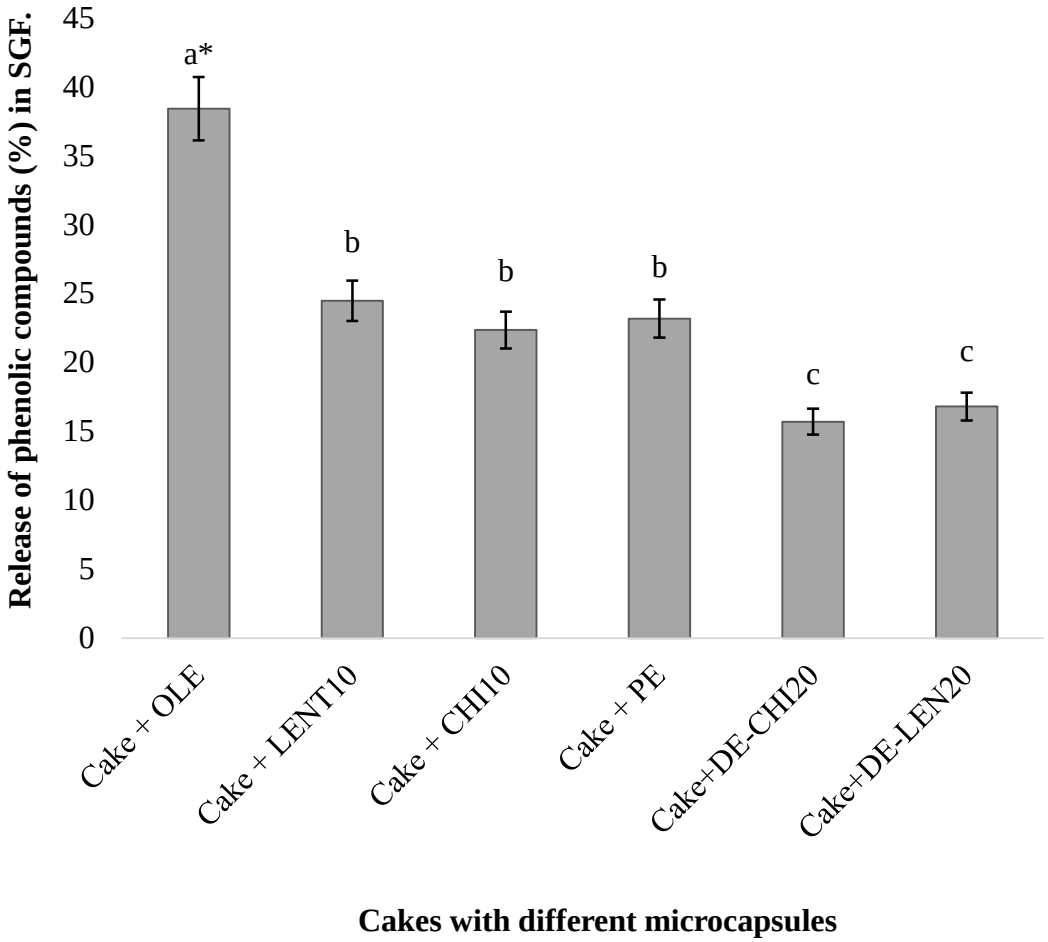

Figure 33 Release of phenolic compounds (%) from uncoated and encapsulated OLE in simulated gastric fluid (SGF). *Bars with different letters represent significant difference between results at $p \leq 0.05$.

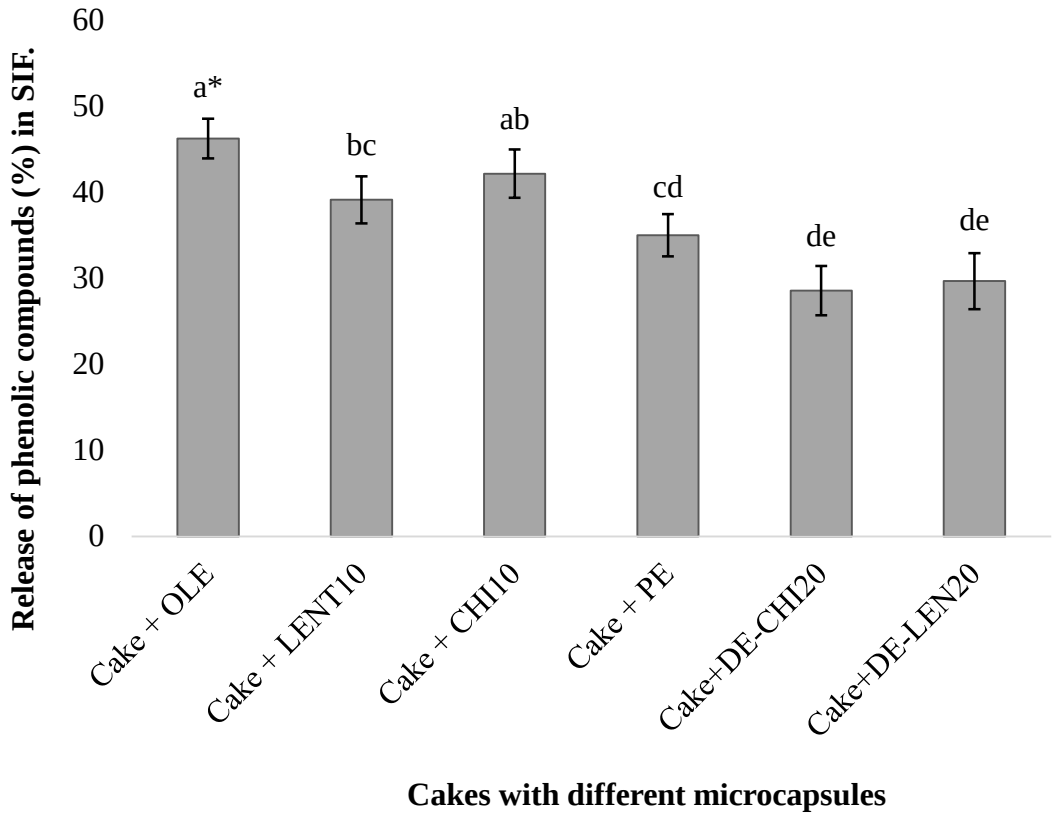

Figure 34 Release of phenolic compounds (%) from uncoated and encapsulated OLE in simulated intestinal fluid (SIF). *Bars with different letters represent significant difference between results at $p \leq 0.05$.

3.5 Correlation between Experimental Results

Correlation coefficients between experimental results are given in Table 13. DE-CHI_{20} and DE-LEN_{20} results were used for correlation. A high "r" value, which is *Pearson* correlation coefficient, represent a correlation between results (r > 0.9); however, p-value should also be analyzed with "r" value. The highlighted values, which are not enough for correlation, are not included in the interpretation.

Table 13 Correlations between experimental results with Pearson correlations (r) and p-values. T_1 and T_2 are NMR relaxometry results, K: consistency coefficient, IS: instant stability, SS: storage stability, D_{50}, D_{32} and D_{43} are particle size results, BS: baking stability, EE: encapsulation efficiency. The highlighted values, which are not enough for correlation, are not included in the interpretation.

		T1	T2	K	IS	SS	EE	D_{50}	D_{32}	D_{43}
T2	r	0.962								
	p	0.038								
K	r	-0.983	-0.982							
	p	0.017	0.018							
IS	r	-0.964	-0.892	0.959						
	p	0.036	0.108	0.041						
SS	r	-0.967	-0.973	0.997	0.959					
	p	0.033	0.027	0.003	0.041					
EE	r	-0.955	-0.976	0.993	0.941	0.998				
	p	0.045	0.024	0.007	0.059	0.002				
D_{50}	r	0.967	0.999	-0.987	-0.903	-0.978	0.980			
	p	0.033	0.001	0.013	0.097	0.022	0.020			
D_{32}	r	0.911	0.949	-0.971	-0.917	-0.986	-0.992	0.953		
	p	0.089	0.051	0.029	0.083	0.014	0.008	0.047		
D_{43}	r	0.994	0.984	-0.996	-0.954	-0.986	-0.979	0.988	0.946	
	p	0.006	0.016	0.004	0.046	0.014	0.021	0.012	0.054	
BS	r	0.999	0.950	-0.972	-0.960	-0.953	-0.938	0.956	0.889	0.987
	p	0.001	0.050	0.028	0.040	0.047	0.062	0.044	0.111	0.013
		T1	T2	K	IS	SS	EE	D_{50}	D_{32}	D_{43}

It can be seen from the Table 13 that;

- T_1 is highly positively correlated with T_2, D_{50}, D_{43}, BS, and inversely correlated with K, IS, SS, EE,
- T_2 is highly positively correlated with T_1, D_{50}, D_{43}, and inversely correlated with K, SS, EE,
- K is highly correlated with all the results,
- IS is highly positively correlated with K, SS and inversely correlated with T_1, D_{43}, BS,
- SS is highly correlated with all the results,
- EE is highly correlated with T_1, T_2, K, SS, D_{50}, D_{32} and D_{43},
- D_{50} is highly correlated with all the results except IS,
- D_{32} is highly positively correlated with D_{50} and inversely correlated with K, SS and EE,
- D_{43} is highly positively correlated with T_1, T_2, D_5 , BS and inversely correlated with K, IS, SS and EE,
- BS is highly positively correlated with T_1, D_{50}, D_{43} and inversely correlated with K, IS and SS.

It can be concluded that NMR relaxometry results are in correlation between consistency coefficients, storage stability, encapsulation efficiency, D_{50} and D_{43} results. NMR gives us information about mobility of protons. Consistency coefficient, storage stability and efficiency results may change if any difference in proton density occurs.

CHAPTER 4

CONCLUSION

Natural phenolic powder obtained from strawberry pomace was successfully encapsulated by using HSH, and HPH with different pressures and process cycles. The higher efficiencies were found for the microcapsules that were treated by HPH (5 and 7 cycles) than those treated by HSH. Applying 70 MPa pressure was found to be better than applying 50 MPa pressure during HPH. The efficiencies increased when MD:GA 6:4 was used as coating material. Coating with MD:GA 6:4 and using HPH at 70 MPa with 5 cycles were found as the optimum condition for the encapsulation of phenolic powder from strawberry pomace.

Besides, OLE was successfully encapsulated with different coatings by using HSH, HPH, single emulsion and double emulsion techniques. Encapsulation of OLE by HPH was found to be more efficient than HSH. Lentil flour was found to be a more efficient coating material as compared to chickpea flour in HPH treatment for dry capsules. Additionally, OLE was successfully encapsulated by DE method with different types of flour mixtures.

NMR results were found to be in correlation between the results of consistency coefficient, storage stability, encapsulation efficiency, D_{50} and D_{43}. The digestion of phenolic compounds and capsules showed that encapsulation prevented the degradation of phenolics at low pH values. The coating material formed a good barrier and caused to increase ingestion during intestinal absorption.

Results showed that DE was a better encapsulation technique than PE method in terms of storage stability, release rate, baking stability and *in vitro* release tests. Besides, DE also caused higher encapsulation efficiencies than entrapment technique.

To conclude, DE-CHI_{20} can be recommended to be incorporated into functional foods since the storage stability of DE-CHI_{20} was higher than that of DE-LEN_{20}.

As a future work, Fourier transform infrared (FTIR) spectroscopy can be suggested to analyze the emulsions with flours to determine the structural changes of proteins upon pressurization. FTIR can provide a better understanding of secondary structure changes of proteins after the influence of high pressure. Besides, further research can be focused on usage of double emulsions in high-fat containing foods such as mayonnaise and ice-cream or in flour containing baking products to produce gluten-free baking goods for the purpose of increasing antioxidant contents and reducing fat contents at the same time.

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APPENDICES

A. CALIBRATION CURVES

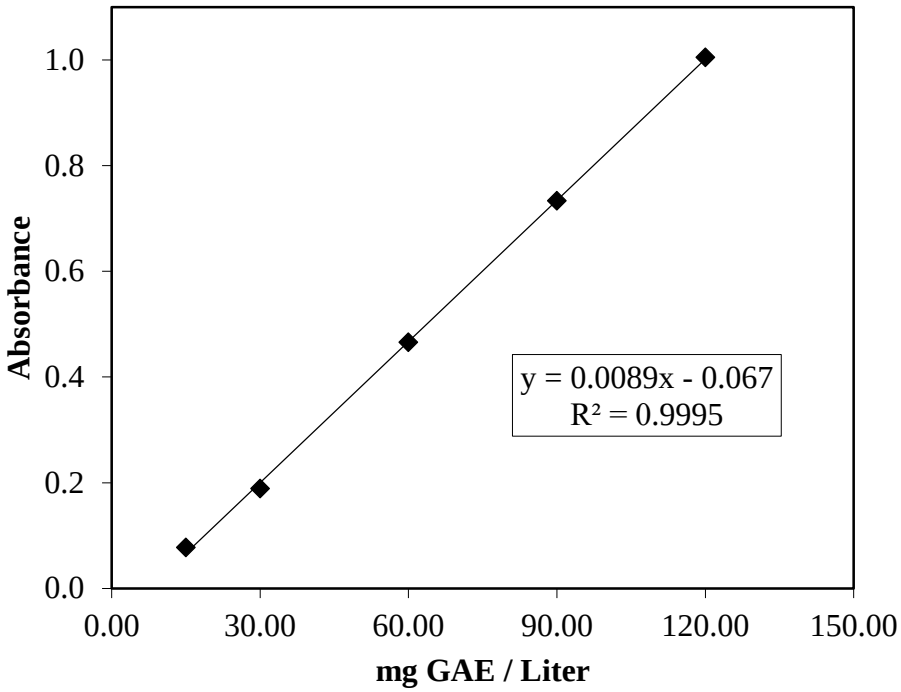

Figure A.1 Calibration curve prepared by gallic acid in ethanol:acetic acid:water mixture (50:50 v/v) for determination of total phenolic contents.

Absorbance (760 nm) = 0.0089 * (mg GAE / L) - 0.067

$R^2 = 0.9995$

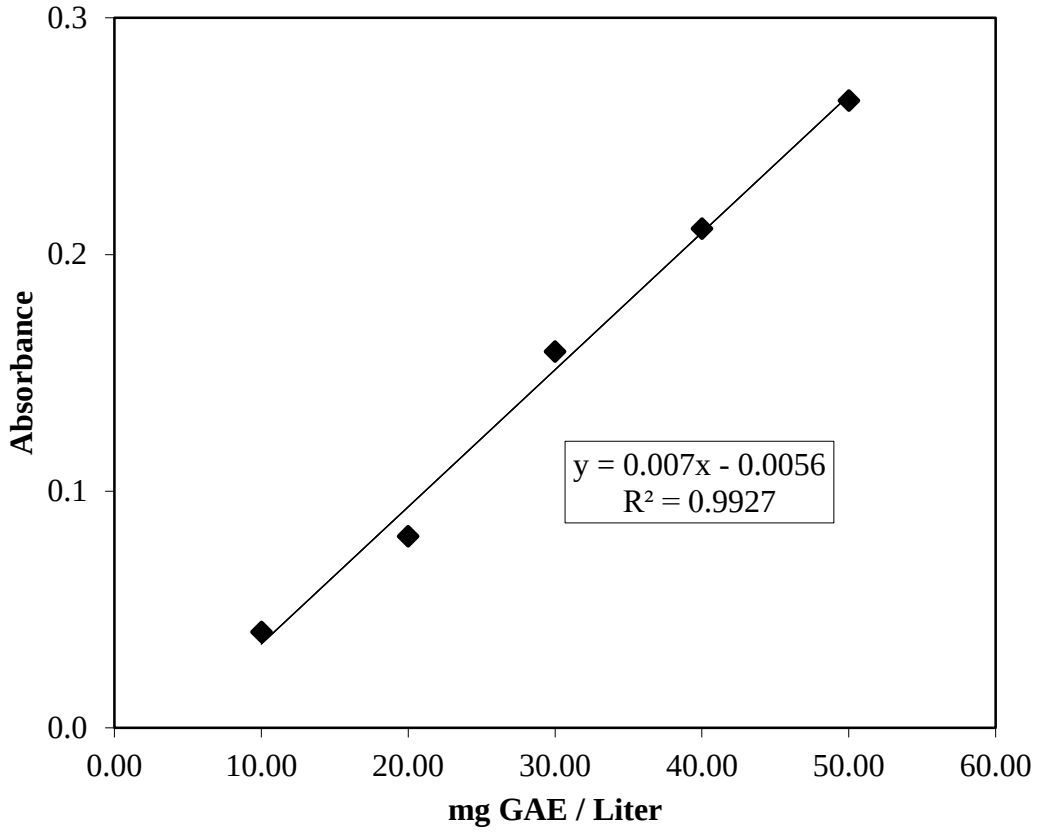

Figure A.2 Calibration curve prepared by gallic acid in ethanol:methanol mixture (50:8:42 v/v) for determination of surface phenolic contents.

Absorbance (760 nm) = 0.007 * (mg GAE / L) - 0.0056

$R^2 = 0.9927$

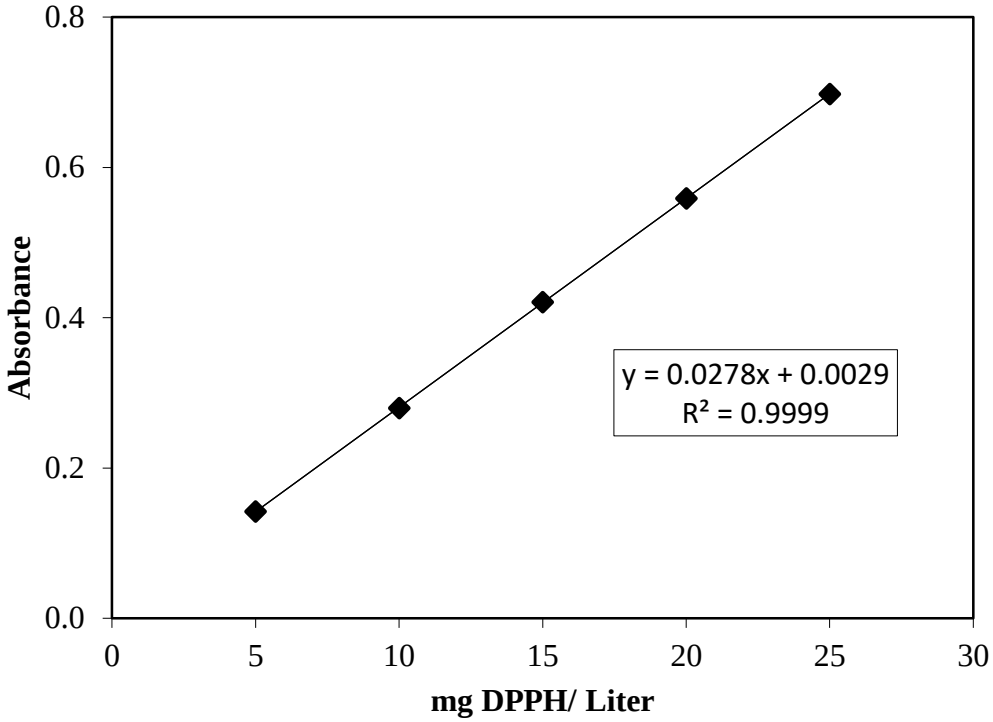

Figure A.3 Calibration curve prepared by DPPH˙ radical in methanol for determination of antioxidant activity.

Absorbance (517 nm) = 0.0278 * (mg DPPH / L) + 0.0029

$R^2 = 0.9999$

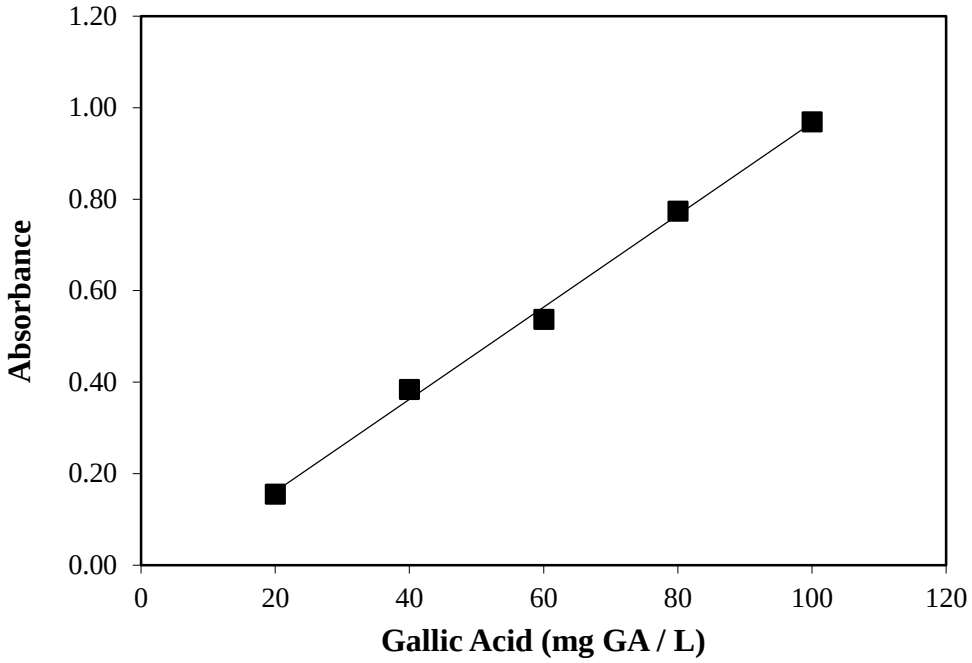

Figure A.4 Calibration curve prepared by gallic acid in SGF for determination of total phenolic contents of microcapsules.

Absorbance (760 nm) = 0.010 * (mg GAE / L) - 0.042

R^2 = 0.9932

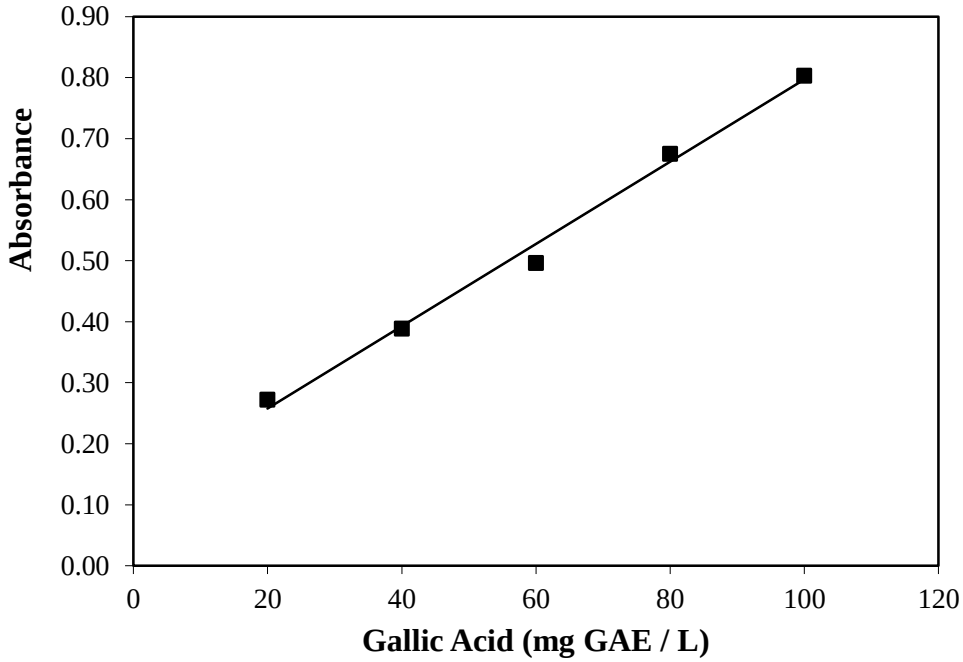

Figure A.5 Calibration curve prepared by gallic acid in SIF for determination of total phenolic contents of microcapsules

Absorbance (760 nm) = 0.008 * (mg GAE / L) + 0.1428

$R^2 = 0.9932$

B. STATISTICAL ANALYSES

Table B.1 Surface phenolic content of microcapsules of strawberry pomace

X1 MD:GA (6:4; 8:2; 10:0)
X2 Method (HSH, HPH)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	1 2

Number of Observations Read	12
Number of Observations Used	12

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	217.3144833	54.3286208	104.37	<.0001
Error	7	3.6439167	0.5205595		
Corrected Total	11	220.9584000			

R-Square	Coeff Var	Root MSE	Y Mean
0.983509	9.109825	0.721498	7.920000

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	197.9656333	197.9656333	380.29	<.0001
X2	2	16.3488500	8.1744250	15.70	0.0026

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	197.9656333	197.9656333	380.29	<.0001
X2	2	16.3488500	8.1744250	15.70	0.0026

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.52056

Number of Means 2

Critical Range .9850

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	11.9817	6	1
B	3.8583	6	2

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.52056

Number of Means	2	3
Critical Range	1.206	1.254

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	9.5600	4	10
B	7.2625	4	8
B	6.9375	4	6

Table B.2 Encapsulation efficiency of microcapsules of strawberry pomace

X1 MD:GA (6:4; 8:2; 10:0)

X2 HSH HPH

Class Level Information

Class	Levels	Values
X1	2	6 8 10
X2	2	1 2

Number of Observations Read	12
Number of Observations Used	12

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	693.1234333	173.2808583	40.17	<.0001
Error	7	30.1936583	4.3133798		
Corrected Total	11	723.3170917			

R-Square	Coeff Var	Root MSE	Y Mean
0.958257	2.578705	2.076868	80.53917

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	552.5704083	552.5704083	128.11	<.0001
X2	2	118.1670167	59.0835083	13.70	0.0038
X3	1	22.3860083	22.3860083	5.19	0.0568

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	552.5704083	552.5704083	128.11	<.0001
X2	2	118.1670167	59.0835083	13.70	0.0038

X3	1	22.3860083	22.3860083	5.19	0.0568

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 4.31338

Number of Means 2

Critical Range 2.835

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	91.325	6	2
B	72.753	6	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 4.31338

Number of Means 2 3

Critical Range 3.472 3.611

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	83.180	4	6
A	82.308	4	8
B	76.130	4	10

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 4.31338

Number of Means 2

Critical Range 2.835

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	81.905	6	20
A	79.173	6	15

Table B.3 Encapsulation efficiency of capsules

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X2 HPH (50 and 70 MPa)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	50 70

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	54.57303333	18.19101111	4.79	0.1776
Error	2	7.59790000	3.79895000		
Corrected Total	5	62.17093333			

R-Square	Coeff Var	Root MSE	Y Mean
0.877790	2.642714	1.949090	73.75333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	46.91163333	23.45581667	6.17	0.1394
X2	1	7.66140000	7.66140000	2.02	0.2914

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	46.91163333	23.45581667	6.17	0.1394
X2	1	7.66140000	7.66140000	2.02	0.2914

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05
Error Degrees of Freedom 2
Error Mean Square 3.79895

Number of Means 2 3
Critical Range 8.386 8.013

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	76.110	2	6
A	75.325	2	8
A	69.825	2	10

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05
Error Degrees of Freedom 2
Error Mean Square 3.79895

Number of Means 2
Critical Range 6.847

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2

A	74.883	3	70
A	72.623	3	50

Table B.4 Encapsulation efficiency of capsules

X1 MD:GA (6:4; 8:2; 10:0)
X2 HPH (3 5 7 passes)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	3	3 5 7

Number of Observations Read	9
Number of Observations Used	9

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	101.7980444	25.4495111	4.08	0.1011
Error	4	24.9603778	6.2400944		
Corrected Total	8	126.7584222			

R-Square	Coeff Var	Root MSE	Y Mean
0.803087	2.842389	2.498018	87.88444

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	80.77242222	40.38621111	6.47	0.0557
X2	2	21.02562222	10.51281111	1.68	0.2946

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	80.77242222	40.38621111	6.47	0.0557

X2	2	21.02562222	10.51281111	1.68	0.2946

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	4	
Error Mean Square	6.240094	

Number of Means	2	3
Critical Range	5.663	5.787

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	90.517	3	6
A	89.443	3	8
B	83.693	3	10

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	4	
Error Mean Square	6.240094	

Number of Means	2	3
Critical Range	5.663	5.787

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	89.003	3	3
A	88.927	3	5
A	85.723	3	7

Table B.5 Surface phenolic content of capsules having core to coating ratio 1:20, MD:GA 8:2 and HPH 700 MPa.

X1 HPH passes

Class Level Information

Class	Levels	Values
X1	6	3 5 7

Number of Observations Read 13
Number of Observations Used 13

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	5	12.67630641	2.53526128	15.72	0.0011
Error	7	1.12921667	0.16131667		
Corrected Total	12	13.80552308			

R-Square	Coeff Var	Root MSE	Y Mean
0.918205	10.00259	0.401642	4.015385

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	5	12.67630641	2.53526128	15.72	0.0011

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	5	12.67630641	2.53526128	15.72	0.0011

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	7
Error Mean Square	0.161317
Harmonic Mean of Cell Sizes	2.117647

NOTE: Cell sizes are not equal.

Number of Means	2	3	
Critical Range	.9229	.9597	.

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	5.8250	2	3
B	4.6400	2	5
B	4.4400	2	7

Table B.6 Encapsulation efficiency of capsules having core to coating ratio 1:20, MD:GA 8:2 HPH 5 Passes.

Class Level Information

Class	Levels	Values
X1	6	5 10 15 20 25 30

Number of Observations Read	13
Number of Observations Used	13

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	5	136.9851269	27.3970254	15.70	0.0011
Error	7	12.2161500	1.74516433		
Corrected Total	12	149.2012769			

R-Square	Coeff Var	Root MSE	Y Mean
0.918123	1.521889	1.321047	86.80308

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	5	136.9851269	27.3970254	15.70	0.0011

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	5	136.9851269	27.3970254	15.70	0.0011

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	7
Error Mean Square	1.745164
Harmonic Mean of Cell Sizes	2.117647

NOTE: Cell sizes are not equal.

Number of Means	2	3	4	5	6
Critical Range	3.036	3.157	3.221	3.257	3.278

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	90.130	3	15
A	89.565	2	25
A	88.450	2	20
B	85.405	2	10
B	84.750	2	5
C	80.855	2	30

Table B.7 Surface phenolic content of capsules

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	3	15 20 25

Number of Observations Read	9
Number of Observations Used	9

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	9.29777778	2.32444444	3.96	0.1057
Error	4	2.34984444	0.58746111		
Corrected Total	8	11.64762222			

R-Square	Coeff Var	Root MSE	Y Mean
0.798255	20.74005	0.766460	3.695556

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	7.40948889	3.70474444	6.31	0.0580
X2	2	1.88828889	0.94414444	1.61	0.3074

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	7.40948889	3.70474444	6.31	0.0580
X2	2	1.88828889	0.94414444	1.61	0.3074

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	0.587461

Number of Means	2	3
Critical Range	1.738	1.776

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	4.9633	3	10
B A	3.2333	3	8
B	2.8900	3	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	0.587461

Number of Means	2	3
Critical Range	1.738	1.776

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	4.3433	3	15
A	3.3733	3	20
A	3.3700	3	25

Table B.8 Antioxidant activity (DPPH˙) of capsules

X1 core to coating ratio (1, 1:10; 2, 1:20)

X2 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X3 UT (min)

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	6 8 10
X3	2	15 20

Number of Observations Read	12
Number of Observations Used	12

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	2.56010000	0.64002500	217.57	<.0001
Error	7	0.02059167	0.00294167		
Corrected Total	11	2.58069167			

R-Square	Coeff Var	Root MSE	Y Mean
0.992021	2.313706	0.054237	2.344167

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	2.49340833	2.49340833	847.62	<.0001
X2	2	0.04501667	0.02250833	7.65	0.0173
X3	1	0.02167500	0.02167500	7.37	0.0300

Source	DF	Type III SS	Mean Square	F Value	Pr > F

X1	1	2.49340833	2.49340833	847.62	<.0001
X2	2	0.04501667	0.02250833	7.65	0.0173
X3	1	0.02167500	0.02167500	7.37	0.0300

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.002942

Number of Means 2

Critical Range .07404

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	2.80000	6	1
B	1.88833	6	2

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.002942

Number of Means	2	3
Critical Range	.09068	.09430

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	2.42000	4	10
B A	2.34250	4	8
B	2.27000	4	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	7
Error Mean Square	0.002942

Number of Means	2
Critical Range	.07404

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	2.38667	6	15
B	2.30167	6	20

Table B.9 Antioxidant activity (DPPH) of capsules

X1 core to coating ratio (1, 1:10; 2, 1:20)

X2 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X3 UT (min)

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	6 8 10
X3	2	15 20

Number of Observations Read	12
Number of Observations Used	12

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	9	12362.95417	1373.66157	887.66	0.0011
Error	2	3.09500	1.54750		
Corrected Total	11	12366.04917			

R-Square	Coeff Var	Root MSE	Y Mean
0.999750	0.877333	1.243986	141.7917

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	12204.94083	12204.94083	7886.88	0.0001

X2	2	18.56167	9.28083	6.00	0.1429
X3	1	65.80083	65.80083	42.52	0.0227
X1*X2	2	1.47167	0.73583	0.48	0.6777
X1*X3	1	46.80750	46.80750	30.25	0.0315
X2*X3	2	25.37167	12.68583	8.20	0.1087

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	12204.94083	12204.94083	7886.88	0.0001
X2	2	18.56167	9.28083	6.00	0.1429
X3	1	65.80083	65.80083	42.52	0.0227
X1*X2	2	1.47167	0.73583	0.48	0.6777
X1*X3	1	46.80750	46.80750	30.25	0.0315
X2*X3	2	25.37167	12.68583	8.20	0.1087

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 2

Error Mean Square 1.5475

Number of Means 2

Critical Range 3.090

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1

A	173.6833	6	1
B	109.9000	6	2

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	1.5475

Number of Means	2	3
Critical Range	3.785	3.616

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	143.5500	4	10
A	140.9500	4	8
A	140.8750	4	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	1.5475

Number of Means	2
Critical Range	3.090

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	144.1333	6	15
B	139.4500	6	20

Table B.10 Antioxidant activity (DPPH˙) of capsules of OLE.

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X2 HPH

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	15 20

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	0.03550000	0.01183333	2.30	0.3176
Error	2	0.01030000	0.00515000		
Corrected Total	5	0.04580000			

R-Square	Coeff Var	Root MSE	Y Mean
0.775109	2.562982	0.071764	2.800000

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	0.01390000	0.00695000	1.35	0.4256
X2	1	0.02160000	0.02160000	4.19	0.1771

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	0.01390000	0.00695000	1.35	0.4256
X2	1	0.02160000	0.02160000	4.19	0.1771

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.00515

Number of Means	2	3
Critical Range	.3088	.2950

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	2.85000	2	10
A	2.81500	2	8
A	2.73500	2	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.00515

Number of Means	2
Critical Range	.2521

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	2.86000	3	15
A	2.74000	3	20

Table B.11 Antioxidant activity (DPPH˙) of capsules of OLE

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X2 HSH

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	15 20 25

Number of Observations Read	9
Number of Observations Used	9

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	0.06111111	0.01527778	1.70	0.3108
Error	4	0.03604444	0.00901111		
Corrected Total	8	0.09715556			

R-Square	Coeff Var	Root MSE	Y Mean
0.629003	5.002001	0.094927	1.897778

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	0.05575556	0.02787778	3.09	0.1542
X2	2	0.00535556	0.00267778	0.30	0.7580

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	0.05575556	0.02787778	3.09	0.1542
X2	2	0.00535556	0.00267778	0.30	0.7580

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	0.009011

Number of Means	2	3
Critical Range	.2152	.2199

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	2.00667	3	10
A	1.86333	3	6
A	1.82333	3	8

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	0.009011

Number of Means	2	3
Critical Range	.2152	.2199

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	1.91667	3	25
A	1.91333	3	15
A	1.86333	3	20

Table B.12 Antioxidant activity (TEAC) of capsules having core to coating ratio 1:10

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X2 UT (min)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	15 20

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	126.6050000	42.2016667	6.14	0.1432
Error	2	13.7433333	6.8716667		
Corrected Total	5	140.3483333			

R-Square	Coeff Var	Root MSE	Y Mean
0.902077	1.509291	2.621386	173.6833

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	14.8033333	7.4016667	1.08	0.4814
X2	1	111.8016667	111.8016667	16.27	0.0563

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	14.8033333	7.4016667	1.08	0.4814
X2	1	111.8016667	111.8016667	16.27	0.0563

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	6.871667

Number of Means	2	3
Critical Range	11.28	10.78

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	175.900	2	10
A	172.700	2	6
A	172.450	2	8

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	6.871667

Number of Means	2
Critical Range	9.209

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	178.000	3	15
A	169.367	3	20

Table B.13 Antioxidant activity (TEAC) of capsules having core to coating ratio 1:20

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X2 UT (min)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	3	15 20 25

Number of Observations Read	9
Number of Observations Used	9

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	41.28666667	10.32166667	2.72	0.1775
Error	4	15.15333333	3.78833333		
Corrected Tot	8	56.44000000			

R-Square	Coeff Var	Root MSE	Y Mean
0.731514	1.793333	1.946364	108.5333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	6.86000000	3.43000000	0.91	0.4739
X2	2	34.42666667	17.21333333	4.54	0.0934

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	6.86000000	3.43000000	0.91	0.4739

X2	2	34.42666667	17.21333333	4.54	0.0934

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	3.788333

Number of Means	2	3
Critical Range	4.412	4.509

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	109.700	3	10
A	108.300	3	8
A	107.600	3	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	3.788333

Number of Means	2	3
Critical Range	4.412	4.509

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	110.267	3	15
A	109.533	3	20
A	105.800	3	25

Table B.14 D32 values of capsules of strawberry pomace

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)
X2 UT (min)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	20 25

Number of Observations Read 6
Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	0.01990125	0.00663375	1.23	0.4781
Error	2	0.01079908	0.00539954		
Corrected Total	5	0.03070033			

R-Square	Coeff Var	Root MSE	Y Mean
0.648242	4.755063	0.073482	1.545333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	0.00810858	0.00405429	0.75	0.5711
X2	1	0.01179267	0.01179267	2.18	0.2775

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	0.00810858	0.00405429	0.75	0.5711
X2	1	0.01179267	0.01179267	2.18	0.2775

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.0054

Number of Means	2	3
Critical Range	.3162	.3021

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	1.59725	2	10
A	1.52175	2	6
A	1.51700	2	8

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.0054
Number of Means	2
Critical Range	.2581

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	1.58967	3	20
A	1.50100	3	25

Table B.15 Span of capsules of strawberry pomace

X1 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X2 UT (min)

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	20 25

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	0.13151250	0.04383750	1.27	0.4687
Error	2	0.06895833	0.03447917		
Corrected Total	5	0.20047083			

R-Square	Coeff Var	Root MSE	Y Mean
0.656018	4.754060	0.185686	3.905833

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	0.06000833	0.03000417	0.87	0.5347
X2	1	0.07150417	0.07150417	2.07	0.2865

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	0.06000833	0.03000417	0.87	0.5347

X2	1	0.07150417	0.07150417	2.07	0.2865

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 2

Error Mean Square 0.034479

Number of Means	2	3
Critical Range	.7989	.7633

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	3.9875	2	8
A	3.9650	2	6
A	3.7650	2	10

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.Alpha 0.05

Error Degrees of Freedom 2

Error Mean Square 0.034479

Number of Means 2

Critical Range .6523

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	4.0150	3	25
A	3.7967	3	20

Table B.16 Span of capsules of OLE by entrapment

X1 Flour (CHI, LEN)

X2 HSH HPH

Class Level Information

Class	Levels	Values
X1	3	6 8 10
X2	2	20 25

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	0.93502817	0.31167606	3.25	0.2439
Error	2	0.19156933	0.09578467		
Corrected Total	5	1.12659750			

R-Square	Coeff Var	Root MSE	Y Mean
0.829958	5.181066	0.309491	5.973500

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	0.93122800	0.46561400	4.86	0.1706
X2	1	0.00380017	0.00380017	0.04	0.8605

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	0.93122800	0.46561400	4.86	0.1706

X2	1	0.00380017	0.00380017	0.04	0.8605

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 2

Error Mean Square 0.095785

Number of Means	2	3
Critical Range	1.332	1.272

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	6.3805	2	6
A	6.0995	2	10
A	5.4405	2	8

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.Alpha 0.05

Error Degrees of Freedom 2

Error Mean Square 0.095785

Number of Means 2

Critical Range 1.087

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	5.9987	3	25
A	5.9483	3	20

Table B.17 L* values of microcapsules

X1 Flour Type (1, 2)
X2 SP OLE (1, 2)
X3 HSH HPH

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	6 8
X3	3	15 20 25

Number of Observations Read	15
Number of Observations Used	15

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	5	53.41971167	10.68394233	5.93	0.0107
Error	9	16.22198167	1.80244241		
Corrected Total	14	69.64169333			

R-Square	Coeff Var	Root MSE	Y Mean
0.767065	2.765713	1.342551	48.54267

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	49.03272111	49.03272111	27.20	0.0006
X2	2	3.56449333	1.78224667	0.99	0.4091
X3	2	0.82249722	0.41124861	0.23	0.8005

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	44.19840833	44.19840833	24.52	0.0008

X2	2	3.56449333	1.78224667	0.99	0.4091
X3	2	0.82249722	0.41124861	0.23	0.8005

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 1.802442

Harmonic Mean of Cell Sizes 7.2

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 1.601

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	50.0189	9	2
B	46.3283	6	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 1.802442

Number of Means 2 3

Critical Range 1.921 2.005

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	49.1360	5	8
A	48.5500	5	10
A	47.9420	5	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 1.802442

Harmonic Mean of Cell Sizes 4.5

NOTE: Cell sizes are not equal.

Number of Means 2 3

Critical Range 2.025 2.113

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	49.7233	3	25
A	48.4367	6	20
A	48.0583	6	15

Table B.18 a* values of microcapsules

X1 Flour Type (1, 2)

X2 SP OLE (1, 2)

X3 HSH HPH PE DE

Class	Levels	Values
X1	2	1 2
X2	3	6 8 10
X3	3	15 20 25 30 35

Number of Observations Read	15
Number of Observations Used	15

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	5	9.82702667	1.96540533	12.26	0.0008
Error	9	1.44334667	0.16037185		
Corrected Total	14	11.27037333			

R-Square	Coeff Var	Root MSE	Y Mean
0.871934	2.157830	0.400465	18.55867

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	9.76144000	9.76144000	60.87	<.0001
X2	2	0.05225333	0.02612667	0.16	0.8521
X3	2	0.01333333	0.00666667	0.04	0.9595

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	8.13453333	8.13453333	50.72	<.0001

X2	2	0.05225333	0.02612667	0.16	0.8521
X3	2	0.01333333	0.00666667	0.04	0.9595

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 0.160372

Harmonic Mean of Cell Sizes 7.2

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range .4774

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	19.5467	6	1
B	17.9000	9	2

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 0.160372

Number of Means 2 3

Critical Range .5729 .5980

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	18.6100	5	6
A	18.5900	5	10
A	18.4760	5	8

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 0.160372

Harmonic Mean of Cell Sizes

NOTE: Cell sizes are not equal.

Number of Means 2 3

Critical Range .6039 .6304

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	18.7567	6	15
A	18.6900	6	20
B	17.9000	3	25
A	18.6900	6	30
B	17.9000	3	35

Table B.19 b* values of microcapsules

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	6 8 10
X3	3	15 20 25

Number of Observations Read 15

Number of Observations Used 15

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	5	0.23278000	0.04655600	0.87	0.5342
Error	9	0.47898000	0.05322000		
Corrected Total	14	0.71176000			

R-Square	Coeff Var	Root MSE	Y Mean
0.327048	1.690814	0.230695	13.64400

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	0.02401000	0.02401000	0.45	0.5187
X2	2	0.13072000	0.06536000	1.23	0.3376
X3	2	0.07805000	0.03902500	0.73	0.5070

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	0.00270000	0.00270000	0.05	0.8268
X2	2	0.13072000	0.06536000	1.23	0.3376
X3	2	0.07805000	0.03902500	0.73	0.5070

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 0.05322

Harmonic Mean of Cell Sizes 7.2

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range .2750

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	13.6767	9	2
A	13.5950	6	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 0.05322

Number of Means 2 3
Critical Range .3300 .3445

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	13.7760	5	8
A	13.5800	5	10
A	13.5760	5	6

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05
Error Degrees of Freedom 9
Error Mean Square 0.05322
Harmonic Mean of Cell Sizes 4.5
NOTE: Cell sizes are not equal.

Number of Means 2 3
Critical Range .3479 .3631

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	13.7800	3	25
A	13.6600	6	20
A	13.5600	6	15

Table B.20 ΔE* values of microcapsules

X1 core to coating ratio (1, 1:10; 2, 1:20)

X2 MD:GA (6, 6:4; 8, 8:2; 10, 10:0)

X3 UT (min)

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	6 8 10
X3	3	15 20 25

Number of Observations Read 15

Number of Observations Used 15

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	5	58.40208000	11.68041600	6.78	0.0069
Error	9	15.51581333	1.72397926		
Corrected Total	14	73.91789333			

R-Square	Coeff Var	Root MSE	Y Mean
0.790094	2.329288	1.313004	56.36933

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	54.63127111	54.63127111	31.69	0.0003
X2	2	3.00645333	1.50322667	0.87	0.4507
X3	2	0.76435556	0.38217778	0.22	0.8054

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	48.96480000	48.96480000	28.40	0.0005

X2	2	3.00645333	1.50322667	0.87	0.4507
X3	2	0.76435556	0.38217778	0.22	0.8054

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 1.723979

Harmonic Mean of Cell Sizes 7.2

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 1.565

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	58.7067	6	1
B	54.8111	9	2

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 1.723979

Number of Means 2 3

Critical Range 1.878 1.961

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	56.9280	5	6
A	56.3480	5	10
A	55.8320	5	8

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 9

Error Mean Square 1.723979

Harmonic Mean of Cell Sizes 4.5

NOTE: Cell sizes are not equal.

Number of Means 2 3

Critical Range 1.980 2.067

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	56.8667	6	15
A	56.5067	6	20
A	55.1000	3	25

Table B.21 Retention of TPC in simulated gastric fluid

X1 type of sample (1, microcapsule with OLE; 2, microcapsule with DE-CHI; 3, microcapsule with DE-LEN)

Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	298.2091000	149.1045500	242.04	0.0005
Error	3	1.8481000	0.6160333		
Corrected Total	5	300.0572000			

R-Square	Coeff Var	Root MSE	Y Mean
0.993841	4.666337	0.784878	16.82000

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	298.2091000	149.1045500	242.04	0.0005

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	298.2091000	149.1045500	242.04	0.0005

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	3
Error Mean Square	0.616033

Number of Means	2	3
Critical Range	2.498	2.506

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	26.7900	2	3
B	11.8750	2	2
B	11.7950	2	1

Table B.22 Retention of TPC in simulated intestinal fluid

X1 type of sample (1, microcapsule with OLE; 2, microcapsule with DE-CHI; 3, microcapsule with DE-LEN)

Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	269.8082333	134.9041167	12.84	0.0338
Error	3	31.5287000	10.5095667		
Corrected Total	5	301.3369333			

R-Square	Coeff Var	Root MSE	Y Mean
0.895371	8.598301	3.241846	37.70333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	269.8082333	134.9041167	12.84	0.0338

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	269.8082333	134.9041167	12.84	0.0338

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	3
Error Mean Square	10.50957

Number of Means	2	3
Critical Range	10.32	10.35

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	47.070	2	3
B	34.305	2	1
B	31.735	2	2

Table B.23 Release rates of DE and PE of OLE

X1 type of sample

Class Level Information

Class	Levels	Values
X1	5	0 6 8 10 100

Number of Observations Read 10

Number of Observations Used 10

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	419.0810600	104.7702650	20.51	0.0027
Error	5	25.5451500	5.1090300		
Corrected Total	9	444.6262100			

R-Square	Coeff Var	Root MSE	Y Mean
0.942547	3.636463	2.260316	62.15700

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	4	419.0810600	104.7702650	20.51	0.0027

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	4	419.0810600	104.7702650	20.51	0.0027

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	5
Error Mean Square	5.10903

Number of Means	2	3	4	5
Critical Range	5.810	5.991	6.068	6.095

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	74.995	2	0
B	60.045	2	10
B	59.315	2	100
B	58.960	2	6
B	57.470	2	8

Table B.1 Microencapsulation of OLE

Class	Levels	Values
X1	2	1 2

Number of Observations Read	4
Number of Observations Used	4

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	1	0.03062500	0.03062500	0.05	0.8469
Error	2	1.27525000	0.63762500		
Corrected Total	3	1.30587500			

R-Square	Coeff Var	Root MSE	Y Mean
0.023452	0.883483	0.798514	90.38250

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	0.03062500	0.03062500	0.05	0.8469

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	0.03062500	0.03062500	0.05	0.8469

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.637625
Number of Means	2
Critical Range	3.436

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	90.4700	2	2
A			
A	90.2950	2	1

Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read 10

Number of Observations Used 10

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	2.35221000	1.17610500	2.45	0.1563
Error	7	3.36295000	0.48042143		
Corrected Total	9	5.71516000			

R-Square	Coeff Var	Root MSE	Y Mean
0.411574	0.748288	0.693124	92.62800

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	2.35221000	1.17610500	2.45	0.1563

Source	dF	Type III SS	Mean Square	F Value	Pr > F
X1	2	2.35221000	1.17610500	2.45	0.1563

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.480421

Harmonic Mean of Cell Sizes 3

NOTE: Cell sizes are not equal.

Number of Means	2	3
Critical Range	1.338	1.391

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	93.4300	2	1
A			
A	92.7325	4	2
A			
A	92.1225	4	3

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Class Level Information

Class	Levels	Values
X1	2	1 2

Number of Observations Read	6
Number of Observations Used	6

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	1	22.19520000	22.19520000	1.32	0.3142
Error	4	67.11360000	16.77840000		
Corrected Total	5	89.30880000			

R-Square	Coeff Var	Root MSE	Y Mean
0.248522	5.509273	4.096145	74.35000

Source	DF	Type I SS	Mean Square	F Value	Pr > F

X1	1	22.19520000	22.19520000	1.32	0.3142

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	22.19520000	22.19520000	1.32	0.3142

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	4
Error Mean Square	16.7784
Harmonic Mean of Cell Sizes	2.666667
Number of Means	2
Critical Range	9.849

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	75.710	4	2
A			
A	71.630	2	1

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	1 2 3

Number of Observations Read	20
Number of Observations Used	20

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	6.93293750	2.31097917	1.38	0.2835
Error	16	26.70344250	1.66896516		
Corrected Total	19	33.63638000			

R-Square	Coeff Var	Root MSE	Y Mean
0.206114	1.388093	1.291884	93.06900

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	3.88962000	3.88962000	2.33	0.1464
X2	2	3.04331750	1.52165875	0.91	0.4217

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	3.88962000	3.88962000	2.33	0.1464
X2	2	3.04331750	1.52165875	0.91	0.4217

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	16
Error Mean Square	1.668965
Number of Means	2
Critical Range	1.225

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	93.5100	10	1
A			
A	92.6280	10	2

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	16
Error Mean Square	1.668965
Harmonic Mean of Cell Sizes	6

NOTE: Cell sizes are not equal.

Number of Means	2	3
Critical Range	1.581	1.658

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	93.5238	8	3
A			
A	92.8750	8	2
A			
A	92.5475	4	1

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Class Level Information

Class	Levels	Values
X1	9	1 2 3 4 5 6 7 8 10

Number of Observations Read	11
Number of Observations Used	11

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	8	54.45922273	6.80740284	146.00	0.0068
Error	2	0.09325000	0.04662500		

Corrected Total	10	54.55247273

R-Square	Coeff Var	Root MSE	Y Mean
0.998291	0.780498	0.215928	27.66545

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	8	54.45922273	6.80740284	146.00	0.0068

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	8	54.45922273	6.80740284	146.00	0.0068

The GLM Procedure

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.046625
Harmonic Mean of Cell Sizes	1.125

Number of Means	2	3	4	5	6	7	8	9
Critical Range	1.239	1.184	1.131	1.085	1.045	1.009	.9773	.9490

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	31.9300	1	1
A			
B A	30.9700	1	2
B			
B C	29.8600	1	3
C			
C	28.7500	1	4

	D	27.3100	1	5
	D			
E	D	26.2100	2	10
E	D			
E	D	26.1300	1	7
E				
E		25.9250	2	8
E				
E		25.1000	1	6

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Class Level Information

Class	Levels	Values
X1	7	1 2 3 4 5 6 7

Number of Observations Read 14

Number of Observations Used 14

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	6	19.44188571	3.24031429	5.13	0.0249
Error	7	4.42500000	0.63214286		
Corrected Total	13	23.86688571			

R-Square	Coeff Var	Root MSE	Y Mean
0.814597	3.009690	0.795074	26.41714

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	6	19.44188571	3.24031429	5.13	0.0249

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	6	19.44188571	3.24031429	5.13	0.0249

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.632143

Number of Means	2	3	4	5	6	7

Critical Range	1.880	1.955	1.995	2.017	2.030	2.036

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	28.0500	2	5
A			
A	27.5000	2	4
A			
A	26.9600	2	3
A			
A	26.1300	2	6
A			
A	26.1000	2	2
A			
A	26.0600	2	1
B	24.1200	2	7

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Class Level Information

Class	Levels	Values
X1	9	1 2 3 4 5 6 7 8 10

Number of Observations Read 11

Number of Observations Used 11

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	8	13.76347273	1.72043409	2.17	0.3535
Error	2	1.58600000	0.79300000		
Corrected Total	10	15.34947273			

R-Square	Coeff Var	Root MSE	Y Mean
0.896674	7.254896	0.890505	12.27455

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	8	13.76347273	1.72043409	2.17	0.3535

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	8	13.76347273	1.72043409	2.17	0.3535

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	0.793
Harmonic Mean of Cell Sizes	1.125

Number of Means	2	3	4	5	6	7	8	9
Critical Range	5.109	4.881	4.665	4.475	4.308	4.161	4.031	3.914

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	14.310	1	1
A	14.020	1	2
A	13.200	1	3
A	12.860	1	7
A	12.270	1	6
A	12.080	1	4
A	11.430	2	10
A	11.230	2	8
A	10.960	1	5

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Class Level Information

Class	Levels	Values
X1	7	1 2 3 4 5 6 7

Number of Observations Read 21

Number of Observations Used 21

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	6	17.25424890	2.87570815	6.53	0.0019
Error	14	6.16433223	0.44030944		
Corrected Total	20	23.41858113			

R-Square	Coeff Var	Root MSE	Y Mean

0.736776 1.006315 0.663558 65.93938

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	6	17.25424890	2.87570815	6.53	0.0019

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	6	17.25424890	2.87570815	6.53	0.0019

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 14

Error Mean Square 0.440309

Number of Means	2	3	4	5	6	7
Critical Range	1.162	1.218	1.252	1.275	1.292	1.303

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	67.6909	3	4
B	66.5011	3	5
B	66.1815	3	1
C B	65.7612	3	2
C B	65.4987	3	6
C B	65.2944	3	3
C	64.6480	3	7

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Class Level Information

Class	Levels	Values

X1	7	1 2 3 5 6 7 8
Number of Observations Read		21
Number of Observations Used		21

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	6	30.08678095	5.01446349	24.34	<.0001
Error	14	2.88460000	0.20604286		
Corrected Total	20	32.97138095			

R-Square	Coeff Var	Root MSE	Y Mean
0.912512	1.660739	0.453919	27.33238

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	6	30.08678095	5.01446349	24.34	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	6	30.08678095	5.01446349	24.34	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	14
Error Mean Square	0.206043

Number of Means	2	3	4	5	6	7
Critical Range	.7949	.8329	.8564	.8723	.8836	.8917

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1

A	28.5000	3	6
A	28.0667	3	5
A	28.0067	3	2
A	27.9667	3	3
A	27.7067	3	1
B	26.1333	3	7
C	24.9467	3	8

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Class Level Information

Class	Levels	Values
X1	7	1 2 3 4 5 6 7

Number of Observations Read 21

Number of Observations Used 21

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	6	9.88872381	1.64812063	3.63	0.0219
Error	14	6.35553333	0.45396667		
Corrected Total	20	16.24425714			

R-Square	Coeff Var	Root MSE	Y Mean
0.608752	1.626343	0.673770	41.42857

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	6	9.88872381	1.64812063	3.63	0.0219

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	6	9.88872381	1.64812063	3.63	0.0219

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 14

Error Mean Square 0.453967

Number of Means	2	3	4	5	6	7
Critical Range	1.180	1.236	1.271	1.295	1.311	1.324

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	42.3400	3	3
A	41.9800	3	7
A	41.7500	3	2
A	41.5167	3	5
A	41.3000	3	6
B A	41.0633	3	1
B	40.0500	3	4

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Class Level Information

Class	Levels	Values
X1	9	1 2 3 4 5 6 7 8 10

Number of Observations Read 11

Number of Observations Used 11

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Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F

Model	8	62.96366364	7.87045795	0.73	0.6913
Error	2	21.50650000	10.75325000		
Corrected Total	10	84.47016364			

R-Square	Coeff Var	Root MSE	Y Mean
0.745395	5.261745	3.279215	62.32182

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	8	62.96366364	7.87045795	0.73	0.6913

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	8	62.96366364	7.87045795	0.73	0.6913

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	2
Error Mean Square	10.75325
Harmonic Mean of Cell Sizes	1.125

NOTE: Cell sizes are not equal.

Number of Means	2	3	4	5	6	7	8	9
Critical Range	18.81	17.97	17.18	16.48	15.86	15.32	14.84	14.41

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	65.840	1	7
A	64.630	1	2
A	63.610	1	6
A	62.975	2	10
A	62.780	1	3
A	62.645	2	8
A	61.710	1	1

A 58.920 1 4

A 56.810 1 5

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Class Level Information

Class	Levels	Values
X1	7	1 2 3 4 5 6 7

Number of Observations Read 14

Number of Observations Used 14

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	6	85.16034286	14.19339048	72.79	<.0001
Error	7	1.36500000	0.19500000		
Corrected Total	13	86.52534286			

R-Square	Coeff Var	Root MSE	Y Mean
0.984224	1.021114	0.441588	43.24571

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	6	85.16034286	14.19339048	72.79	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	6	85.16034286	14.19339048	72.79	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 7

Error Mean Square 0.195

Number of Means	2	3	4	5	6	7
Critical Range	1.044	1.086	1.108	1.120	1.127	1.131

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	48.9700	2	3
B	43.8600	2	1
C B	42.8200	2	2
C D	42.2500	2	5
C D	41.9500	2	7
D	41.5500	2	4
D	41.3200	2	6

Table B.25. Microencapsulation of OLE through emulsification emthods.

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Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	1 2 3

Number of Observations Read	24
Number of Observations Used	24

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	505.9150958	168.6383653	8.42	0.0008
Error	20	400.6547000	20.0327350		
Corrected Total	23	906.5697958			

R-Square	Coeff Var	Root MSE	Y Mean

0.558054 9.880795 4.475794 45.29792

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	419.5884375	419.5884375	20.95	0.0002
X2	2	86.3266583	43.1633292	2.15	0.1421

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	419.5884375	419.5884375	20.95	0.0002
X2	2	86.3266583	43.1633292	2.15	0.1421

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 20

Error Mean Square 20.03273

Number of Means 2

Critical Range 3.812

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	49.479	12	1
B	41.117	12	2

The GLM Procedure

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 20

Error Mean Square 20.03273

Number of Means 2 3

Critical Range 4.668 4.900

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	47.976	8	3
A	44.083	8	1
A	43.835	8	2

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	1 2 3

Number of Observations Read 24

Number of Observations Used 24

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	144.3028750	48.1009583	1.95	0.1546
Error	20	494.2537250	24.7126863		
Corrected Total	23	638.5566000			

R-Square	Coeff Var	Root MSE	Y Mean
0.225983	9.011485	4.971186	55.16500

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	106.0921500	106.0921500	4.29	0.0514
X2	2	38.2107250	19.1053625	0.77	0.4749

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	106.0921500	106.0921500	4.29	0.0514
X2	2	38.2107250	19.1053625	0.77	0.4749

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	20
Error Mean Square	24.71269
Number of Means	2
Critical Range	4.233

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	57.268	12	2
A	53.063	12	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	20	
Error Mean Square	24.71269	
Number of Means	2	3
Critical Range	5.185	5.442

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	56.881	8	2
A	54.730	8	3
A	53.884	8	1

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	1 2 3

Number of Observations Read 24

Number of Observations Used 24

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	1594.256637	531.418879	16.40	<.0001
Error	20	648.105158	32.405258		
Corrected Total	23	2242.361796			

R-Square	Coeff Var	Root MSE	Y Mean
0.710972	11.57211	5.692562	49.19208

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	1565.096504	1565.096504	48.30	<.0001
X2	2	29.160133	14.580067	0.45	0.6440

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	1565.096504	1565.096504	48.30	<.0001
X2	2	29.160133	14.580067	0.45	0.6440

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 20

Error Mean Square 32.40526

Number of Means 2

Critical Range 4.848

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	57.268	12	2
B	41.117	12	1

The GLM Procedure

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	20	
Error Mean Square	32.40526	
Number of Means	2	3
Critical Range	5.937	6.232

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	50.314	8	3
A	49.569	8	2
A	47.694	8	1

Class Level Information

Class	Levels	Values
X1	2	1 2
X2	3	1 2 3

Number of Observations Read	24
Number of Observations Used	24

Dependent Variable: Y

Sum of

Source	DF	Squares	Mean Square	F Value	Pr > F
Model	3	95.2018000	31.7339333	1.96	0.1527
Error	20	324.0203833	16.2010192		
Corrected Total	23	419.2221833			

R-Square	Coeff Var	Root MSE	Y Mean
0.227092	7.850563	4.025049	51.27083

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	77.04166667	77.04166667	4.76	0.0413
X2	2	18.16013333	9.08006667	0.56	0.5797

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	77.04166667	77.04166667	4.76	0.0413
X2	2	18.16013333	9.08006667	0.56	0.5797

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	20
Error Mean Square	16.20102
Number of Means	2
Critical Range	3.428

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	53.063	12	2
B	49.479	12	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	20
Error Mean Square	16.20102

Number of Means	2	3
Critical Range	4.198	4.407

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	52.393	8	3
A	51.148	8	2
A	50.273	8	1

Class Level Information

Class	Levels	Values
X1	5	0 10 11 30 31
X2	3	3 4 7
X3	11	50 51 52 53 54 55 56 57 58 59 60

Number of Observations Read	143
Number of Observations Used	143

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	16	27431.97521	1714.49845	324.59	<.0001
Error	126	665.53284	5.28201		
Corrected Total	142	28097.50805			

R-Square	Coeff Var	Root MSE	Y Mean
0.976313	11.96471	2.298262	19.20867

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	4	27.95202	6.98800	1.32	0.2650
X2	2	0.09188	0.04594	0.01	0.9913
X3	10	27403.93131	2740.39313	518.82	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	4	26.36848	6.59212	1.25	0.2941
X2	2	0.09188	0.04594	0.01	0.9913
X3	10	27403.93131	2740.39313	518.82	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007
Harmonic Mean of Cell Sizes	27.5

NOTE: Cell sizes are not equal.

Number of Means	2	3	4	5
Critical Range	1.227	1.291	1.334	1.365

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	20.1664	22	11
B A	19.2558	33	31
B A	19.0836	33	0
B A	19.0345	22	10
B	18.7642	33	30

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007
Harmonic Mean of Cell Sizes	45

NOTE: Cell sizes are not equal.

Number of Means	2	3
Critical Range	0.959	1.009

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	19.2925	55	7
A	19.2407	55	4
A	19.0155	33	3

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	1.784	1.878	1.940	1.986	2.021	2.050	2.074	2.094	2.111	2.127

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	44.4908	13	50
B	38.8285	13	51

	C	31.6808	13	52
	D	27.9938	13	53
	E	20.7708	13	54
	F	15.4385	13	55
	G	9.7946	13	56
	H	6.8569	13	57
I	H	5.2992	13	58
I	H	5.2492	13	59
I		4.8923	13	60

Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3
X3	11	1 2 3 4 7 11 15 25 30 60 120

Number of Observations Read 66

Number of Observations Used 66

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	12	38400.84858	3200.07072	21.03	<.0001
Error	53	8063.45543	152.14067		
Corrected Total	65	46464.30401			

R-Square	Coeff Var	Root MSE	Y Mean
0.826459	16.22486	12.33453	76.02242

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	22166.52502	22166.52502	145.70	<.0001
X2	1	2092.21882	2092.21882	13.75	0.0005
X3	10	14142.10475	1414.21047	9.30	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.00000	.	.	.
X2	1	2092.21882	2092.21882	13.75	0.0005
X3	10	14142.10475	1414.21047	9.30	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 53

Error Mean Square 152.1407

Harmonic Mean of Cell Sizes 29.33333

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 6.460

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	88.981	44	5
B	50.105	22	4

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 53

Error Mean Square 152.1407

Number of Means 2 3

Critical Range 7.460 7.846

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	95.877	22	2
B	82.085	22	3
C	50.105	22	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	53
Error Mean Square	152.1407

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	14.28	15.02	15.51	15.87	16.14	16.35	16.53	16.68	16.81	16.92

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	95.443	6	1
A	92.033	6	2
A	89.415	6	3
A	88.283	6	4
B A	84.803	6	7
B A	81.098	6	11
B C	72.548	6	15
C	59.363	6	25
C	57.778	6	30
C	57.760	6	60
C	57.720	6	120

Table B.26. Encapsulation of OLE by DE during storage.

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Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	66
Number of Observations Used	66

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	24258.74384	12129.37192	34.41	<.0001
Error	63	22205.56017	352.46921		
Corrected Total	65	46464.30401			

R-Square	Coeff Var	Root MSE	Y Mean
0.522094	24.69556	18.77416	76.02242

Sourc	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	24258.74384	12129.37192	34.41	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	24258.74384	12129.37192	34.41	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	63
Error Mean Square	352.4692

Number of Means	2	3
Critical Range	11.31	11.90

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	95.877	22	2
B	82.085	22	3
C	50.105	22	1

Class Level Information

Class	Levels	Values
X1	2	1 2

Number of Observations Read 8

Number of Observations Used 8

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	1	15.23520000	15.23520000	22.27	0.0033
Error	6	4.10415000	0.68402500		
Corrected Total	7	19.33935000			

R-Square	Coeff Var	Root MSE	Y Mean
0.787782	0.884341	0.827058	93.52250

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	15.23520000	15.23520000	22.27	0.0033

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	1	15.23520000	15.23520000	22.27	0.0033

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 6

Error Mean Square 0.684025

Number of Means 2

Critical Range 1.431

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	94.9025	4	1
B	92.1425	4	2

Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	190.6585333	95.3292667	73.55	0.0028
Error	3	3.8884000	1.2961333		
Corrected Total	5	194.5469333			

R-Square	Coeff Var	Root MSE	Y Mean
0.980013	1.192832	1.138479	95.44333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	189.2896333	189.2896333	146.04	0.0012
X2	1	1.3689000	1.3689000	1.06	0.3797

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.00000000	.	.	.
X2	1	1.36890000	1.36890000	1.06	0.3797

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 1.296133

Harmonic Mean of Cell Sizes 2.666667

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 3.138

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	99.4150	4	5
B	87.5000	2	4

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	3
Error Mean Square	1.296133

Number of Means	2	3
Critical Range	3.623	3.635

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	100.000	2	2
A	98.830	2	3
B	87.500	2	1

Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Souce	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	569.0949333	284.5474667	114.07	0.0015
Error	3	7.4836000	2.4945333		
Corrected Total	5	576.5785333			

R-Square	Coeff Var	Root MSE	Y Mean
0.987021	1.716127	1.579409	92.03333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	564.1665333	564.1665333	226.16	0.0006
X2	1	4.9284000	4.9284000	1.98	0.2545

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.00000000	.	.	.
X2	1	4.92840000	4.92840000	1.98	0.2545

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 2.494533

Harmonic Mean of Cell Sizes 2.666667

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 4.353

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	98.890	4	5
B	78.320	2	4

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	3
Error Mean Square	2.494533

Number of Means	2	3
Critical Range	5.026	5.043

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	100.000	2	2
A	97.780	2	3
B	78.320	2	1

Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Surce	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	771.8923000	385.9461500	70.95	0.0030
Error	3	16.3182500	5.4394167		
Corrected Total	5	788.2105500			

R-Square	Coeff Var	Root MSE	Y Mean
0.979297	2.608349	2.332256	89.41500

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	742.2987000	742.2987000	136.47	0.0013
X2	1	29.5936000	29.5936000	5.44	0.1019

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.00000000	.	.	.
X2	1	29.59360000	29.59360000	5.44	0.1019

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 5.439417

Harmonic Mean of Cell Sizes 2.666667

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 6.428

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	97.280	4	5
B	73.685	2	4

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square	5.439417	
Number of Means	2	3
Critical Range	7.422	7.447

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	100.000	2	2
A	94.560	2	3
B	73.685	2	1

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Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	797.1092333	398.5546167	69.29	0.0031
Error	3	17.2557000	5.7519000		
Corrected Total	5	814.3649333			

R-Square	Coeff Var	Root MSE	Y Mean
0.978811	2.716608	2.398312	88.28333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	759.2252083	759.2252083	132.00	0.0014

X2	1	37.8840250	37.8840250	6.59 0.0828

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.00000000	.	.	.
X2	1	37.88402500	37.88402500	6.59	0.0828

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 5.7519

Harmonic Mean of Cell Sizes 2.666667

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 6.610

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	96.238	4	5
B	72.375	2	4

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 5.7519

Number of Means	2	3
Critical Range	7.633	7.658

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	99.315	2	2
A	93.160	2	3
B	72.375	2	1

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Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	633.0410333	316.5205167	54.67	0.0044
Error	3	17.3703000	5.7901000		
Corrected Total	5	650.4113333			

R-Square	Coeff Var	Root MSE	Y Mean
0.973293	2.837462	2.406263	84.80333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	463.3904083	463.3904083	80.03	0.0029
X2	1	169.6506250	169.6506250	29.30	0.0124

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.0000000	.	.	.
X2	1	169.6506250	169.6506250	29.30	0.0124

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 5.7901

Harmonic Mean of Cell Sizes 2.666667

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 6.632

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	91.018	4	5
B	72.375	2	4

The GLM Procedure

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 5.7901

Number of Means 2 3

Critical Range 7.658 7.683

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	97.530	2	2
B	84.505	2	3
C	72.375	2	1

Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	6508.484133	3254.242067	452.57	0.0002
Error	3	21.571750	7.190583		
Corrected Total	5	6530.055883			

R-Square	Coeff Var	Root MSE	Y Mean
0.996697	4.641059	2.681526	57.77833

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	6046.234133	6046.234133	840.85	<.0001
X2	1	462.250000	462.250000	64.29	0.0041

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.0000000	.	.	.
X2	1	462.2500000	462.2500000	64.29	0.0041

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 7.190583

Harmonic Mean of Cell Sizes 2.666667

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 7.391

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	80.225	4	5
B	12.885	2	4

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	3	
Error Mean Square	7.190583	
Number of Means	2	3
Critical Range	8.534	8.562

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	90.975	2	2
B	69.475	2	3
C	12.885	2	1

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Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3

Number of Observations Read	66
Number of Observations Used	66

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	24258.74384	12129.37192	34.41	<.0001
Error	63	22205.56017	352.46921		
Corrected Total	65	46464.30401			

R-Square	Coeff Var	Root MSE	Y Mean
0.522094	24.69556	18.77416	76.02242

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	22166.52502	22166.52502	62.89	<.0001
X2	1	2092.21882	2092.21882	5.94	0.0177

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	0	0.000000	.	.	.
X2	1	2092.218820	2092.218820	5.94	0.0177

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	63
Error Mean Square	352.4692
Harmonic Mean of Cell Sizes	29.33333

NOTE: Cell sizes are not equal.

Number of Means	2
Critical Range	9.797

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	88.981	44	5
B	50.105	22	4

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Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	63
Error Mean Square	352.4692

Number of Means	2	3
Critical Range	11.31	11.90

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	95.877	22	2
B	82.085	22	3
C	50.105	22	1

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Class Level Information

Class	Levels	Values
X1	5	0 10 11 30 31
X2	3	3 4 7
X3	11	50 51 52 53 54 55 56 57 58 59 60

Number of Observations Read	143
Number of Observations Used	143

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	16	27431.97521	1714.49845	324.59	<.0001
Error	126	665.53284	5.28201		
Corrected Total	142	28097.50805			

R-Square	Coeff Var	Root MSE	Y Mean
0.976313	11.96471	2.298262	19.20867

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	4	27.95202	6.98800	1.32	0.2650
X2	2	0.09188	0.04594	0.01	0.9913
X3	10	27403.93131	2740.39313	518.82	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	4	26.36848	6.59212	1.25	0.2941
X2	2	0.09188	0.04594	0.01	0.9913
X3	10	27403.93131	2740.39313	518.82	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007
Harmonic Mean of Cell Sizes	27.5

NOTE: Cell sizes are not equal.

Number of Means	2	3	4	5
Critical Range	1.227	1.291	1.334	1.365

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	20.1664	22	11
B A	19.2558	33	31
B A	19.0836	33	0
B A	19.0345	22	10
B	18.7642	33	30

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007
Harmonic Mean of Cell Sizes	45

NOTE: Cell sizes are not equal.

Number of Means	2	3
Critical Range	0.959	1.009

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	19.2925	55	7
A	19.2407	55	4
A	19.0155	33	3

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	1.784	1.878	1.940	1.986	2.021	2.050	2.074	2.094	2.111	2.127

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	44.4908	13	50

	B	38.8285	13	51
	C	31.6808	13	52
	D	27.9938	13	53
	E	20.7708	13	54
	F	15.4385	13	55
	G	9.7946	13	56
	H	6.8569	13	57
I	H	5.2992	13	58
I	H	5.2492	13	59
I		4.8923	13	60

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Class Level Information

Class	Levels	Values
X1	5	0 10 11 30 31
X2	3	3 4 7
X3	11	50 51 52 53 54 55 56 57 58 59 60

Number of Observations Read 143

Number of Observations Used 143

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	16	27431.97521	1714.49845	324.59	<.0001
Error	126	665.53284	5.28201		
Corrected Total	142	28097.50805			

R-Square	Coeff Var	Root MSE	Y Mean
0.976313	11.96471	2.298262	19.20867

Source	DF	Type I SS	Mean Square	F Value	Pr > F

X1	4	27.95202	6.98800	1.32	0.2650
X2	2	0.09188	0.04594	0.01	0.9913
X3	10	27403.93131	2740.39313	518.82	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	4	26.36848	6.59212	1.25	0.2941
X2	2	0.09188	0.04594	0.01	0.9913
X3	10	27403.93131	2740.39313	518.82	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	126
Error Mean Square	5.282007
Harmonic Mean of Cell Sizes	27.5

NOTE: Cell sizes are not equal.

Number of Means	2	3	4	5
Critical Range	1.227	1.291	1.334	1.365

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	20.1664	22	11
B A	19.2558	33	31
B A	19.0836	33	0
B A	19.0345	22	10
B	18.7642	33	30

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 126

Error Mean Square 5.282007

Harmonic Mean of Cell Sizes 45

NOTE: Cell sizes are not equal.

Number of Means	2	3
Critical Range	0.959	1.009

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X2
A	19.2925	55	7
A	19.2407	55	4
A	19.0155	33	3

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 126

Error Mean Square 5.282007

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	1.784	1.878	1.940	1.986	2.021	2.050	2.074	2.094	2.111	2.127

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	44.4908	13	50
B	38.8285	13	51
C	31.6808	13	52
D	27.9938	13	53

	E	20.7708	13	54
	F	15.4385	13	55
	G	9.7946	13	56
	H	6.8569	13	57
I	H	5.2992	13	58
I	H	5.2492	13	59
I		4.8923	13	60

Class Level Information

Class	Levels	Values
X1	2	4 5
X2	3	1 2 3
X3	11	1 2 3 4 7 11 15 25 30 60 120

Number of Observations Read	66
Number of Observations Used	66

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	12	38400.84858	3200.07072	21.03	<.0001
Error	53	8063.45543	152.14067		
Corrected Total	65	46464.30401			

R-Square	Coeff Var	Root MSE	Y Mean
0.826459	16.22486	12.33453	76.02242

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	1	22166.52502	22166.52502	145.70	<.0001
X2	1	2092.21882	2092.21882	13.75	0.0005
X3	10	14142.10475	1414.21047	9.30	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F

X1	0	0.00000	.	.	.
X2	1	2092.21882	2092.21882	13.75	0.0005
X3	10	14142.10475	1414.21047	9.30	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 53

Error Mean Square 152.1407

Harmonic Mean of Cell Sizes 29.33333

NOTE: Cell sizes are not equal.

Number of Means 2

Critical Range 6.460

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	88.981	44	5
B	50.105	22	4

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 53

Error Mean Square 152.1407

Number of Means 2 3

Critical Range 7.460 7.846

Means with the same letter are not significantly different.

Duncan Grouping Mean N X2

A	95.877	22	2
B	82.085	22	3
C	50.105	22	1

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	53
Error Mean Square	152.1407

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	14.28	15.02	15.51	15.87	16.14	16.35	16.53	16.68	16.81	16.92

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X3
A	95.443	6	1
A	92.033	6	2
A	89.415	6	3
A	88.283	6	4
B A	84.803	6	7
B A	81.098	6	11
B C	72.548	6	15
C	59.363	6	25
C	57.778	6	30
C	57.760	6	60
C	57.720	6	120

Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	190.6585333	95.3292667	73.55	0.0028
Error	3	3.8884000	1.2961333		
Corrected Total	5	194.5469333			

R-Square	Coeff Var	Root MSE	Y Mean
0.980013	1.192832	1.138479	95.44333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	190.6585333	95.3292667	73.55	0.0028

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	190.6585333	95.3292667	73.55	0.0028

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	3	
Error Mean Square	1.296133	
Number of Means	2	3
Critical Range	3.623	3.635

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	100.000	2	2
A			

A	98.830	2	3
B	87.500	2	1

Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	569.0949333	284.5474667	114.07	0.0015
Error	3	7.4836000	2.4945333		
Corrected Total	5	576.5785333			

R-Square	Coeff Var	Root MSE	Y Mean
0.987021	1.716127	1.579409	92.03333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	569.0949333	284.5474667	114.07	0.0015

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	569.0949333	284.5474667	114.07	0.0015

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	3	
Error Mean Square	2.494533	
Number of Means	2	3
Critical Range	5.026	5.043

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	100.000	2	2
A	97.780	2	3
B	78.320	2	1

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Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read 6

Number of Observations Used 6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	771.8923000	385.9461500	70.95	0.0030
Error	3	16.3182500	5.4394167		
Corrected Total	5	788.2105500			

R-Square	Coeff Var	Root MSE	Y Mean
0.979297	2.608349	2.332256	89.41500

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	771.8923000	385.9461500	70.95	0.0030

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	771.8923000	385.9461500	70.95	0.0030

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 3

Error Mean Square 5.439417

Number of Means 2 3

Critical Range 7.422 7.447

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	100.000	2	2
A	94.560	2	3
B	73.685	2	1

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Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read 6

Number of Observations Used 6

The GLM Procedure

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	797.1092333	398.5546167	69.29	0.0031
Error	3	17.2557000	5.7519000		
Corrected Total	5	814.3649333			

R-Square	Coeff Var	Root MSE	Y Mean
0.978811	2.716608	2.398312	88.28333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	797.1092333	398.5546167	69.29	0.0031

Source	DF	Type III SS	Mean Square	F Value	Pr > F

X1	2	797.1092333	398.5546167	69.29	0.0031

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the exrimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	3	
Error Mean Square	5.7519	
Number of Means	2	3
Critical Range	7.633	7.658

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	99.315	2	2
A	93.160	2	3
B	72.375	2	1

Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

The GLM Procedure

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	633.0410333	316.5205167	54.67	0.0044
Error	3	17.3703000	5.7901000		
Corrected Total	5	650.4113333			

R-Square	Coeff Var	Root MSE	Y Mean
0.973293	2.837462	2.406263	84.80333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	633.0410333	316.5205167	54.67	0.0044

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	633.0410333	316.5205167	54.67	0.0044

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	3	
Error Mean Square	5.7901	
Number of Means	2	3
Critical Range	7.658	7.683

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	97.530	2	2
B	84.505	2	3
C	72.375	2	1

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Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F

Model	2	6316.436633	3158.218317	470.75	0.0002
Error	3	20.126700	6.708900		
Corrected Total	5	6336.563333			

R-Square	Coeff Var	Root MSE	Y Mean
0.996824	4.363223	2.590154	59.36333

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	6316.436633	3158.218317	470.75	0.0002

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	6316.436633	3158.218317	470.75	0.0002

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05	
Error Degrees of Freedom	3	
Error Mean Square	6.7089	
Number of Means	2	3
Critical Range	8.243	8.270

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	92.985	2	2
B	69.595	2	3
C	15.510	2	

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Class Level Information

Class	Levels	Values
X1	3	1 2 3

Number of Observations Read	6
Number of Observations Used	6

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	6485.287300	3242.643650	451.08	0.0002
Error	3	21.565700	7.188567		
Corrected Total	5	6506.853000			

R-Square	Coeff Var	Root MSE	Y Mean
0.996686	4.645097	2.681150	57.72000

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	2	6485.287300	3242.643650	451.08	0.0002

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	2	6485.287300	3242.643650	451.08	0.0002

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	3
Error Mean Square	7.188567
Number of Means	2 3

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	90.800	2	2
B	69.475	2	3
C	12.885	2	1

Class Level Information

Class	Levels	Values
X1	11	1 2 3 4 7 11 15 25 30 60 120

Number of Observations Read 22

Number of Observations Used 22

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	10	301.6658860	30.1665886	1620.41	<.0001
Eror	11	0.2047827	0.0186166		
Corrected Total	21	301.8706687			

R-Square	Coeff Var	Root MSE	Y Mean
0.999322	0.142309	0.136443	95.87774

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	10	301.6658860	30.1665886	1620.41	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	10	301.6658860	30.1665886	1620.41	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 11

Error Mean Square 0.0186

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	.3003	.3141	.3224	.3277	.3314	.3340	.3358	.3370	.3378	.3383

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	100.0000	2	1
A	100.0000	2	2
A	100.0000	2	3
B	99.3103	2	4
C	97.5287	2	7
D	96.9540	2	11

E	95.2874	2	15
F	92.9885	2	25
G	90.9770	2	30
G	90.8046	2	60
G	90.8046	2	120

Class Level Information

Class	Levels	Values
X1	11	1 2 3 4 7 11 15 25 30 60 120

Number of Observations Read 22

Number of Observations Used 22

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	10	301.6658860	30.1665886	1620.41	<.0001
Error	11	0.2047827	0.0186166		
Corrected Total	21	301.8706687			

R-Square	Coeff Var	Root MSE	Y Mean
0.999322	0.142309	0.136443	95.87774

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	10	301.6658860	30.1665886	1620.41	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	10	301.6658860	30.1665886	1620.41	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 11

Error Mean Square 0.018617

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	.3003	.3141	.3224	.3277	.3314	.3340	.3358	.3370	.3378	.3383

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	100.0000	2	1
A	100.0000	2	2
A	100.0000	2	3
B	99.3103	2	4
C	97.5287	2	7
D	96.9540	2	11
E	95.2874	2	15
F	92.9885	2	25
G	90.9770	2	30
G	90.8046	2	60
G	90.8046	2	120

Class Level Information

Class	Levels	Values
X1	11	1 2 3 4 7 11 15 25 30 60 120

Number of Observations Read 22

Number of Observations Used 22

Dependent Variable: Y

		Sum of			
Model	10	2962.681445	296.268145	436.82	<.0001
Error	11	7.460700	0.678245		
Corrected Total	21	2970.142145			

R-Square	Coeff Var	Root MSE	Y Mean
0.997488	1.003292	0.823557	82.08545

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	10	2962.681445	296.268145	436.82	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	10	2962.681445	296.268145	436.82	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha	0.05
Error Degrees of Freedom	11
Error Mean Square	0.678245

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	1.813	1.896	1.946	1.978	2.000	2.016	2.027	2.034	2.039	2.042

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1
A	98.8300	2	1
A	97.7800	2	2
B	94.5600	2	3
B	93.1600	2	4
C	84.5050	2	7
D	79.8250	2	11
E	76.1400	2	15
F	69.5950	2	25
F	69.5950	2	60
F	69.4750	2	30
F	69.4750	2	120

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Class Level Information

Class	Levels	Values
X1	11	1 2 3 4 7 11 15 25 30 60 120

Number of Observations Read 22

Number of Observations Used 22

Dependent Variable: Y

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	10	18759.08000	1875.90800	118.64	<.0001
Error	11	173.93261	15.81206		
Corrected Total	21	18933.01261			

R-Square	Coeff Var	Root MSE	Y Mean
0.990813	7.936174	3.976438	50.10522

Source	DF	Type I SS	Mean Square	F Value	Pr > F
X1	10	18759.08000	1875.90800	118.64	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
X1	10	18759.08000	1875.90800	118.64	<.0001

Duncan's Multiple Range Test for Y

NOTE: This test controls the Type I comparisonwise error rate, not the experimentwise error rate.

Alpha 0.05

Error Degrees of Freedom 11

Error Mean Square 15.81206

Number of Means	2	3	4	5	6	7	8	9	10	11
Critical Range	8.752	9.154	9.395	9.552	9.659	9.733	9.785	9.821	9.844	9.859

Means with the same letter are not significantly different.

Duncan Grouping	Mean	N	X1

	A	87.500	2	1
	B	78.318	2	2
C	B	73.688	2	3
C	B	72.377	2	4
C	B	72.377	2	7
C		66.512	2	11
	D	46.219	2	15
	E	15.509	2	25
	E	12.886	2	30
	E	12.886	2	60
	E	12.886	2	120

CURRICULUM VITAE

PERSONAL INFORMATION

Surname, Name	: Betül, ÇİLEK TATAR
Nationality	: Turkish (TC)
Date and Place of Birth	: 14 February 1988, Ankara
Phone	: +90 537 619 58 88
E-mail	: bcot0408@gmail.com

EDUCATION

Degree	Institution	Year of Graduation
MS	METU Food Engineering	2012
BS	METU Food Engineering	2010
High School	Gazi Anadolu High School, Ankara	2005

WORK EXPERIENCE

Year	Place	Enrollment
2011-Present	METU Department of Food Eng.	Research Assistant
2009 August	Anadolu Efes Brewery and Malting Inds.	Intern Engineering Student
2008 August	Ankara Sugar Factory	Intern Engineering Student

FOREIGN LANGUAGES

Advanced English, Basic German

PUBLICATIONS

1. Cilek Tatar, B., Sumnu, G., & Oztop, M. (2018). Microcapsule characterization of phenolic powder obtained from strawberry pomace. (accepted from Journal of Food Processing and Preservation)

2. Cilek Tatar, B., Sumnu, G., & Sahin, S. (2017). Rheology of Emulsions. In J. Ahmed, P. Ptaszek, & S. Basu (Eds.), Advances in Food Rheology and Its Applications. Oxford: Woodhead Publishing Ltd. (Book Chapter)

3. Kirtil, E., Tonyali, B., Aydogdu, A., Yildiz Bulut, E., Cilek Tatar, B., & Oztop, M. (2017). Visualization of cakes differing in oil content with magnetic resonance imaging. International Journal of Food Properties, doi: 10.1080/10942912.2017.1326057.

4. Cilek Tatar, B., Sumnu, G., Oztop, M., & Ayaz, E. (2016). Effects of centrifugation, encapsulation method and different coating materials on the total antioxidant activity of the microcapsules of powdered cherry laurels. International Journal of Nutrition and Food Engineering, 10 (12): 901-904.

5. Luca, A., Cilek, B., Hasırcı, V., Sahin, S., & Sumnu, G. (2014). Storage and baking stability of encapsulated sour cherry phenolic compounds prepared from micro- and nano-suspensions. Food and Bioprocess Technology, 7, 204-211.

6. Luca, A., Cilek, B., Hasırcı, V., Sahin, S., & Sumnu, G. (2013). Effect of degritting of phenolic extract from sour cherry pomace on encapsulation efficiency: Production of nano-suspension. Food and Bioprocess Technology, 6, 2494-2502.

7. Cilek, B., Luca, A., Hasırcı, V., Sahin, S., & Sumnu, G. (2012). Microencapsulation of phenolic compounds extracted from sour cherry pomace: Effect of formulation, ultrasonication time and core to coating ratio. European Food Research and Technology, 235(4), 587-596.

8. Çilek, B. (2012). Microencapsulation of Phenolic Compounds Extracted from Sour Cherry (Cerasus prunus L.) Pomace (Unpublished master's thesis). Middle East Technical University, Ankara, Turkey.

9. Cilek Tatar, B., Sumnu, G., Oztop, M., & Ayaz, E. Characterization of encapsulated cherry laurel (Laurocerasus officinalis Roem.) powder. (submitted to LWT – Food Science and Technology)

10. Cilek Tatar, B., Sumnu, G., & Oztop, M. Effects of temperature and storage conditions on the total phenolic content and antioxidant activity of microcapsules including strawberry phenolics. (in progress)

<u>ORAL and POSTER PRESENTATIONS</u>

11. Cilek Tatar, B., Sumnu, G., Oztop, M. (2018, June). Encapsulation of olive leaf extract by double emulsion. Poster session will be presented at the 3rd Food Structure and Functionality Forum Symposium and 3rd IDF Symposium on Microstructure of Dairy Products, Montreal, Canada.

12. Cilek Tatar, B., Sumnu, G., Oztop, M. (2018, April). Characterization of olive leaf extract containing double emulsions using NMR Relaxometry. Poster session presented at the International Eurasian Conference on Biological and Chemical Sciences, Ankara, Turkey.

13. Cilek Tatar, B., Sumnu, G., Oztop, M. (2018, April). Microcapsule characterization of phenolic powder obtained from strawberry pomace. Poster session presented at the International Conference on Raw Materials to Processed Foods, Antalya, Turkey.

14. Cilek Tatar, B., Sumnu, G., Oztop, M. (2018, April). Effects of temperature and storage conditions on the total phenolic content and antioxidant activity of microcapsules including strawberry phenolics. Poster session presented at the International Conference on Raw Materials to Processed Foods, Antalya, Turkey.

15. Cilek Tatar, B., Sumnu, G., Oztop, M. (2017, November). Encapsulation of phenolic compounds: effects of high pressure homogenization and different coating materials. Poster session presented at the 10th Food Engineering Congress, Antalya, Turkey.

16. Cilek Tatar, B., Sumnu, G., Oztop, M., & Ayaz, E. (2016, December). Effects of Centrifugation, Encapsulation Method and Different Coating Materials on the Total Antioxidant Activity of the Microcapsules of Powdered Cherry Laurels. Oral presentation at the 18th International Conference on Food Science and Components, Hong Kong, China.

17. Cilek Tatar, B., Ayaz, E., Sahin, S., & Sumnu, G. (2016, February). Investigation of the Effects of Different Gum Types and Concentrations on the Encapsulation of Cherry Laurel Powders. Poster session presented at the 2nd Food Structure and Functionality Forum Symposium, From Molecules to Functionality, Singex, Singapore.

18. Sumnu, G., Cilek Tatar B., Luca, A., Hasırcı, V., & Sahin, S. (2014, September). Bioaccessibility of the Capsules Prepared from Micro- and Nano-Suspensions of Sour Cherry Pomace Extract Containing Phenolic Compounds. Poster session presented at the XXII International Conference on Bioencapsulation, 21th Bratislava International Conference on Macromolecules, Bratislava, Slovakia.

19. Luca, A., Cilek, B., Hasırcı, V., Sahin, S., & Sumnu, G. (2013, September). Storage and baking stability of encapsulated sour cherry phenolics prepared from micro and nano-suspensions. Poster session presented at the 19th International Symposium on Microencapsulation, Pamplona, Spain.

20. Cilek, B., Luca, A., Hasırcı, V., Sahin, S., Sumnu, G. (2012, October). Characterization of microcapsules containing phenolic compounds extracted from sour cherry pomace. Poster session presented at the 11th Turkey Food Congress, Hatay, Turkey.

21. Luca, A., Cilek, B., Sumnu, G., Sahin, S., Hasırcı, V. (2012, October). Effect of purification of cherry pomace extract on encapsulation efficiency: preparation of nanoemulsion and microemulsion. Poster session presented at the 11th Turkey Food Congress, Hatay, Turkey.

22. Sahin, S., Cilek, B., Luca, A., Hasırcı, V., Sumnu, G. (2012, April). Microencapsulation of phenolic compounds extracted from sour cherry pomace. Poster session presented at the 6th International Symposium on Food Rheology and Structure. Zurich, Switzerland.

23. Sumnu, G., Luca, A., Cilek, B., Hasırcı, V., Sahin, S. (2012, April). Role of purification of extract of sour cherry pomace on the encapsulation efficiency: nanoemulsion and microemulsion production. Poster session presented at the 6th International Symposium on Food Rheology and Structure. Zurich, Switzerland.

HOBBIES

Skiing, Off-road motor sports