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**Faculty of Graduate Studies**

# **Rheological Properties for Olive Oil in Palestine**

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the Degree of Master of Physics, Faculty of Graduate Studies, An-  
Najah National University - Nablus, Palestine.**

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## Rheological Properties for Olive Oil in Palestine

By

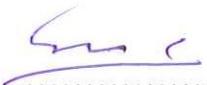
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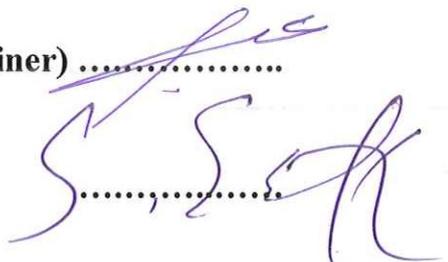
  
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III

**Dedication**

*This thesis is dedicated to my parents, as well as,  
to my brothers and sisters.*

*With respect and love.*

## **Acknowledgement**

I would like to express my sincere appreciation to my supervisors, Prof. Dr. Issam Rashid Abdelraziq for his helpful comments and continual encouragement, and Dr. Sharif Mohammed Musameh, for his cooperation which helped me in the completion of this research.

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## الإقرار

أنا الموقع أدناه مقدم الرسالة التي تحمل العنوان:

### **Rheological Properties for Olive Oil in Palestine**

أقر بأن ما اشتملت عليه هذه الرسالة إنما هو نتاج جهدي الخاص، باستثناء ما تمت الإشارة إليه حيثما ورد، وان هذه الرسالة ككل أو جزء منها لم يقدم من قبل لنيل أية درجة أو بحث علمي أو بحثي لدى أية مؤسسة تعليمية أو بحثية أخرى .

#### **Declaration**

The work provided in this thesis, unless otherwise referenced, is the researcher's own work, and has not been submitted elsewhere for any other degree or qualification.

**Student's name:**

اسم الطالب:

**Signature:**

التوقيع:

**Date:**

التاريخ:

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**List of Abbreviations**

AAD%	Percentage of Average Absolute Deviation
ANOVA	Analysis of Variance
CGS	Centimeter-Gram-Second
cP	Centipoise
cSt	Centistokes
E <sub>a</sub>	Activation Energy
Eq.	Equation
EVOO	Extra Virgin Olive Oil
VOO	Virgin Olive Oil
FFA	Free Fatty Acids
Fig.	Figure
IOOC	International Olive Oil Council
N	Newton
P	poise
Pa	Pascal
P-value	Probability
R	Gas Constant
RPM	Revolution Per Minute
SD	Standard Deviation
SI	Système International d' Unités
SMC	Spindle Multiplier Constant
SP	Spindle
T	Temperature in Kelvin
L <sub>1</sub>	Saida Region
L <sub>2</sub>	Allar Region
L <sub>3</sub>	Beit Lid Region
L <sub>4</sub>	NazlatIssa Region
L <sub>5</sub>	Meithaloon Region
L <sub>6</sub>	Jeet Region
L <sub>7</sub>	Ti'innik Region
L <sub>8</sub>	Yasid Region
L <sub>9</sub>	Borqa Region
L <sub>10</sub>	Arraba Region
L <sub>11</sub>	Asira Region
L <sub>12</sub>	Beta Region
L <sub>13</sub>	Jenin Region
$\rho$	Density
$\eta$	Dynamic Viscosity
$\nu$	Kinematic Viscosity
$\eta_{cal}$	Calculated Dynamic Viscosity
$\eta_{exp}$	Measured Dynamic Viscosity

XVI  
**Rheological Properties for Olive Oil in Palestine**  
**By**  
**Ahmad Mustafa Bahti**  
**Supervisor**  
**Prof. Issam Rashid Abdelraziq**  
**Co - Supervisor**  
**Dr. Sharif Mohammad Musameh**

**Abstract**

In this study, olive oil samples of different storage ages and regions in Palestine were studied. The density, refractive index, acidity and viscosity of the samples were measured. The refractive index of the olive oil samples were studied against storage ages and results showed that the refractive index decreases as a function of storage age. The acidity of olive oil samples from different regions and different crops showed that acidity increases as a function of storage age. Most of olive oil samples (storage age  $\leq 12$  years) acidity did not exceed the international quality standards ( $< 3.3\%$ ). It is worth noting that olive oil can be stored until 12 years without exceeding the international quality standards of acidity in proper conditions.

The viscosity of olive oil samples of 2012 crop from different regions was studied, and the results showed that most of the olive oil samples are classified to be extra - virgin.

Two and three constant equations were proposed to obtain more suitable prediction of temperature dependence of dynamic viscosity of olive oil.

The experimental results of viscosity were compared with the power law equation, and the behavior of olive oil was found to be Newtonian.

# **Chapter One**

## **Introduction**

### **1.1 Olive Oil**

Olive oil is a fat obtained from the olive fruit by mechanical or chemical means. Olive oil is commonly used in cooking, cosmetics, pharmaceuticals, soaps, and as a fuel for traditional oil lamps. Olive oil is used throughout the world, but especially in the Mediterranean countries and, in particular, in Greece, which has the highest consumption per person (NAOOA, 2013).

Olives are very important for the Palestinian, not only because they are the biggest crop in what remains a largely agricultural economy, but also for their deep cultural significance as a symbol of traditional society and ties to the land. It is estimated that olive trees account for nearly 45 percent of cultivated land in Palestine and in good years can contribute as much as 15 - 19 percent of agriculture output. Given that agriculture accounts for nearly 25 percent of gross domestic product, olives are an important element of the Palestinian economy and estimates suggest that about 100,000 families depend to some extent upon the olive harvest for their livelihoods (The World Bank, 2012).

### **1.2 Previous Studies**

Vegetable oils have become increasingly important for nutritional purposes and in a wide range of industrial applications which include fuels, skin care products, high pressure lubricants and alkyd resins for paint. These

applications require extensive studies on the physic - chemical properties of oils in order to ascertain their suitability as raw materials. Such properties include viscosity and acidity which are an important parameters in the design of process equipment for oils (Eromosele, and Paschal,2003). (Nierat *et al.*, 2012).

In 1988, a model was used by Patil and his group to describe the liquid - liquid thermal hypothesis of vegetable oils. Extensive data on hydrolysis equilibrium and rate have been obtained (Patil *et al.*, 1988).

Noureddin and his group have presented the range of temperatures in which the viscosity and temperature of vegetable oils are correlated (Noureddini *et al.*, 1992). Van Wazer and his group discussed the sensitivity of the eye in judging viscosity of Newtonian liquids (James,1996).

Studies by Bayrak on vegetable oils determined the relationship between viscosity and average molecular weight (Bayrak *et al.*, 1997). Hsieh predicted viscosity of vegetable oils from density data (Hsieh *et al.*, 1999). The palm oil was proved to be a good diesel - generator fuel by Almeidaa and his group, they found that the performance of diesel generator is increased by increasing the palm oil temperature (Almeidaa *et al.*, 2002).

In a given range of temperatures, Farhoosh found that the natural logarithm of the kinetic rate of five different vegetable oils varies linearly with respect to temperature (Farhoosh *et al.*, 2008).

In his study, Ahmad evaluated the viscosity changes of vegetable oils, and fitted the viscosity with well - known rheological equations (Ahmad *et al.*, 2009). He identified model limitation through graphical and numerical

observations. Vegetable oils were subjected to viscometer measurements of viscosity at shear rate (3 - 100 RPM) and temperature (40 - 100°C).

Stanciu proposed four relationships of dynamic viscosity temperature dependence for vegetable oils. In his studies he found a polynomial or exponential dependence between temperature and dynamic viscosity of vegetable oil using the Andrade's equation changes (Stanciu, 2012).

Adnan and his group studied the characterization of different oils and their rheological properties. Eight different natural oils, namely olive, coconut, almond, castor, sesame, cotton seed, sunflower, and paraffin oils. All the oils investigated were found to possess Non - Newtonian behavior (Adnan *et al.*, 2009).

Effect of fatty acid composition on dynamic and steady shear rheology of oils was studied by Hasan Yalcin and his group (Yalcin *et al.*, 2012).

Studies in New Zealand by Sims indicated that vegetable oils, particularly rapeseed oil, could be used as a replacement for diesel fuel (Sims *et al.*, 1981).

Reid and his group evaluated the chemical and physical properties of 14 vegetable oils. These injection studies pointed out that the oils behave very differently from petroleum - based fuels (Reid *et al.*, 1989).

Goering and his group studied the characteristic properties of eleven vegetable oils to determine which oil would be the best suited for use as an alternative fuel source (Goering *et al.*, 1981).

Bruwer and his group studied the use of sunflower seed oil as a renewable energy source. When operating tractors with 100% sunflower oil instead of

diesel fuel, an 8% power loss occurred after 1000 hours of operation (Bruwer *et al.*, 1981).

Viscosity of oil samples used by Fasina and Colley was shown to decrease with temperature, in the same study they found that the specific heat capacity increases with increasing temperature (Fasina and Colley, 2008).

In their work, Toscano and his group investigated the effects of two analytical chemical parameters, their results confirmed the incidence of molecular characteristics of triglycerides of an oil with respect to its viscosity (Toscano *et al.*, 2012). The concentration of polyunsaturated fatty acids (PUFAs) was found to be a predominant parameter that influences the low - temperature properties of vegetable oil - based lubricants (Quinchia *et al.*, 2012).

Many researches concerned on olive oil as a vegetable oil, Lupi and his group prepared different samples of olive oil based organogels by using cocoa butter, dynamic temperature ramp tests, carried out at 5 °C/min, allowed the determination of rheological characteristics (Lupi *et al.*, 2012).

To study the influence of operative conditions adopted during the malaxation of pastes on the quality of resulting oils, Angerosa and his coworkers found that low temperatures and times, ranging between 30 and 45 min, according to the rheology of the olive pastes, were the optimal operative conditions for the malaxation (Angerosa *et al.*, 2001).

The relationship between density, viscosity, oil/water interfacial tension and structure of vegetable oils after heating at frying temperatures were studied by Adolfo. He aimed to explore the possibility of reusing waste

vegetable oils as solid agglomerants for different purposes. Commercial olive and sunflower oils were heated at 150 and 225 °C in the time interval of 1 – 15 days to achieve a wide range of alteration degrees. Structural changes in the oils were monitored, of the two vegetable oils studied, sunflower oil was found to be more sensitive to thermal treatment, undergoing greater changes in its properties, especially in viscosity, which may show a marked increase (Adolfo *et al.*, 2006).

### **1.3 Objectives of the Study**

- The goal of this work is to check whether olive oil in Palestine shows Newtonian or Non - Newtonian behavior.
- The physical properties (viscosity, acidity, density, refractive index) of olive oil in Palestine will be measured and compared with standard values.
- The viscosity of olive will be measure as a function of temperature.
- Equation of the behavior will be suggested to describe the rheological effect.
- The experimental data will be fitted by using SPSS and Excel programs to get the relationship of dynamic viscosity as a function of temperature.

### **1.4 Organization of the Thesis:**

Given below is a brief outline of the topics discussed in this thesis:

- 1- Introduction: the characteristics of olive oil are presented here, previous studies concerning the problems into account, and the objectives of the study.

- 2- Theoretical formulation: the theory of rheology, viscosity, stress and strain rate, acidity of oils, refractive index and mass density.
- 3- Methodology: the samples used in the research and the function of the equipments used in measurements.
- 4- Results and analysis of data obtained.
- 5- Discussion about different results obtained by analysis of measured data.

## Chapter Two

### Theoretical Formulation

#### 2.1 Rheology

Rheology is defined as the branch of physics that studies the deformation and flow of matter (Larson, 1999).

Rheology applies to substances which have a complex microstructure, such as muds, suspensions, polymers and other glass formers, it also applies to many foods and additives, bodily fluids and other biological materials or other materials which belong to the class of soft matter (Themelis,1995).

The role of rheology is important in the field of cosmetic science, especially in the field of emulsions and lotions (Martin *et al.*, 2004).

Since the different creams have different consistencies and they are used for long terms, the effects of different rheological parameters of oils are studied for understanding the performance of the system (Remington, 2006).

#### 2.2 Viscosity

Viscosity is a measure of the resistance to flow or shear. Viscosity can also be termed as a drag force and is a measurement of the frictional properties of the fluid. It can be expressed in two distinct forms:

- a. Dynamic viscosity ( $\eta$ )
- b. Kinematic viscosity ( $\nu$ )

Dynamic viscosity is defined as the ratio of shear stress (force over cross section area) to the rate of deformation (the difference of velocity over a sheared distance), and it is presented as:

$$\eta = \frac{\tau}{\frac{\partial u}{\partial x}} \quad (2.1)$$

Where,  $\eta$  is the dynamic viscosity in Pascal-second (Pa.s);  $\tau$  is shear stress ( $\text{N/m}^2$ ); and,  $\frac{\partial u}{\partial x} = \gamma$  is rate of deformation or velocity gradient or better

known as shear rate (1/s) (Dutt N. *et al*, 2007).

The Kinematic viscosity requires knowledge of mass density of the liquid ( $\rho$ ) at that temperature and pressure. It is defined as:

$$\nu = \frac{\eta}{\rho} \quad (2.2)$$

Where,  $\nu$  is kinematic viscosity in centistokes (cSt),  $\rho$  is in  $\text{g/cm}^3$  (Dutt N. *et al*, 2007).

The flow characteristics of liquids are mainly dependent on viscosity and are broadly divided into two categories:

- 1- Newtonian systems.
- 2- Non - Newtonian systems.

### **2.2.1 Newtonian Systems**

These fluids have the same viscosity at different shear rates (different revolution per minute) (rpm). These fluids are called Newtonian over the shear rate range they are measured. Water is an example of these fluids (James F., 1996).

### **2.2.2 Non - Newtonian Systems**

These fluids have different viscosity at different shear rates. They are classified into two groups:

### a) Time Independent

Time independent means that the viscosity behavior does not change as a function of time when it is measuring at a specific shear rate. Pseudoplastic materials such as lava, ketchup, whipped cream, and blood are examples of such fluids which display decrease in viscosity with an increase in shear rate. This type of fluid is known as "shear thinning".

### b) Time Dependent

Time Dependent means that the viscosity behavior changes as a function of time when measuring at a specific shear rate (the duration for which the fluid has been subjected to shearing as well as their previous kinematic history). A thixotropic material is an example of that fluid which has decreasing viscosity under constant shear rate. Many gels are classified to be thixotropic material (James, 1996).

In this study, the viscosity of different olive oil samples will be measured as a function of shear rates over a given shear rate range and at different temperatures.

The nature of liquids is complex, so there has been no comprehensive theory explaining the relationship between liquid viscosity and other properties, so empirical methods are used in addition to mathematical expressions to get the best fit of the experimental data.

## 2.3 The Dependence on Temperature

Clements and his group was the first who fitted the dependence of viscosity on temperature using the Arrhenius - type relationship which is given by:

$$\eta = \eta_{\infty,T} e^{\frac{E_a}{RT}} \quad (2.3)$$

Where  $\eta$  is the dynamic viscosity in Pa.s,  $\eta_{\infty,T}$  is the viscosity at infinite - temperature in Pa.s,  $E_a$  is the exponential constant that is known as activation energy (J/mol); R is the gas constant (J/mol.K) and T is the absolute temperature in Kelvin (Ahmad, 2009; Clements *et al.*, 2006).

When applied to real phenomena, equation (2.3) failed to provide good representation, so new models are needed.

### 2.3.1 The Andrade's Equation

The Andrade's equation is the simplest form of representation of liquid dynamic viscosity as a function of temperature, it takes the form:

$$\eta = Ae^{\frac{B}{T}} \quad (2.4)$$

Where  $\eta$  is the dynamic viscosity in cP, T is the temperature in Kelvin, A is a constant in cP, and B is a constant Celsius, A and B are characteristics of each substance (De Guzman, 1913; Andrade, 1930).

The constants of Andrade's equation were evaluated experimentally for a number of substances by several researchers such as Duhne, Dutt and his group, Visvwanath and Natarjan (Duhne, 1979; Dutt *et al.*, 2007; Natarajan *et al.*, 1989)

### 2.3.2 Abramovic's Equations

Abramovic proposed the following equations to describe the dependence of dynamic viscosity on temperature:

Abramovic's equation (formula 1):

$$\log \eta = \frac{A}{T} - B \quad (2.5)$$

Abramovic's equation (formula 2):

$$\eta = A - B \log t \quad (2.6)$$

Where  $\eta$  is the dynamic viscosity in cP, T is the temperature in Kelvin, t is the temperature in degrees Celsius. A and B are constants.

Table 2.1 shows the values of the constants of the above two equations for olive oil (Abramovic *et al.*, 1998)

**Table (2.1): Constants of equations 2.5 and 2.6**

Oil	$\log \eta = \frac{A}{T} - B$			$\eta = A - B \log t$		
	A (K)	B	$\eta$ (cP) at 298.15 K	A (cP)	B (cP)	$\eta$ (cP) at 25.15 °C
Olive oil	1558.2	3.433	62.12	235.4	124.1	61.59
Refined corn oil	1464.1	3.207	50.54	186.6	97.4	50.19
Refined sunflower oil	1443.3	3.157	48.29	177.2	92.3	47.93

### 2.3.3 Three Constant Andrade's Equation

Three constants equation was proposed by Andrade which is used by Abramovic. The equation has the following form :

$$\ln \eta = A + \frac{B}{T} + CT \quad (2.7)$$

Where  $\eta$  is the dynamic viscosity in cP, T is the temperature in Kelvin. A, B and C are constants (Vogel, 1921; Andrade, 1930; Abramovic, 1998).

Table (2.2) shows the values of the constants A, B, and C of equation 2.7 (Andrade, 1930; Abramovic *et al.*, 1998).

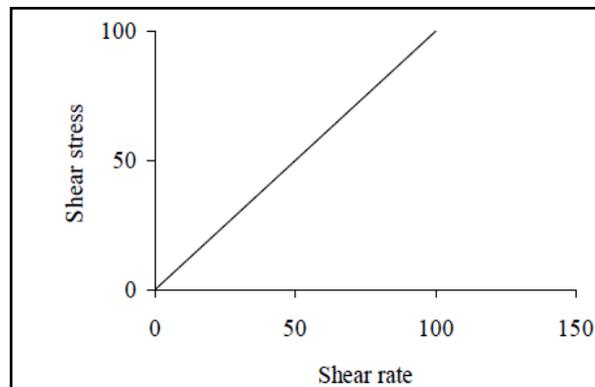
**Table (2.2): Constants of equations 2.7**

Oil	$\ln \eta = A + \frac{B}{T} + CT$			
	A	B (K)	C (K <sup>-1</sup> )	$\eta$ (cP) at 298.15 K
Olive oil	-32.72	7462.27	0.04	69.03
Refined corn oil	-27.89	6572.41	0.03	22.16
Refined sunflower oil	-28.09	6575.60	0.03	18.34

## 2.4 Models Regarding Stress and Strain Rate

### 2.4.1 Newtonian Fluid

Newtonian fluid means that when shear stress is plotted against shear rate at a given temperature, the plot shows a straight line with a constant slope that is independent of shear rate. (Fig.2.1)



**Fig. (2.1):** Flow curve of a Newtonian fluid at power law

The simplest constitutive equation is Newton's law of viscosity:

$$\tau = \eta \dot{\gamma} \quad (2.8)$$

where  $\eta$  is the Newtonian viscosity and  $\dot{\gamma}$  is the shear rate or the rate of strain. The Newtonian fluid is the basis for classical fluid mechanics. Gases, for example, exhibit characteristics of Newtonian viscosity.

### 2.4.2 Non - Newtonian Fluid Models

One of the most widely used forms of the general non - Newtonian constitutive relation is a power law model, which can be described as (Middleman, 1968; Munson *et al.*, 1998; Bird *et al.*, 1987):

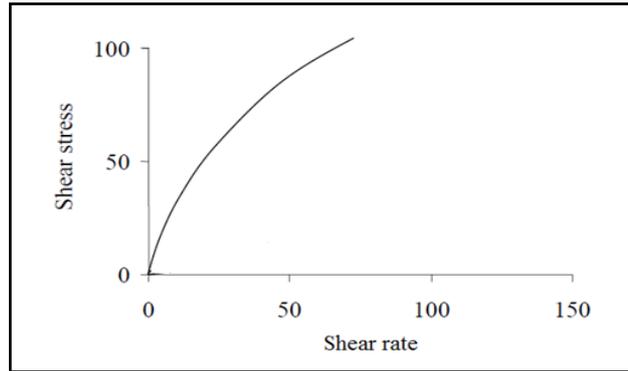
$$\tau = m\dot{\gamma}^n \quad (2.9)$$

Where  $\tau$  is stress and  $\dot{\gamma}$  is strain rate,  $m$  and  $n$  are power-law model constants. The constant,  $m$  is a measure of the consistency of the fluid with dimensions of  $\text{cP}\cdot(\text{s})^{n-1}$ , the higher the  $m$  is, the more viscous the fluid is.  $n$  is a measure of the degree of non - Newtonian behavior. The greater the departure from the unity, the more pronounced the non - Newtonian properties of the fluid are.

The viscosity for the power - law fluid can be expressed as (Middleman, 1968; Munson *et al.*, 1998; Bird *et al.*, 1987):

$$\eta = m\dot{\gamma}^{n-1} \quad (2.10)$$

Where  $\eta$  is non - Newtonian apparent viscosity, if  $n = 1$ , a Newtonian fluid is obtained. If  $n$  deviates from 1, a non - Newtonian fluid is obtained. (Fig.2.2).



**Fig.(2.2):** Flow curve of a Non - Newtonian fluid at power law

Herschel - Bulkley described the behavior of fluids by the following equation:

$$\begin{aligned} \tau &= m\dot{\gamma}^n + \tau_y, & \text{where } \tau \geq \tau_y \\ \tau &= 0, & \text{where } \tau \leq \tau_y \end{aligned} \quad (2.11)$$

Where  $\tau$  is stress and  $\dot{\gamma}$  is strain rate,  $m$  and  $n$  are model constants,  $\tau_y$  is a constant that is interpreted as yield stress.

The model shows both yield stress and shear - thinning non - Newtonian viscosity, and is used to describe the rheological behavior of food products and biological liquids (James F., 1996).

## 2.5 Olive Oil Acidity

The acidity of olive oil is effected by different parameters such as degree of ripeness, industrial processes employed for oil extraction, altitude, the cultivator, climate and other factors.

Olive oil is classified qualitatively according to its acidity into many classes as given in Table (2.3) (IOOC, 2000).

**Table (2.3): Classification of olive oil according to FFA%**

Category	FFA%
Extra virgin olive oil	$\leq 0.8$
Virgin olive oil	$\leq 2.0$
Ordinary virgin olive oil	$\leq 3.3$
Lampante oil	$> 3.3$

## 2.6 Refractive Index

Refractive index ( $n$ ) of a medium is defined as the ratio of the speed of light in a vacuum to the speed of light traveling through this medium, and mathematically it is written as:

$$n = \frac{c}{v} \quad (2.12)$$

Where  $c$  is the speed of light in vacuum and  $v$  is the speed of light in the substance. The refractive index for olive oil extends from 1.4677 to 1.4707 at 20 °C (IOOC, 2000).

## 2.7 Mass Density

Mass density is defined as the ratio of mass of the material in grams and the volume in  $\text{cm}^3$ . Robert has measured the density of olive oil to be 0.918  $\text{gm}/\text{cm}^3$  at 15 °C and at atmospheric pressure (Robert *et al.*, 1979).

## Chapter Three

### Methodology

Olive oil samples were collected from different region in Palestine, they were all produced by Palestinian industrial olive oil mills, from the crop of 1994 until the crop of 2012 at least four samples were collected from each region representing different olive oil ages.

The samples were collected from different regions, these are:

L<sub>1</sub>, L<sub>2</sub>, L<sub>3</sub>, L<sub>4</sub>, L<sub>5</sub>, L<sub>6</sub>, L<sub>7</sub>, L<sub>8</sub>, L<sub>9</sub>, L<sub>10</sub>, L<sub>11</sub>, L<sub>12</sub>, and L<sub>13</sub>. The samples were kept in closed glass bottles in dark place at 25 °C.

The viscosity of each olive oil sample was measured in wide range of temperatures extend from 8 °C to 73 °C. Each time acidity, refractive index, and mass density were measured.

The viscosity was measured using the ND - 1 rotational viscometer. The refractive index was measured using the refractometer. Chemical titration was used to measure the acidity, while temperature of the samples was measured by using Digital Prima Long Thermometer.

Measured data were analyzed and relationships between different parameters were studied. In addition the relationship between density and refractive index, refractive index and age, acidity and age, viscosity and age, viscosity and temperature, and density and age were plotted. The curves representing the relationship between viscosity and temperature were fitted using previously used equations. A comparison with experimental data was done. Moreover, new equations were suggested to fit the experimental data of viscosity versus temperature.

Finally, the relationship between viscosity and shear rate was studied to determine whether the olive oil samples under study are Newtonian or Non Newtonian.

### 3.1 Measurement Equipment

#### 3.1.1 Viscosity Apparatus

The viscosity was measured by using NDJ - 1 Rotational Viscometer. It has four spindles (RV SPINDLE SET) and accuracy of 5%. The rotational speeds of the spindles are: 6, 12, 30, 60 RPM, the ranges of viscosities measured by the spindles are 0.1 to 100000 cP.



**Fig.(3.1):** NDJ - 1 Rotational Viscometer

The viscosity was measured using a appropriate spindle for each rotational speed: 6, 12, 30, 60 RPM at different temperatures. The temperature ranges from 8 °C to 73 °C.

#### 3.1.2 Temperature Apparatus

Digital Prima Long Thermometer was used to measure the temperature of olive oil samples. The accuracy of this apparatus is  $\pm 1\%$ . It measures temperature ranges from  $- 20$  °C to  $+ 100$  °C.

The temperature of the olive oil samples was incremented using the Fried Electric Model WB - 23.

### 3.1.3 Density Apparatus

The density of olive oil samples will be measured using a 2ml Pycnometer. The Pycnometer was first weighted empty and then weighted full of olive oil then the difference was divided by 2 ml to get the density.



**Fig.(3.2):** Pycnometer

The error in measuring the density is calculated using the following equation:

$$\Delta\rho = \pm \rho \left[ \frac{\Delta m}{m} + \frac{\Delta V}{V} \right] \quad (3.1)$$

The analytical balance HR - 200 with accuracy  $\pm 0.00005$  was used to measure the mass.

### 3.1.4 Refractive Index Apparatus

The index of refraction of the olive oil samples was measured using the way - 2s ABBE digital refractometer.



**Fig.(3.3):** The way - 2s ABBE digital refractometer

The measurement range of the device extends from 1.3000 - 1.7000 with accuracy equals to  $\pm 0.0002$ .

### **3.1.5 Acidity Measurement**

The acid value of olive oil was determined by the titrimetric method used in (AOAC 1997). The acid value of olive oil is equal to the mass of KOH in mg required to neutralize 1 g of olive oil dissolved in ethanol - ether mixture, and titrated with standard KOH solution.

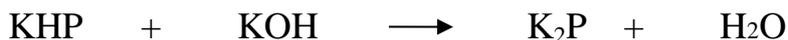
Three main steps were followed to measure the acid value of olive oil:

Firstly: a 0.1 M of ethanolic KOH solution is prepared and standardized as follows:

- A 0.56 g of solid KOH is transferred into a 100 - mL volumetric flask and dissolved in absolute ethanol (to get roughly 0.01 moles with 0.1 M of KOH ethanol solution).
- A 0.204 g of dry primary standard KHP (Molar mass = 204.23 g/mol) was weighted into a 250 mL conical flask and was dissolved in 50 mL of distilled water (to get accurately 0.01 moles with 0.2 M of KHP solution).
- 3 drops of phenolphthalein are added (in order not to increase volume and correspondingly changing values of molarity) and

titrated dropwise in the vicinity of the end point with KOH until a pink color is obtained and persisted for 30 seconds.

- The last two steps were repeated three times.
- The average molar concentration of KOH solution was calculated.



Where the exact molarity of KOH is  $\frac{\text{weight of KHP (g)} \times 100\%}{204.23 \times \text{mL of KOH}}$

Secondly: Ethanol - ether mixture was prepared:

- 50 mL of absolute ethanol and 50 mL of ether were mixed in a conical flask, and 3 drops of phenolphthalein solution then ethanolic KOH were added to faint pink color.

Thirdly: The acid value of olive oil was determined:

- 5 - 10 mg oil was weighted into 250 mL conical flask, then 50 mL of ethanol ether mixture and 3 drops phenolphthalein solution are added.
- The resulted solution is titrated the standard ethanolic KOH solution until permanent faint pink appears and persists for 30 seconds.

Then the acid value was calculated according to the following equation:

$$\text{Acid value} = \frac{\text{mL KOH standard solution} \times \text{molarity of KOH standard solution} \times 56.1}{\text{wt of sample (g)}}$$

The acid value may be expressed in terms of % free fatty acids as follows:

$$\text{FFA}\% = \frac{\text{Acid value}}{1.99}$$

## Chapter Four

### Results and Data Analysis

#### 4.1 Density Results

The densities in ( $\text{gm}/\text{cm}^3$ ) of the collected olive oil samples were measured. The overall density results are given in Table 4.1. The density results are tabulated according to the region of the olive oil sample and the sample storage age in years.

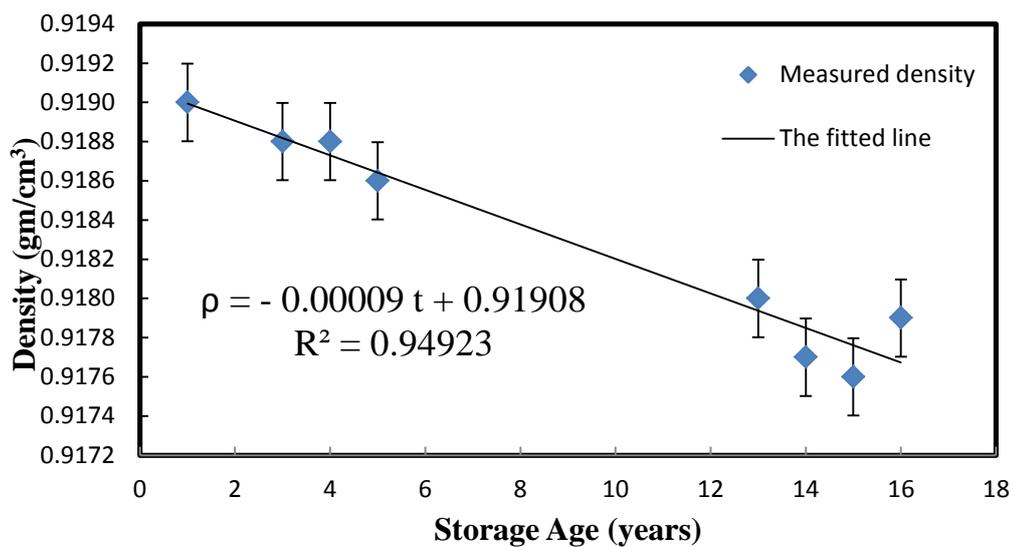
**Table (4.1): Measured density ( $\text{gm}/\text{cm}^3$ ) of olive oil samples in different regions and for different storage ages**

Storage Age (years)	L <sub>1</sub>	L <sub>2</sub>	L <sub>3</sub>	L <sub>4</sub>	L <sub>5</sub>	L <sub>6</sub>	L <sub>7</sub>	L <sub>8</sub>	L <sub>9</sub>	L <sub>10</sub>	L <sub>11</sub>	L <sub>12</sub>	L <sub>13</sub>	Average
1	0.9190	0.9189	0.9183	0.9184	0.9189			0.9191						0.9187
2		0.9189	0.9178	0.9184				0.9191	0.9185			0.9185		0.9185
3	0.9188	0.9185		0.9183	0.9188	0.9183	0.9188	0.9189	0.9182		0.9185	0.9185	0.9188	0.9185
4	0.9188			0.9182	0.9186	0.9182	0.9186						0.9185	0.9184
5	0.9186						0.9185		0.9177					0.9182
6					0.9186		0.9181	0.9177				0.9180		0.9181
7					0.9182				0.9175					0.9178
8								0.9177			0.9180			0.9178
12						0.9177							0.9179	0.9178
13	0.9180					0.9176								0.9178
14	0.9177	0.9160	0.9176			0.9176				0.9179			0.9179	0.9174
15	0.9176	0.9155	0.9175			0.9175					0.9174		0.9179	0.9172
16	0.9179					0.9175				0.9165			0.9175	0.9173
19							0.9174							0.9174
Average	0.9183	0.9176	0.9178	0.9183	0.9186	0.9178	0.9183	0.9185	0.9180	0.9172	0.9180	0.9183	0.9181	0.9180

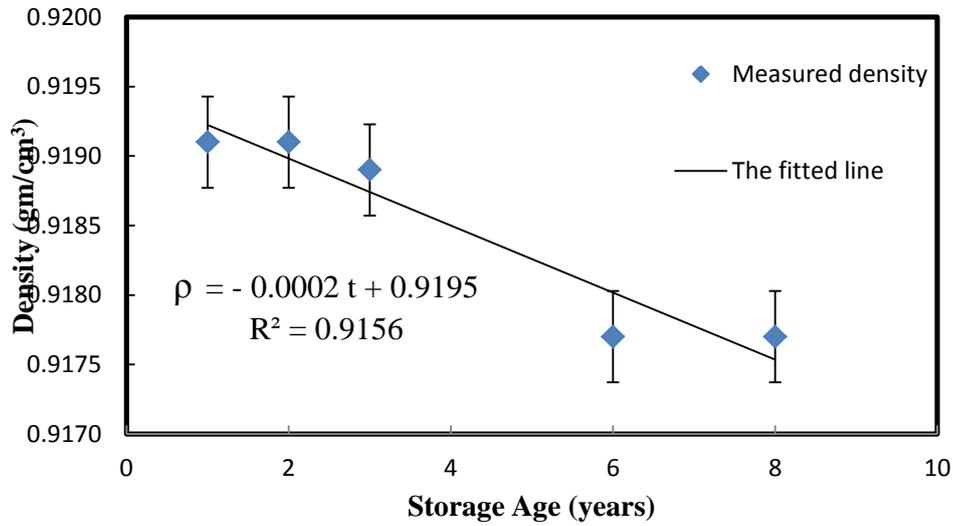
The overall average density of olive oil samples is  $0.9180 \text{ gm/cm}^3$ .

The highest values of density recorded were the 1 year storage age and 2 years storage age of  $L_8$  samples ( $0.9190 \text{ gm/cm}^3$ ). While the lowest value was found for 15 years storage age of  $L_2$  samples ( $0.9155 \text{ gm/cm}^3$ ). The maximum value of the average density was found for  $L_5$  samples ( $0.9186 \text{ gm/cm}^3$ ) while the minimum average value of density was found for  $L_{10}$  samples ( $0.9172 \text{ gm/cm}^3$ ).

The relationship between density of olive oil samples of  $L_1$  and  $L_2$  and storage age is given in Figs. (4.1, 4.2), respectively.



**Fig.(4.1):** Measured density versus storage age of olive oil samples of  $L_1$



**Fig.(4.2):** Measured density versus storage age of olive oil samples of L<sub>8</sub>

The density shows a linear proportional relationship with sample storage age. The relationship between density and storage age for the rest of regions are shown in Appendix A.

#### **4.2 Refractive Index Results**

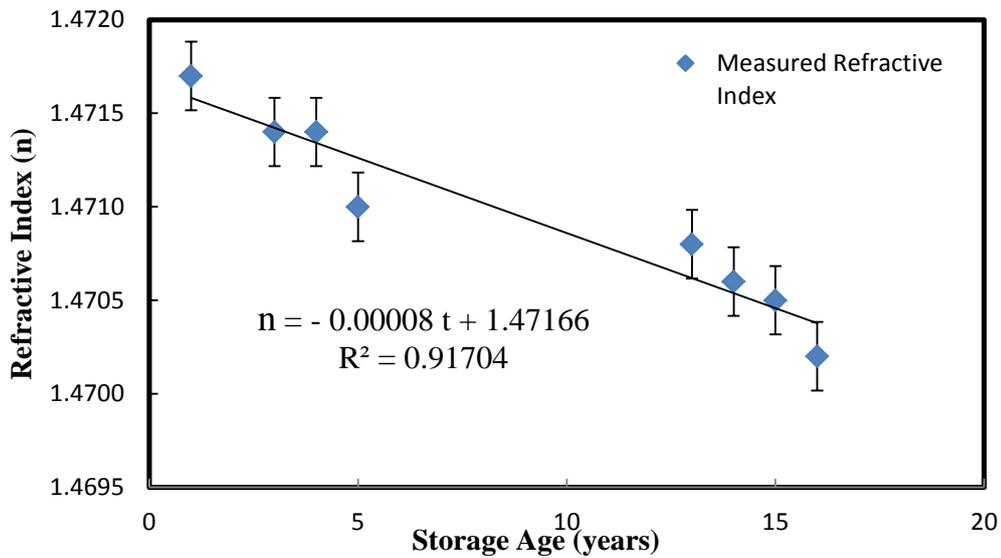
The measured refractive indexes of olive oil for all samples, from all regions are given in Table 4.2.

**Table (4.2): Measured refractive index of olive oil samples of different regions and different storage ages**

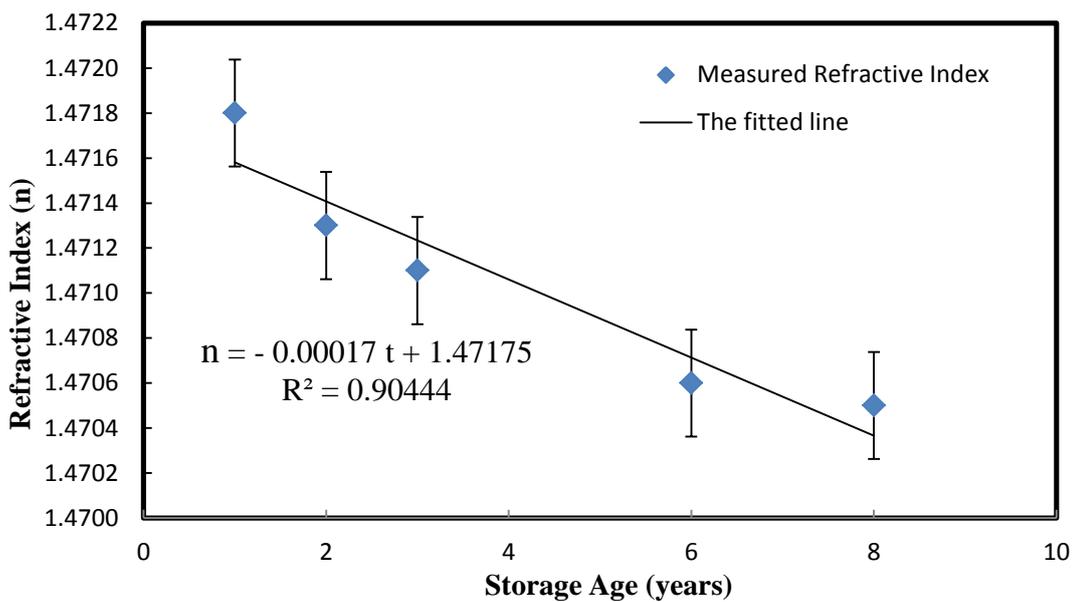
Storage Age (years)	L <sub>1</sub>	L <sub>2</sub>	L <sub>3</sub>	L <sub>4</sub>	L <sub>5</sub>	L <sub>6</sub>	L <sub>7</sub>	L <sub>8</sub>	L <sub>9</sub>	L <sub>10</sub>	L <sub>11</sub>	L <sub>12</sub>	L <sub>13</sub>	Average
1	1.4717	1.4712	1.4710	1.4717	1.4716			1.4718						1.4715
2			1.4710	1.4716				1.4713	1.4713			1.4713		1.4713
3	1.4714	1.4711		1.4711	1.4714	1.4711	1.4715	1.4711	1.4713		1.4713	1.4712	1.4711	1.4712
4	1.4714			1.4707	1.4710		1.4714						1.4705	1.4710
5	1.4710						1.4712		1.4712					1.4711
6					1.4708		1.4712	1.4706				1.4709		1.4709
7					1.4707				1.4709					1.4708
8					1.4707			1.4705			1.4711			1.4708
12						1.4705							1.4703	1.4704
13	1.4708					1.4702								1.4705
14	1.4706	1.4708	1.4709			1.4701				1.4708			1.4701	1.4706
15	1.4705	1.4706	1.4708			1.4704				1.4698	1.4710		1.4702	1.4705
16	1.4702					1.4700				1.4690			1.4700	1.4698
19							1.4711							1.4711
Average	1.4710	1.4709	1.4709	1.4713	1.4710	1.4704	1.4713	1.4711	1.4712	1.4699	1.4711	1.4711	1.4704	1.4708

The average value of refractive index of all olive oil samples is 1.4708. The range of refractive index of all samples extends from 1.4690 (16 years storage age L<sub>10</sub> sample) to 1.4718 (1 year storage age L<sub>8</sub> sample).

The relationship between refractive index and storage age for samples collected from L<sub>1</sub> and L<sub>8</sub> are shown in Figs 4.3 and 4.4.



**Fig.(4.3):** Measured refractive index versus storage age of olive oil samples of L<sub>1</sub>



**Fig.(4.4):** Measured refractive index versus storage age of olive oil samples of L<sub>8</sub>

One can notice from Figs 4.3 and 4.4 that the refractive index decreases as the storage age of the olive oil sample increases. The relationship between refractive index and storage age for the rest of regions are showed in Appendix B.

### **4.3 Acidity Results**

The results of olive oil samples acidity for samples collected from all regions and different storage ages are given in Table 4.3.

**Table 4.3: Measured acidity in FFA% of olive oil samples in different regions for different storage ages**

Storage Age (years)	L <sub>1</sub>	L <sub>2</sub>	L <sub>3</sub>	L <sub>4</sub>	L <sub>5</sub>	L <sub>6</sub>	L <sub>7</sub>	L <sub>8</sub>	L <sub>9</sub>	L <sub>10</sub>	L <sub>11</sub>	L <sub>12</sub>	L <sub>13</sub>
1	1.18	1.72	0.94	0.51	0.45			0.44					
2			0.70	0.53				0.50	0.67			0.77	
3	1.55	2.53		0.56	1.49	1.03	0.80	1.96	0.96		0.96	1.88	0.56
4	2.64			1.96	2.07	1.80	1.12						1.03
5	3.97						1.56		1.18				
6					2.40		2.50	4.77				2.00	
7					2.88				1.20				
8					2.94			5.25			3.92		
12						3.18							2.31
13	8.79									4.40			
14	9.09	5.09	5.98							5.04			
15	11.13	5.77	5.72			3.81					7.07		3.95
16	13.01					4.49				9.49			4.60
19						5.02	4.00						5.22

The range of acidity extends from 0.44% for the 1 year storage age (L<sub>8</sub>) to 13.01% for the 16 years storage (L<sub>1</sub>).

Table 4.4 shows the acidity of olive oil samples from different regions as a function of storage age. The Classification of olive oil is given according to Table 2.3.

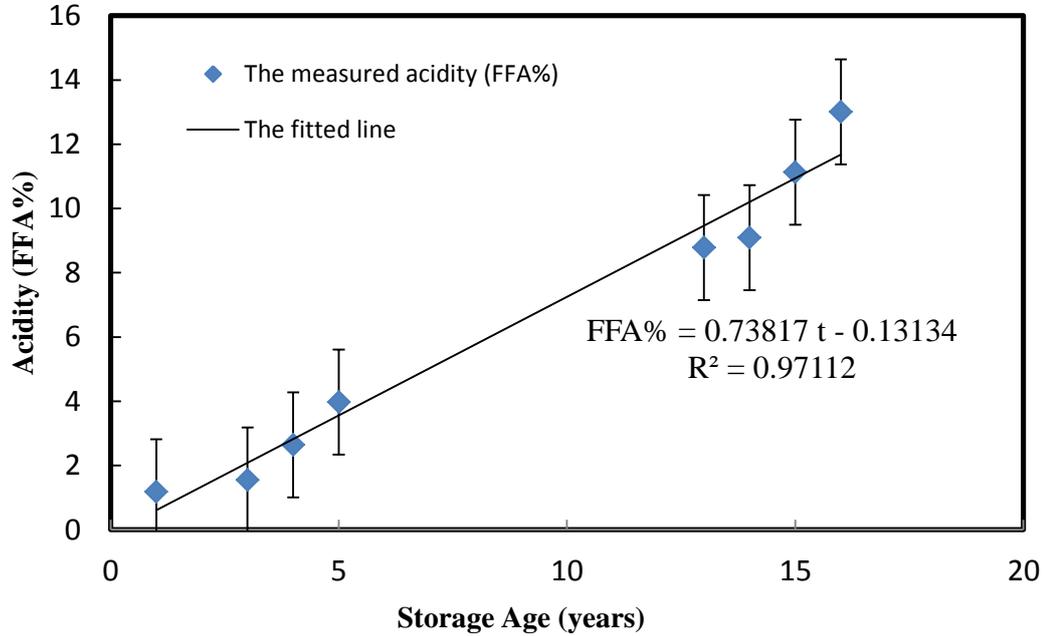
**Table 4.4: The acidity of olive oil samples from different regions of different storage ages**

Region	Storage Age(years)	Acidity	Olive Oil Classification
L <sub>3</sub>	2	0.70	Extra virgin olive oil
L <sub>4</sub>	1	0.51	Extra virgin olive oil
L <sub>4</sub>	2	0.53	Extra virgin olive oil
L <sub>4</sub>	3	0.56	Extra virgin olive oil
L <sub>5</sub>	1	0.45	Extra virgin olive oil
L <sub>8</sub>	1	0.44	Extra virgin olive oil
L <sub>8</sub>	2	0.50	Extra virgin olive oil
L <sub>9</sub>	2	0.67	Extra virgin olive oil
L <sub>12</sub>	2	0.77	Extra virgin olive oil
L <sub>13</sub>	3	0.56	Extra virgin olive oil
L <sub>1</sub>	1	1.18	Virgin olive oil
L <sub>1</sub>	3	1.55	Virgin olive oil
L <sub>2</sub>	1	1.72	Virgin olive oil
L <sub>3</sub>	1	0.94	Virgin olive oil
L <sub>4</sub>	4	1.96	Virgin olive oil
L <sub>5</sub>	3	1.49	Virgin olive oil
L <sub>6</sub>	3	1.03	Virgin olive oil
L <sub>6</sub>	4	1.80	Virgin olive oil
L <sub>7</sub>	3	0.80	Virgin olive oil
L <sub>7</sub>	4	1.12	Virgin olive oil
L <sub>7</sub>	5	1.56	Virgin olive oil
L <sub>8</sub>	3	1.96	Virgin olive oil
L <sub>9</sub>	3	0.96	Virgin olive oil
L <sub>9</sub>	5	1.18	Virgin olive oil
L <sub>9</sub>	7	1.20	Virgin olive oil
L <sub>11</sub>	3	0.96	Virgin olive oil
L <sub>12</sub>	3	1.88	Virgin olive oil
L <sub>12</sub>	6	2.00	Virgin olive oil
L <sub>13</sub>	4	1.03	Virgin olive oil

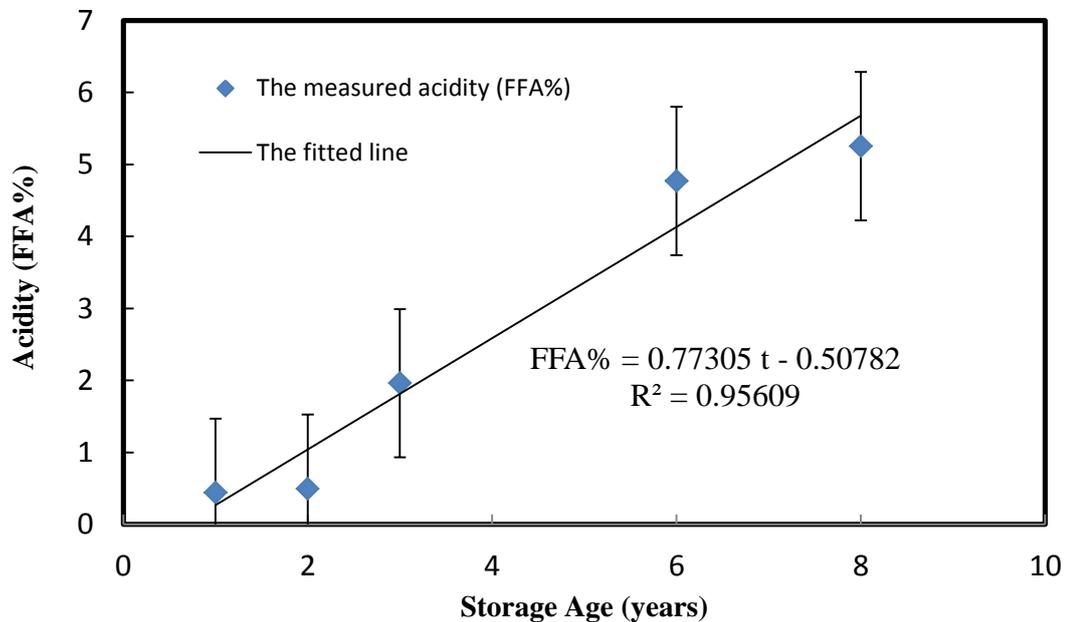
L <sub>1</sub>	4	2.64	Ordinary virgin olive oil
L <sub>2</sub>	3	2.53	Ordinary virgin olive oil
L <sub>5</sub>	4	2.07	Ordinary virgin olive oil
L <sub>5</sub>	6	2.40	Ordinary virgin olive oil
L <sub>5</sub>	7	2.88	Ordinary virgin olive oil
L <sub>5</sub>	8	2.94	Ordinary virgin olive oil
L <sub>6</sub>	12	3.18	Ordinary virgin olive oil
L <sub>7</sub>	6	2.50	Ordinary virgin olive oil
L <sub>13</sub>	12	2.31	Ordinary virgin olive oil
L <sub>1</sub>	5	3.97	Lampante oil
L <sub>1</sub>	13	8.79	Lampante oil
L <sub>1</sub>	14	9.09	Lampante oil
L <sub>1</sub>	15	11.13	Lampante oil
L <sub>1</sub>	16	13.01	Lampante oil
L <sub>2</sub>	14	5.09	Lampante oil
L <sub>2</sub>	15	5.77	Lampante oil
L <sub>3</sub>	14	5.98	Lampante oil
L <sub>3</sub>	15	5.72	Lampante oil
L <sub>6</sub>	14	3.81	Lampante oil
L <sub>6</sub>	15	4.49	Lampante oil
L <sub>6</sub>	16	5.02	Lampante oil
L <sub>7</sub>	19	4.00	Lampante oil
L <sub>8</sub>	6	4.77	Lampante oil
L <sub>8</sub>	8	5.25	Lampante oil
L <sub>10</sub>	13	4.40	Lampante oil
L <sub>10</sub>	14	5.04	Lampante oil
L <sub>10</sub>	16	9.49	Lampante oil
L <sub>11</sub>	8	3.92	Lampante oil
L <sub>11</sub>	15	7.07	Lampante oil
L <sub>13</sub>	14	3.95	Lampante oil
L <sub>13</sub>	15	4.60	Lampante oil
L <sub>13</sub>	16	5.22	Lampante oil

Three samples of crop 2013 (L<sub>4</sub>, L<sub>5</sub>, and L<sub>8</sub>) are extra virgin, while the other three samples of the same crop are virgin (L<sub>1</sub>, L<sub>2</sub>, and L<sub>3</sub>). The five samples of crop 2012 are all extra virgin. In general, one can notice from Table (4.4) that the olive oil samples are good for human consumption up to 10 years storage age.

The relationship between acidity and storage age for the collected samples from  $L_1$  and  $L_8$  are shown in Figs 4.5 and 4.6, respectively.



**Fig.(4.5):** Measured acidity versus storage age of olive oil samples of  $L_1$



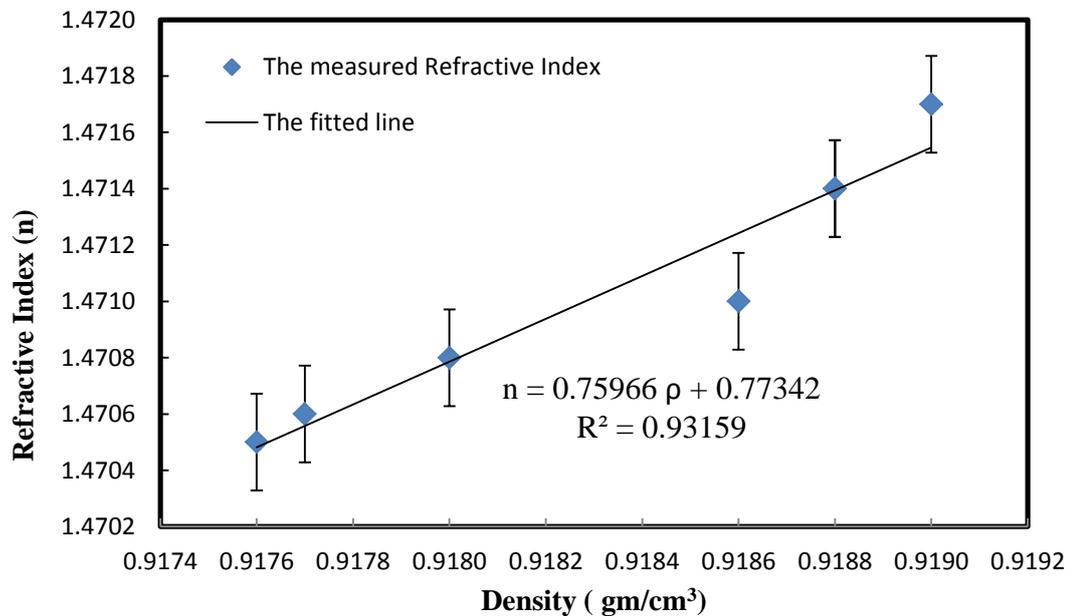
**Fig.(4.6):** Measured acidity versus storage age of olive oil samples of  $L_8$

The olive oil acidity increases as the storage age of the sample increases. The relationship between olive oil acidity and storage age is shown to be

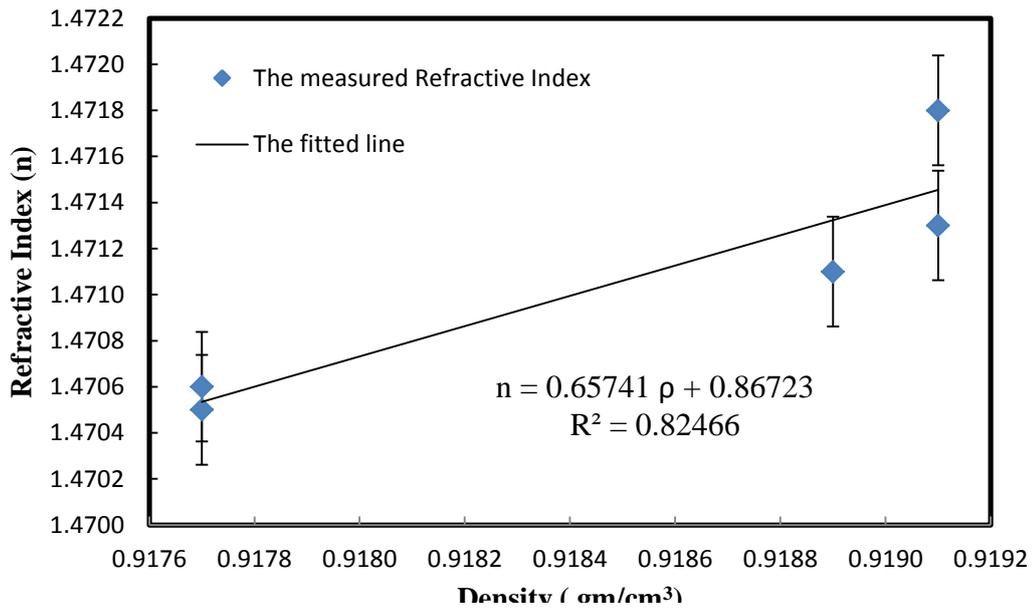
linear. The acidity versus storage age for the rest of the samples of different regions are shown in the Appendix C.

#### 4.4 Refractive Index as a Function of Density of Olive Oil

The refractive index of olive oil samples were plotted against density mass and the results of the  $L_1$  and  $L_8$  are shown in Figs.4.7 and 4.8. The refractive index versus density for the rest of the samples are shown in the Appendix D.



**Fig.(4.7):** Measured refractive index versus density mass of olive oil samples of  $L_1$



**Fig.(4.8):** Measured refractive index versus density mass of olive oil samples of L<sub>8</sub>

The relationship between refractive index and density mass is linear.

The reason for the increasing of acidity with storage age is the definition of acidity. A concentration of free fatty acids, oils and fats are made up of triglycerides and chains of free fatty acids, when chains of free fatty acids are liberated, they can increase the acidity free fatty acids which leads to an increase in acidity. The reason for the breakup of the of fatty chains is an lipase enzyme that gives the association between fatty chains and triglycerides then transformed into free fatty chains. The presence of water with the presence of holes in the olive grain or high oil temperature will lead to increase in the activity of the lipase enzyme and thus higher acidity. As the temperature of the oil increases the spaces between particles increases then their motion became easier which leads to a decrease in viscosity because the viscosity is the resistance of flow of the liquid. The decrease of density storage with storage age is due to the increase of free

fatty acids (i.e., the breakup of the liquid molecules) and thus less density between molecules, leading to a decrease in mass density.

#### 4.5 Viscosity Results

The Kinematic viscosity of olive oil samples was calculated at room temperature (25 °C ). Table 4.5 gives the results of L<sub>1</sub> and L<sub>8</sub> samples at different storage age.

**Table 4.5: Results of kinematic viscosity of L<sub>1</sub> and L<sub>8</sub> samples at different storage age**

	(v) in (cSt)					
t (°C)	L <sub>1</sub>			L <sub>8</sub>		
	Storage age: 2 years	Storage age: 5 years	Storage age: 13 years	Storage age: 3 years	Storage age: 6 years	Storage age: 10 years
25	74.00	74.02	69.71	81.61	80.63	77.36

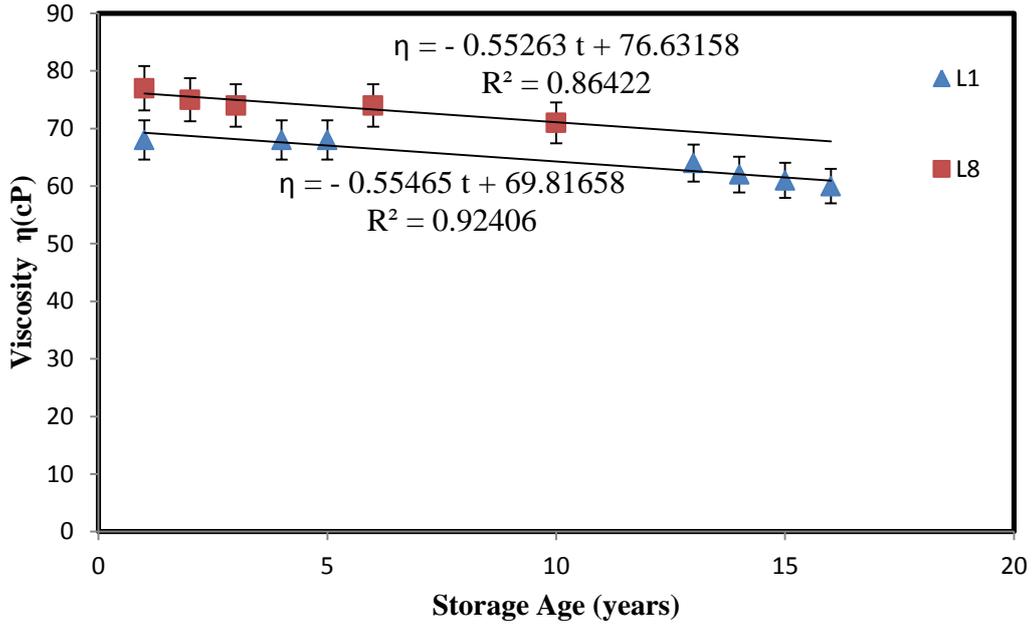
The dynamic viscosity of olive oil samples was measured at different temperatures. Table 4.6 shows the results of L<sub>1</sub> and L<sub>8</sub> samples at different storage age.

**Table 4.6: Results of dynamic viscosity of L<sub>1</sub> and L<sub>8</sub> samples at different storage age**

t (°C)	$(\eta_{\text{exp}})$ in cP					
	L <sub>1</sub>			L <sub>8</sub>		
	Storage age: 2 years	Storage age: 5 years	Storage age: 13 years	Storage age: 3 years	Storage age: 6 years	Storage age: 10 years
23	75	72	70	80	79	78
25	68	68	64	75	74	71
28	62	67	63	72	71	69
30	58	61	59	68	66	67
33	58	55	53	63	63	62
37	56	49	49	59	61	59
40	52	47	45	55	57	57
42	52	43	44	52	57	54
44	45	39	42	45	50	52
47	38	33	42	40	47	50
50	32	31	36	33	46	46
52	31	25	32	28	43	42
55	29	25	28	26	41	36
58	22	21	24	23	38	32
60	17	18	22	20	37	30
63	17	17	22	19	33	25

The dynamic viscosity decreases with storage age sample as a function of temperature. It also decreases with temperature at a given sample age.

The relationship between dynamic viscosity and storage age of the samples from L<sub>1</sub> and L<sub>8</sub> are shown in Fig.4.9.



**Fig.(4.9):** Measured viscosity versus storage age of olive oil samples of L<sub>1</sub> and L<sub>8</sub> at 25 °C

Figure 4.9 shows that the viscosity decreases as the storage age of the olive oil sample increases.

#### 4.5.1 Viscosity Results and Theoretical Predictions

The experimental results of dynamic viscosity versus temperature were compared with equations obtained by Abramovic and Andrade. The percentage of absolute deviations and standard deviation between the measured and theoretical data were calculated.

Three equations were used to fit the experimental data of this work, these are: Abramovic's equation (formula 1)

$$\log \eta = \frac{A}{T} - B \quad (4.1)$$

and Abramovic's equation (formula 2)

$$\eta = A - B \log t \quad (4.2)$$

and Andrade's equation

$$\ln \eta = A + \frac{B}{T} + CT \quad (4.3)$$

**a. Two Constants Abramovic's Equation (Formula 1)**

The dynamic viscosity versus temperature using the two constants Abramovic's equation (formula 1) has been calculated and gives:

$\log \eta = \frac{A}{T} - B$  with  $A = 1558.2$  (K) and  $B = 3.433$  (Abramovic *et al.*, 1998). A comparison between the measured values of dynamic viscosity and the calculated values using Abramovic's equation (formula 1) of four different olive oil samples. The values of AAD% and SD are also given in Table 4.7.

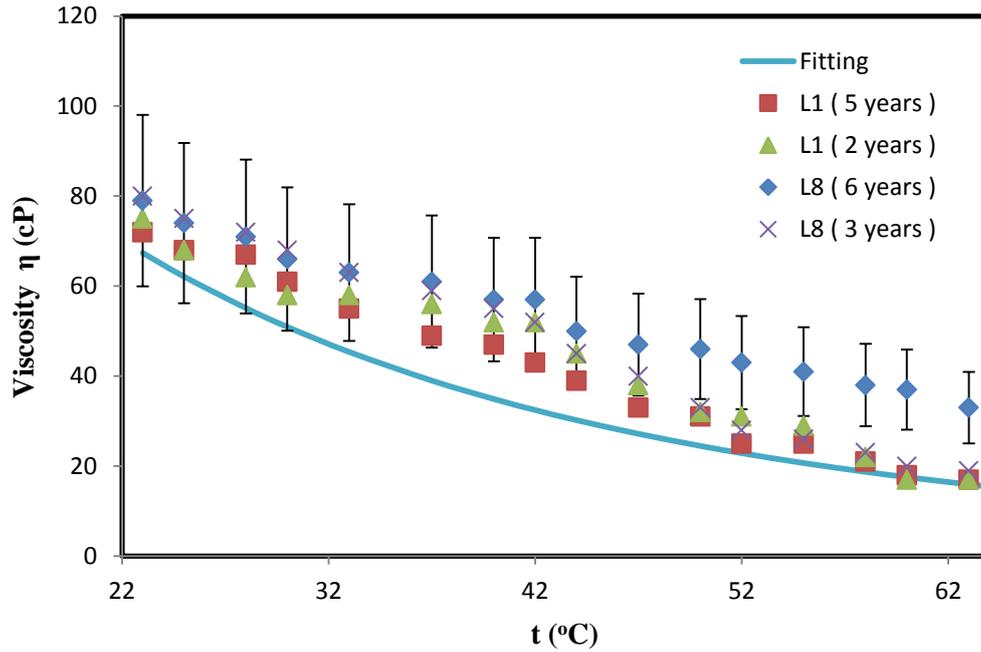
**Table 4.7: The measured and calculated dynamic viscosity of olive oil versus temperature using Abramovic's equation (formula 1)**

$$[\log \eta = \frac{A}{T} - B] \text{ for } L_1 \text{ and } L_8$$

t (°C)	$\eta_{cal}$ (cP) using Abramovic's equation (formula 1)	$\eta_{exp}$ (cP)					
		L <sub>1</sub>			L <sub>8</sub>		
		Storage age: 2 years	Storage age: 5 years	Storage age: 13 years	Storage age: 3 years	Storage age: 6 years	Storage age: 10 years
23	67	75	72	70	80	79	78
25	62	68	68	64	75	74	71
28	55	62	67	63	72	71	69
30	51	58	61	59	68	66	67
33	45	58	55	53	63	63	62
37	39	56	49	49	59	61	59
40	35	52	47	45	55	57	57
42	32	52	43	44	52	57	54
44	30	45	39	42	45	50	52
47	27	38	33	42	40	47	50
50	24	32	31	36	33	46	46
52	23	31	25	32	28	43	42
55	21	29	25	28	26	41	36
58	19	22	21	24	23	38	32
60	18	17	18	22	20	37	30
63	16	17	17	22	19	33	25
AAD%	----	13.2%	12.0%	15.3%	15.8%	24.1%	22.1%
SD	----	1.72	1.24	1.42	2.17	3.03	2.76

The values of AAD% are high for all samples. Their ranges are from 12.0% to 24.1%, which means that the constants of Abramovic's equation (formula1) used by Abramovic do not represent a good fit for our results.

Fig 4.10 shows the relationship between the dynamic viscosity of olive oil versus temperature for L<sub>1</sub> and L<sub>8</sub> samples.



**Fig.(4.10):** Measured and calculated values of dynamic viscosity using Abramovic's equation (formula 1) for four different samples

### b. Two Constants Abramovic's Equation (Formula 2)

The viscosities of olive oil for 2, 5 and 13 years storage age of  $L_1$  sample and 3, 6, and 10 years storage age of  $L_8$  sample were calculated using Abramovic's two constants formula 2 which is:  $\eta = A - B \log t$  with  $A = 235.4$  cP and  $B = 124.1$ cP (Abramovic *et al.*, 1998). The results of calculated and measured values of viscosity are given in Table 4.8.

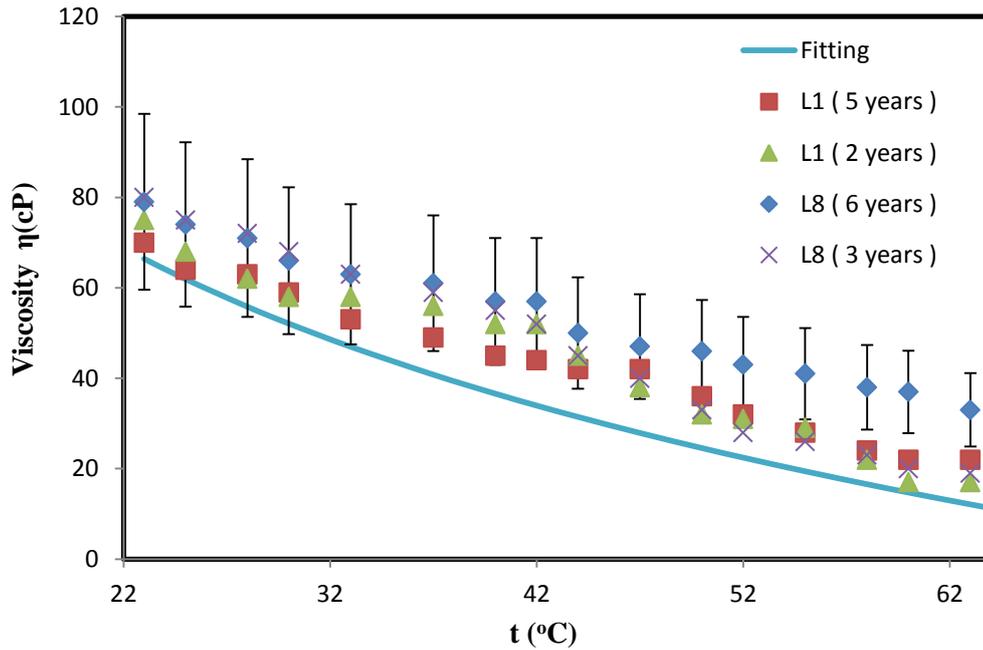
**Table 4.8: The measured and calculated dynamic viscosity of olive oil versus temperature using Abramovic's equation (formula 2)**

$$[\eta = A - B \log t] \text{ for } L_1 \text{ and } L_8$$

t (°C)	using Abramovic's equation (formula 2) $\eta_{cal}$ (cP)	$\eta_{exp}$ (cP)					
		L <sub>1</sub>			L <sub>8</sub>		
		Storage age: 2 years	Storage age: 5 years	Storage age: 13 years	Storage age: 3 years	Storage age: 6 years	Storage age: 10 years
23	66	75	72	70	80	79	78
25	62	68	68	64	75	74	71
28	56	62	67	63	72	71	69
30	52	58	61	59	68	66	67
33	47	58	55	53	63	63	62
37	41	56	49	49	59	61	59
40	37	52	47	45	55	57	57
42	34	52	43	44	52	57	54
44	31	45	39	42	45	50	52
47	28	38	33	42	40	47	50
50	25	32	31	36	33	46	46
52	22	31	25	32	28	43	42
55	19	29	25	28	26	41	36
58	17	22	21	24	23	38	32
60	15	17	18	22	20	37	30
63	25	17	17	22	19	33	25
AAD%	----	14.3%	18.2%	21.3%	17.1%	24.6%	22.8%
SD	----	1.63	1.31	1.51	2.09	3.04	2.73

AAD% values are very high and extend from 14.3% for 2 years storage age of L<sub>1</sub> sample to 24.6% for 6 years storage age sample. This implies a failure fit constants for abramovic's equation (formula 2).

Fig. 4.11 shows the relationship between dynamic viscosity versus temperature for two samples from L<sub>1</sub> and L<sub>8</sub>.



**Fig.(4.11):** Measured and calculated values of dynamic viscosity using Abramovic's equation (formula 2) for four different samples from different regions

### c. Andrade's Equation

Andrade's equation  $\ln \eta = A + \frac{B}{T} + CT$  is used to calculate the dynamic viscosity of olive oil versus temperature. The values of A, B, and C are found to be equal -32.72, 7462.27 K, and 0.04 (  $\text{K}^{-1}$  ), respectively (Andrade, 1930; Abramovic *et al.*, 1998).

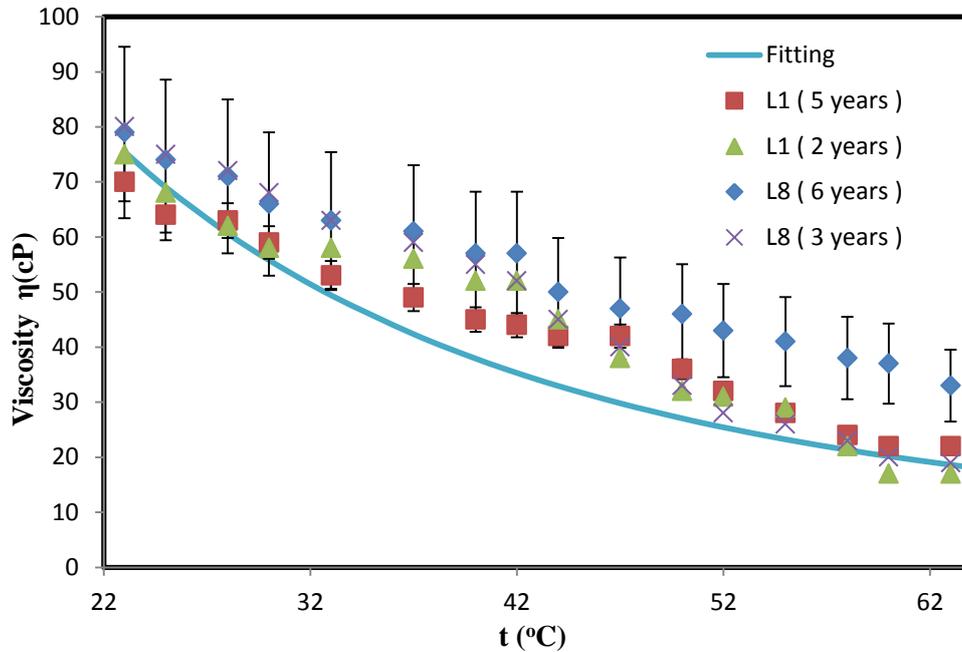
The calculated and experimental values of viscosity versus temperature are given in Table 4.9 for two samples from L<sub>1</sub> and L<sub>8</sub>.

**Table 4.9: The measured and calculated dynamic viscosity using Andrade's equation [ $\ln \eta = A + \frac{B}{T} + CT$ ] of olive oil samples collected from L<sub>1</sub> and L<sub>8</sub> versus temperature**

t (°C)	using $\eta_{cal}$ (cP) equation	$\eta_{exp}$ (cP)					
		L <sub>1</sub>			L <sub>8</sub>		
		Storage age: 2 years	Storage age: 5 years	Storage age: 13 years	Storage age: 3 years	Storage age: 6 years	Storage age: 10 years
23	75	75	72	75	80	79	78
25	69	68	68	69	75	74	71
28	61	62	67	61	72	71	69
30	56	58	61	56	68	66	67
33	49	58	55	49	63	63	62
37	42	56	49	42	59	61	59
40	38	52	47	38	55	57	57
42	35	52	43	35	52	57	54
44	33	45	39	33	45	50	52
47	30	38	33	30	40	47	50
50	27	32	31	27	33	46	46
52	25	31	25	25	28	43	42
55	23	29	25	23	26	41	36
58	21	22	21	21	23	38	32
60	20	17	18	20	20	37	30
63	19	17	17	19	19	33	25
AAD%	----	9.6%	6.4%	9.5%	10.4%	19.7%	17.4%
SD	----	1.30	0.77	1.02	1.63	2.51	2.25

High AAD% values were calculated, 9.6%, 6.4% and 9.5% for the 2 years, 5 years, and 13 years of L<sub>1</sub>, and 10.4%, 19.7%, and 17.4% for the 3 years, 6 years, and 10 years of L<sub>8</sub>. These indicate that Andrade's equation with the given constants is not suitable to describe the experimental results.

Fig. 4.12 shows the measured and calculated dynamic viscosity using Andrade's equation of olive oil samples collected from L<sub>1</sub> and L<sub>8</sub> versus temperature.



**Fig.(4.12):** Measured and calculated values of dynamic viscosity using Andrade's equation for four different samples

#### 4.5.2 Modifications of the Abramovic's and Andrade's equations

In section 4.5.1, Abramovic's first and second formulas and Andrade's equation with their constants failed to describe the relationship between dynamic viscosity and temperature of olive oil samples. Modifications were proposed to these equations by fitting the experimental data with different constants to get good prediction with the least error. In order to achieve that, experimental data was fitted using these equations and new constants are introduced and the values of AAD% and SD were calculated.

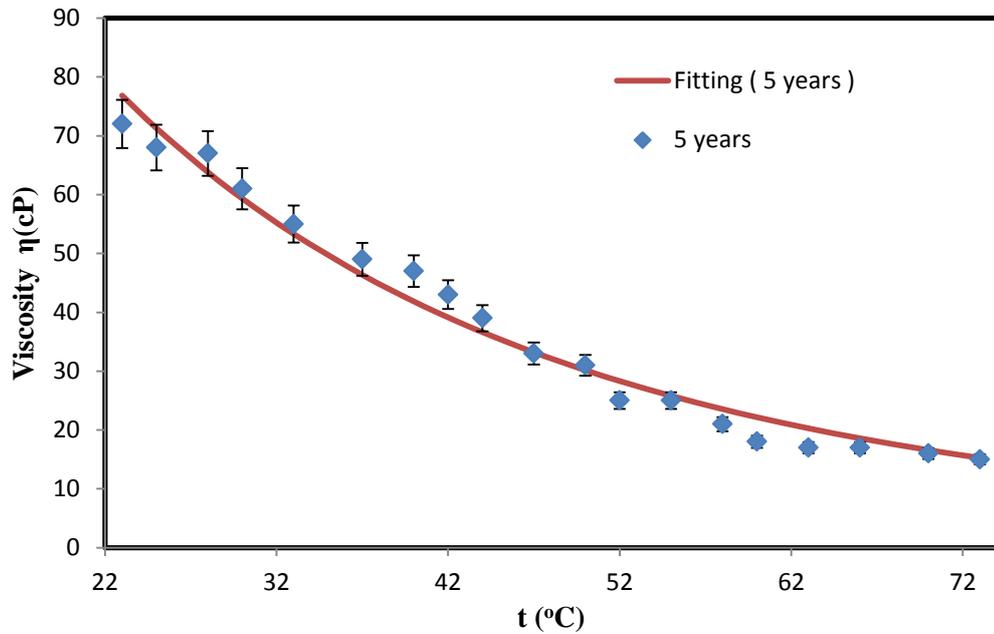
**a. Modification to Abramovic's Equation (Formula 1)**

Measured values of dynamic viscosity versus temperature of two samples from L<sub>1</sub> and L<sub>8</sub> were fitted using Abramovic's equation (formula 1)  $\log \eta = \frac{A}{T} - B$ . The values of A, B for each samples of different storage age were introduced, and values of AAD% and SD were calculated. The results of this fit are given in Table 4.10.

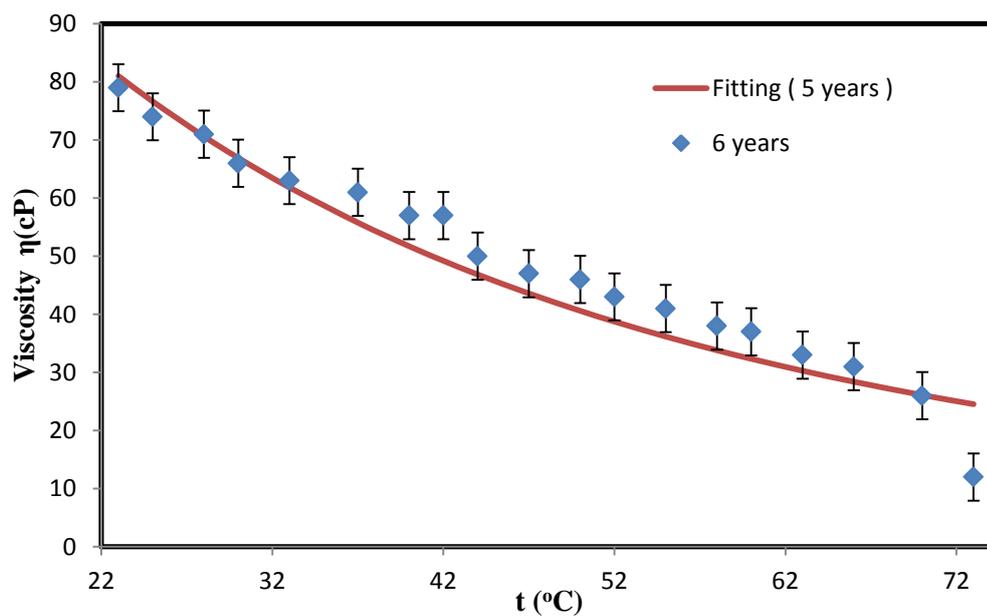
**Table 4.10: The measured and calculated dynamic viscosity using modified Abramovic's equation (formula 1)  $[\log \eta = \frac{A}{T} - B]$  of olive oil samples collected from L<sub>1</sub> and L<sub>8</sub> versus temperature**

t (°C)	L <sub>1</sub>						L <sub>8</sub>					
	Storage age: 2 years		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	$\eta_{\text{exp}}(\text{cP})$	$\eta_{\text{cal}}(\text{cP})$										
23	75	78	72	77	70	73	80	86	79	81	78	81
25	68	72	68	71	64	68	75	80	74	77	71	77
28	62	65	67	64	63	62	72	72	71	72	69	71
30	58	61	61	59	59	58	68	67	66	68	67	67
33	58	55	55	53	53	53	63	60	63	63	62	62
37	56	49	49	46	49	47	59	52	61	58	59	56
40	52	44	47	42	45	43	55	47	57	54	57	52
42	52	42	43	39	44	41	52	44	57	51	54	49
44	45	39	39	37	42	38	45	41	50	49	52	47
47	38	36	33	33	42	35	40	38	47	46	50	44
50	32	33	31	30	36	33	33	34	46	43	46	41
52	31	31	25	28	32	31	28	32	43	41	42	39
55	29	28	25	26	28	29	26	29	41	39	36	36
58	22	26	21	24	24	26	23	27	38	37	32	34
60	17	25	18	22	22	25	20	25	37	35	30	32
63	17	23	17	20	22	23	19	23	33	33	25	30
A(K)	1334.35		1439.45		1234.13		1420.86		970.83		1063.17	
B	2.615		2.975		2.306		2.864		1.369		1.681	
AAD%	7.7%		5.7%		7.3%		6.8%		2.4%		4.6%	
SD	0.9		0.7		0.5		1.2		1.8		1.6	

Figs 4.13 and 4.14 show the relationship between viscosity and temperature for two samples of  $L_1$  and  $L_8$  respectively. In addition, the fitted line using the modified Abramovic's first formula is given.



**Fig.(4.13):** Measured and calculated values of dynamic viscosity of olive oil of  $L_1$  using modified Abramovic's equation (formula 1)



**Fig.(4.14):** Measured and calculated values of dynamic viscosity of olive oil of  $L_8$  using modified Abramovic's equation (formula 1)

The values of AAD% given in Table 4.9 are still high. They are 7.7%, 5.7%, and 7.3%, for the 2 years, 5 years, and 13 years of L<sub>1</sub>, and 6.8%, 2.4%, and 4.6% for the 3 years, 6 years, and 10 years of L<sub>8</sub>. Therefore one can conclude that modified Abramovic's equation (formula 1) was not suitable for describing the relationship between dynamic viscosity and temperature.

**b. Modification to Abramovic's Equation (Formula 2)**

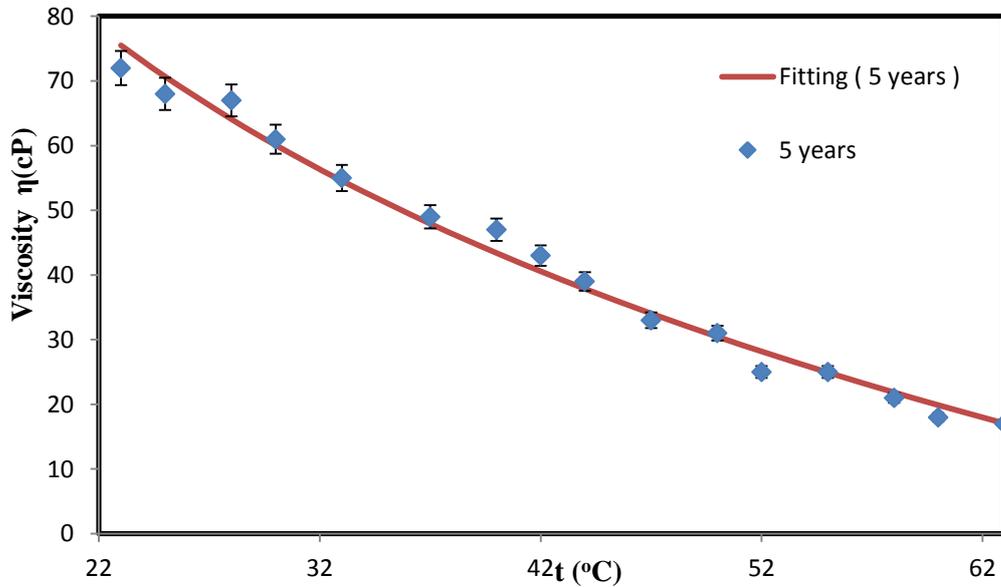
Modification was made on Abramovic's equation (formula 2)

$[\eta = A - B \log t]$  of olive oil behavior of samples from L<sub>1</sub> and L<sub>8</sub>, the results of measured and calculated values of dynamic viscosity are given in Table 4.11.

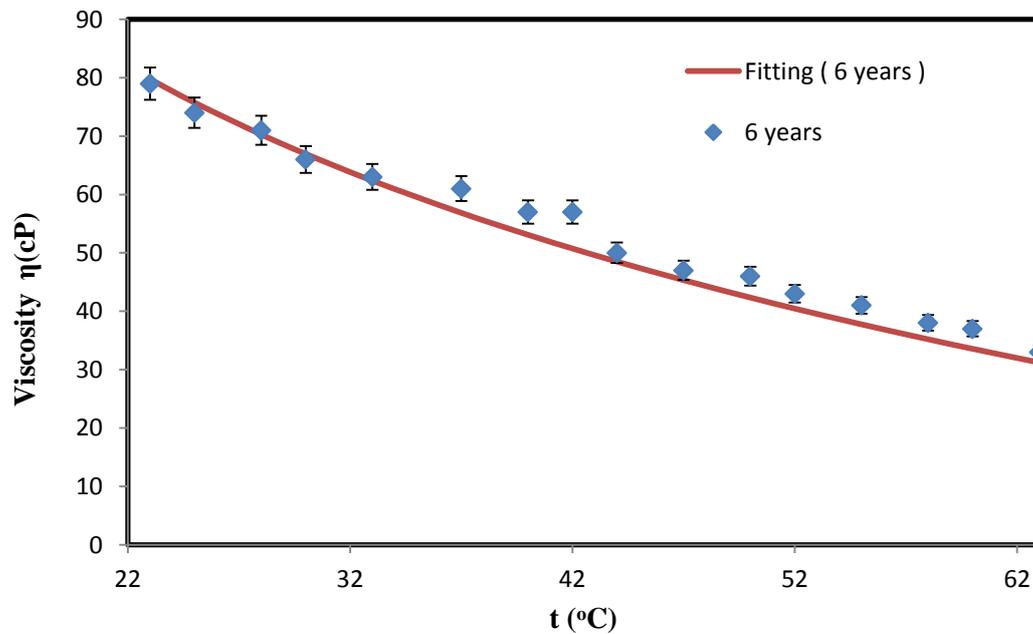
**Table 4.11: The measured and calculated dynamic viscosity using modified Abramovic's equation (formula 2) [ $\eta = A - B \log t$ ] of olive oil samples from L<sub>1</sub> and L<sub>8</sub> versus temperature**

t (°C)	L <sub>1</sub>						L <sub>8</sub>					
	Storage age: 2 years		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$
23	75	77	72	76	70	71	80	85	79	79	78	80
25	68	72	68	71	64	67	75	79	74	76	71	76
28	62	66	67	64	63	62	72	72	71	71	69	70
30	58	62	61	60	59	58	68	68	66	68	67	67
33	58	57	55	55	53	54	63	61	63	64	62	62
37	56	50	49	48	49	48	59	54	61	58	59	57
40	52	46	47	43	45	45	55	49	57	55	57	53
42	52	43	43	41	44	42	52	46	57	53	54	51
44	45	41	39	38	42	40	45	43	50	51	52	48
47	38	37	33	34	42	37	40	39	47	48	50	45
50	32	33	31	30	36	34	33	35	46	45	46	42
52	31	31	25	28	32	32	28	32	43	44	42	40
55	29	28	25	25	28	29	26	28	41	41	36	38
58	22	25	21	22	24	27	23	25	38	39	32	35
60	17	23	18	20	22	25	20	23	37	37	30	34
63	17	21	17	17	22	23	19	20	33	35	25	31
A(cP)	251.06		257.41		221.51		286.70		217.24		230.80	
B(cP)	128.058		133.583		110.397		148.372		101.243		110.926	
AAD%	6.3%		3.7%		3.0%		4.4%		3.5%		4.3%	
SD	0.8		0.6		0.3		1.2		1.7		1.4	

The measured and fitted viscosity - temperature relationship using the modified Abramovic's equation (formula 2) are shown in Figs. 4.15 and 4.16.



**Fig.(4.15):** Measured and calculated values of dynamic viscosity of olive oil of  $L_1$  using modified Abramovic's equation (formula 2)



**Fig.(4.16):** Measured and calculated values of dynamic viscosity of olive oil of  $L_8$  using modified Abramovic's equation (formula 2)

The calculated values of AAD% extend from 3.0% to 6.3%. Which are relatively high values. This implies that modified Abramovic's equation (formula 2) is not proper to fit the relationship between viscosity and temperature.

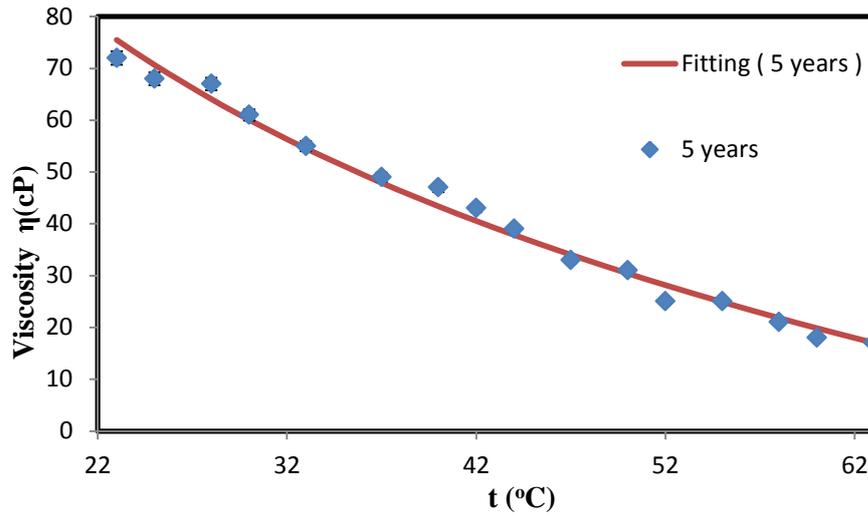
### **c. Modification to Andrade's Equation**

Measured and calculated values of dynamic viscosity versus temperature of two samples from L<sub>1</sub> and L<sub>8</sub> were fitted using Andrade's equation [ $\ln \eta = A + \frac{B}{T} + CT$ ]. The values of A, B, and C for each sample were introduced, and values of AAD% and SD were calculated. The results of this fit are given in Table 4.12.

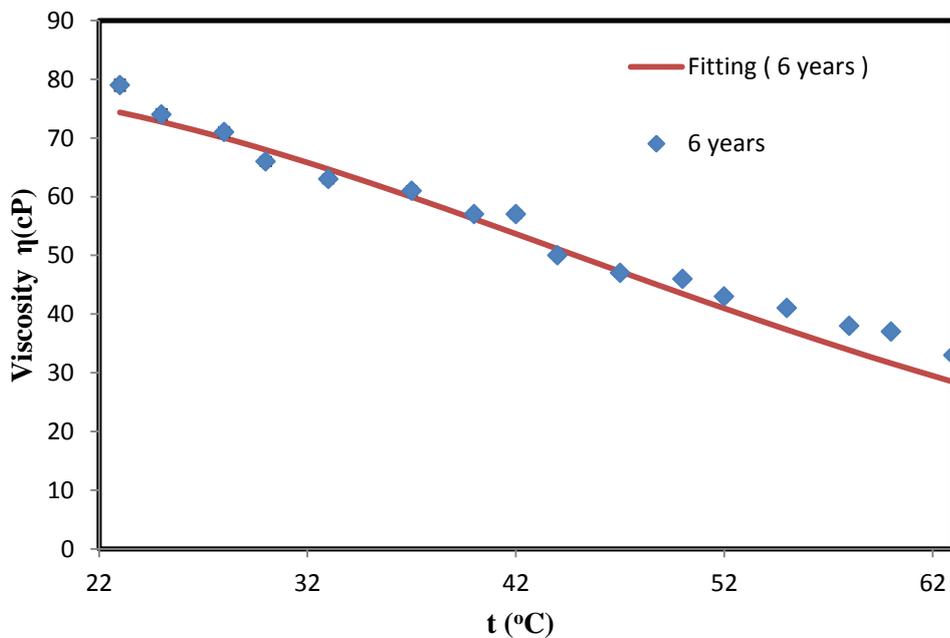
**Table 4.12: The measured and calculated dynamic viscosity using modified Andrade's equation [ $\ln \eta = A + \frac{B}{T} + CT$ ] of olive oil samples from L<sub>1</sub> and L<sub>8</sub> versus temperature**

t (°C)	L <sub>1</sub>						L <sub>8</sub>					
	Storage age: 2 years		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	$\eta_{\text{exp}}(\text{cP})$	$\eta_{\text{cal}}(\text{cP})$										
23	75	70	72	72	70	68	80	78	79	78	78	74
25	68	68	68	69	64	66	75	76	74	75	71	73
28	62	66	67	65	63	62	72	73	71	71	69	70
30	58	64	61	62	59	59	68	70	66	68	67	68
33	58	60	55	57	53	55	63	65	63	64	62	65
37	56	54	49	50	49	50	59	58	61	59	59	60
40	52	50	47	45	45	46	55	53	57	56	57	56
42	52	47	43	42	44	44	52	49	57	54	54	54
44	45	43	39	39	42	41	45	45	50	52	52	51
47	38	39	33	34	42	38	40	40	47	49	50	47
50	32	34	31	30	36	34	33	34	46	46	46	43
52	31	31	25	27	32	32	28	31	43	44	42	41
55	29	27	25	24	28	29	26	26	41	41	36	37
58	22	23	21	20	24	26	23	22	38	38	32	34
60	17	21	18	18	22	24	20	20	37	37	30	32
63	17	17	17	16	22	21	19	16	33	34	25	28
A	143.30		109.15		69.71		155.66		21.16		76.62	
B(K)	-20277.051		-14731.423		-8948.565		-21996.926		-1694.496		-10264.864	
C(K <sup>-1</sup> )	-0.238		-0.186		-0.119		-0.260		-0.037		-0.127	
AAD%	3.6%		1.8%		2.1%		1.8%		1.2%		1.9%	
SD	0.7		0.6		0.3		1.1		1.6		1.4	

Figs. 4.17 and 4.18 show the relationship between viscosity and temperature using the modified Andrade's equation for the 5 years storage age of  $L_1$  and 6 years storage age of  $L_8$  sample.



**Fig.(4.17):** Measured and calculated values of dynamic viscosity of olive oil of  $L_1$  using modified Andrade's equation



**Fig.(4.18):** Measured and calculated values of dynamic viscosity of olive oil of  $L_8$  using modified Andrade's equation

The values of AAD% when new constants are substituted into Andrade's equation are smaller than that of Abramovic's equations as given in Table 4.12. Taking 13 years storage age of  $L_1$  as an example, one can notice from Tables (4.7 - 4.12) that AAD% is 15.3% for Abramovic's equation (formula1)  $[\log \eta = \frac{A}{T} - B]$  where it is 7.3% for modified Abramovic's equation (formula 1), 21.3% for Abramovic's equation (formula 2)  $[\eta = A - B \log t]$  and 3.0% for modified Abramovic's equation (formula 2), and 9.5 % for Andrade's equation  $[\ln \eta = A + \frac{B}{T} + CT]$  and 2.1 % for modified Andrade's equation.

The Abramovic's and Andrade's equations with modified constants were better in describing the viscosity – temperature relationship since low values of AAD% (relative to the same equations with the original constants) were obtained. However, the modification of these equations by imposing new constants did not produce an acceptable description to the experimental data because still they have high values of AAD%.

The using of Abramovic's and Andrade's equations failed to describe the viscosity temperature relationship. New proposed equations should be imposed to describe the experimental data.

In the following section, three new equations are proposed to describe the dynamic viscosity relationship of olive oil versus temperature.

### 4.5.3 Proposed Equations

#### a. Proposed equation (formula 1)

Three order polynomial of equation was proposed as:

$$\eta = A + Bt + Ct^2 + Dt^3 \quad (4.4)$$

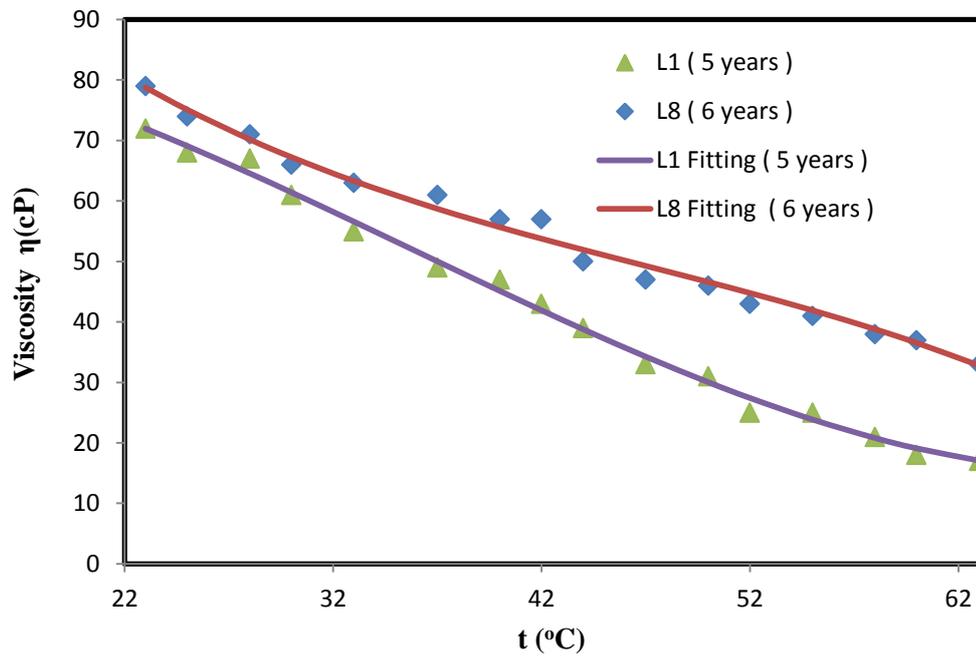
to fit the experimental dynamic viscosity - temperature relation of three samples from L<sub>1</sub> and three samples from L<sub>8</sub>. Table 4.13 shows the results of fitting.

**Table 4.13: The measured and calculated dynamic viscosity using the proposed equation (formula 1)  $[\eta = A + Bt + Ct^2 + Dt^3]$  of olive oil samples collected from  $L_1$  and  $L_8$  versus temperature**

t (°C)	$L_1$						$L_8$					
	Storage age: 2 years		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$
23	75	70	72	72	70	68	80	78	79	79	78	75
25	68	68	68	69	64	66	75	76	74	75	71	73
28	62	66	67	65	63	62	72	72	71	70	69	70
30	58	64	61	61	59	59	68	70	66	67	67	68
33	58	60	55	57	53	55	63	65	63	63	62	65
37	56	54	49	50	49	50	59	58	61	59	59	60
40	52	50	47	45	45	46	55	53	57	56	57	56
42	52	47	43	42	44	44	52	49	57	54	54	54
44	45	43	39	39	42	41	45	45	50	52	52	51
47	38	39	33	34	42	38	40	40	47	49	50	47
50	32	34	31	30	36	34	33	35	46	47	46	44
52	31	31	25	27	32	32	28	31	43	45	42	41
55	29	27	25	24	28	29	26	27	41	42	36	37
58	22	23	21	21	24	26	23	23	38	39	32	33
60	17	21	18	19	22	24	20	21	37	37	30	31
63	17	18	17	17	22	21	19	18	33	33	25	27
A(cP)	51.05		88.06		96.19		64.98		152.52		84.09	
B(cP/°C)	2.616		0.257		-1.077		2.543		-4.812		0.150	
C(cP/°C <sup>2</sup> )	-0.094		-0.053		-0.008		-0.105		0.083		-0.029	
D(cP/°C <sup>3</sup> )	0.00071		0.00049		0.00010		0.00083		-0.00059		0.00019	
AAD%	0.2%		0.3%		0.2%		0.1%		0.2%		0.1%	
SD	0.7		0.7		0.1		0.1		0.4		0.5	

AAD% are relatively very small for all samples, they range from 0.1% to 0.3%. This result implies that the proposed formula 1 is suitable to describe the viscosity - temperature relationship.

Fig. 4.19 shows the experimental and calculated values of the dynamic viscosity versus temperature for two olive oil samples.



**Fig.(4.19):** Measured and calculated values of dynamic viscosity using proposed equation (formula 1) fit for olive oil sample from L<sub>1</sub> and L<sub>8</sub>

One can observe from Fig 4.19 that the fitted lines coincide with most of the experimental data.

### **b. Proposed equation (formula 2)**

The measured dynamic viscosity versus temperature was fitted using the proposed formula 2:

$$\eta = A \ln(-B \ln t) \quad (4.5)$$

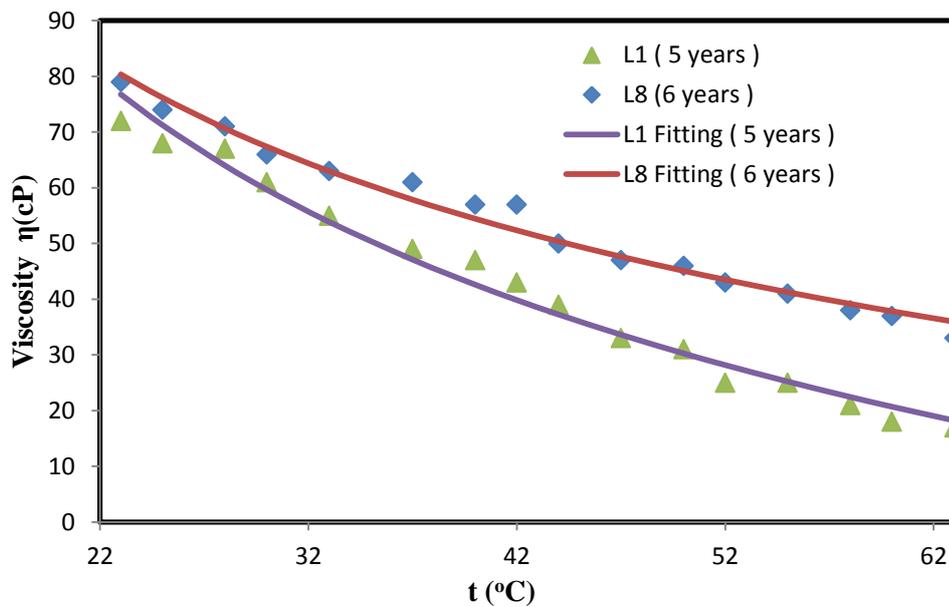
Results of this fitting for L<sub>1</sub> and L<sub>8</sub> samples are given in Table 4.14.

**Table 4.14: The measured and calculated dynamic viscosity using proposed equation (formula 2) [ $\eta = A \ln(-B \ln t)$ ] of olive oil samples collected from  $L_1$  and  $L_8$  versus temperature**

t (°C)	$L_1$						$L_8$					
	Storage age: 2 years		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$
23	75	78	72	77	70	72	80	86	79	80	78	81
25	68	72	68	71	64	68	75	80	74	76	71	76
28	62	66	67	64	63	62	72	72	71	71	69	70
30	58	61	61	60	59	58	68	67	66	67	67	67
33	58	56	55	54	53	53	63	61	63	63	62	62
37	56	49	49	47	49	48	59	53	61	58	59	56
40	52	45	47	43	45	44	55	48	57	54	57	52
42	52	43	43	40	44	42	52	45	57	52	54	50
44	45	40	39	37	42	40	45	42	50	50	52	48
47	38	37	33	34	42	37	40	38	47	48	50	45
50	32	33	31	30	36	34	33	34	46	45	46	42
52	31	31	25	28	32	32	28	32	43	44	42	40
55	29	29	25	25	28	30	26	29	41	41	36	38
58	22	26	21	22	24	27	23	26	38	39	32	36
60	17	24	18	21	22	26	20	24	37	38	30	34
63	17	22	17	18	22	24	19	21	33	36	25	32
A(cP)	-200.60		-210.07		-173.51		-232.78		-159.29		-173.75	
B	-0.216		-0.221		-0.210		-0.220		-0.193		-0.200	
AAD%	0.6%		0.7%		0.5%		0.5%		0.8%		0.4%	
SD	0.2		0.7		0.4		0.4		1.7		0.5	

Good prediction was obtained as given by Table 4.14, where very low values of AAD% are obtained. They are 0.6%, 0.7% and 0.5% for the 2 years, 5 years, and 13 years of  $L_1$ , and 0.5%, 0.8% and 0.4% for the 3 years, 6 years, and 10 years of  $L_8$ . The proposed equation (formula 2) is suitable in describing the viscosity versus temperature relationship.

Experimental and calculated data using the proposed equation (formula 2) of viscosity versus temperature for samples from  $L_1$  and  $L_8$  are shown in Fig.4.20.



**Fig.(4.20):** Measured and calculated values of dynamic viscosity using proposed equation (formula 2) fit for olive oil sample from  $L_1$  and  $L_8$

**c. Proposed equation (formula 3)**

The experimental results for dynamic viscosity versus temperature for four olive oil samples were fitted using the following proposed formula 3:

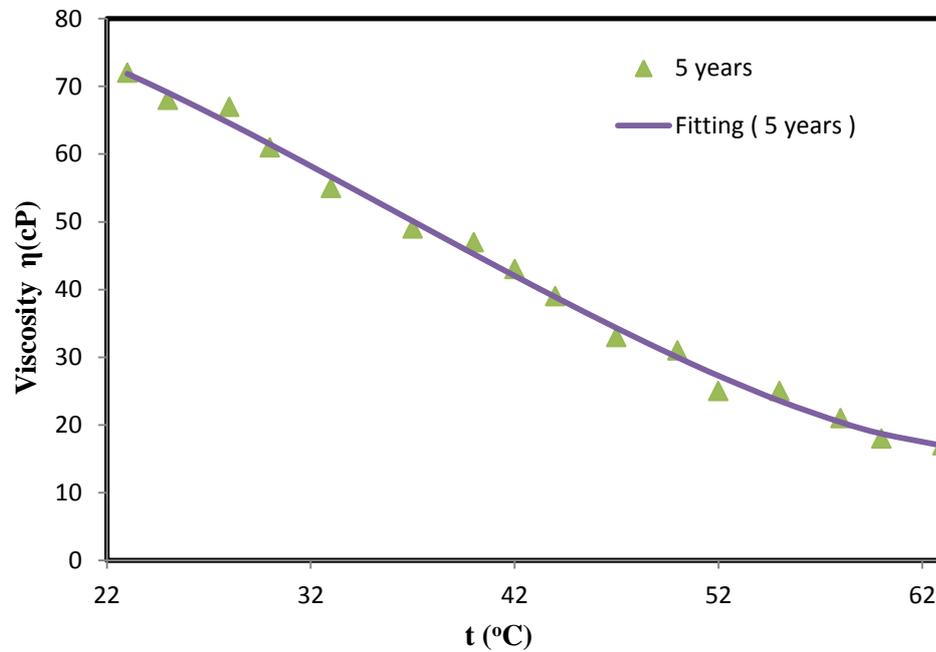
$$\eta = A + Bt + Ct^2 + Dt^3 + Et^4 + Ft^5 \quad (4.6)$$

The results of this fitting in addition to experimental values, constants A, B, C, D, E, F, AAD%, and SD are given in Table 4.15.

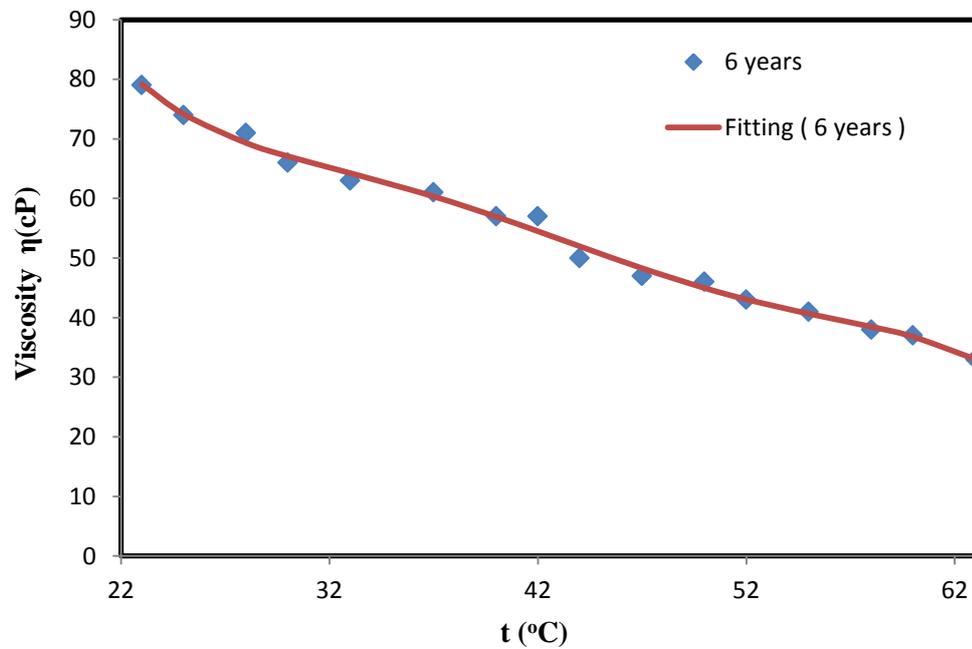
**Table 4.15: The measured and calculated dynamic viscosity using proposed equation (formula 3)  $[\eta = A + Bt + Ct^2 + Dt^3 + Et^4 + Ft^5]$  of olive oil samples collected from L<sub>1</sub> and L<sub>8</sub> versus temperature**

t (°C)	L <sub>1</sub>						L <sub>8</sub>					
	Storage age: 2 years		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$	$\eta_{exp}(cP)$	$\eta_{cal}(cP)$
23	75	75	72	72	70	69	80	80	79	79	78	77
25	68	67	68	69	64	67	75	75	74	74	71	73
28	62	61	67	65	63	62	72	70	71	69	69	68
30	58	60	61	61	59	58	68	68	66	67	67	66
33	58	58	55	57	53	54	63	65	63	64	62	63
37	56	56	49	50	49	49	59	59	61	60	59	59
40	52	52	47	45	45	46	55	54	57	57	57	57
42	52	49	43	42	44	44	52	50	57	54	54	55
44	45	46	39	39	42	43	45	46	50	52	52	53
47	38	40	33	34	42	40	40	40	47	48	50	49
50	32	34	31	30	36	36	33	33	46	45	46	45
52	31	30	25	27	32	33	28	30	43	43	42	42
55	29	26	25	24	28	28	26	25	41	41	36	37
58	22	22	21	20	24	24	23	22	38	38	32	32
60	17	20	18	19	22	22	20	21	37	37	30	29
63	17	16	17	17	22	22	19	19	33	33	25	25
A(cP)	1255.24		64.70		-411.16		724.97		665.72		205.43	
B(cp/K)	-144.179		3.464		70.235		-79.770		-72.015		-9.527	
C(cP/K <sup>2</sup> )	6.804		-0.226		-3.861		3.843		3.469		0.168	
D(cP/K <sup>3</sup> )	-0.156		0.005		0.100		-0.090		-0.083		0.002	
E(cP/K <sup>4</sup> )	0.001734		-0.000059		-0.001250		0.001019		0.000961		-0.000082	
F(cP/K <sup>5</sup> )	-0.000007485		0.000000297		0.000006041		-0.000004412		-0.000004357		0.000000617	
AAD%	0.4%		0.2%		0.1%		0.1%		0.1%		0.1%	
SD	0.7		0.3		0.2		0.1		0.2		0.3	

Figs. 4.21 and 4.22 show the experimental and calculated dynamic viscosity versus temperature of olive oil samples of  $L_1$  and  $L_8$ .



**Fig.(4.21):** Measured and calculated values of dynamic viscosity using proposed equation (formula 3) fit for olive oil sample from  $L_1$



**Fig.(4.22):** Measured and calculated values of dynamic viscosity using proposed equation (formula 3) fit for olive oil sample from  $L_8$

The proposed formula 3 is suitable for viscosity - temperature relationship. The values of AAD% are very small, they are 0.4%, 0.2% and 0.1% for the 2 years, 5 years, and 13 years storage ages of  $L_1$ , and 0.1%, 0.1%, and 0.1% for the 3 years, 6 years and 10 years storage ages of  $L_8$ . This indicates a good theoretical prediction.

The validity of good prediction is emphasized by Fig. 4.21 and 4.22 that show that both the experimental and calculated results are rather identical.

The results of fitting the viscosity temperature relationship using different equation are given in Table 4.16.

**Table 4.16: Values of AAD% obtained from fitting viscosity - temperature relationship using different equations**

Equations	AAD%						Average AAD%
	$L_1$			$L_8$			
	Storage age: 2 years	Storage age: 5 years	Storage age: 13 years	Storage age : 3 years	Storage age: 6 years	Storage age: 10 years	
Abramovic's formula 1	13.2%	12.0%	15.3%	15.8%	24.1%	22.1%	17.08%
Abramovic's formula 2	14.3%	18.2%	21.3%	17.1%	24.6%	22.8%	19.72%
Andrade's formula	9.6%	6.4%	9.5%	10.4%	19.7%	17.4%	12.17%
Modified Abramovic's formula 1	7.7%	5.7%	7.3%	6.8%	2.4%	4.6%	5.75%
Modified Abramovic's formula 2	6.3%	3.7%	3.0%	4.4%	3.5%	4.3%	4.20%
Modified Andrade's formula	3.6%	1.8%	2.1%	1.8%	1.2%	1.9%	2.07%
Our proposed formula 1	0.2%	0.3%	0.2%	0.1%	0.2%	0.1%	0.18%
Our proposed formula 2	0.6%	0.7%	0.5%	0.5%	0.8%	0.4%	0.58%
Our proposed formula 3	0.4%	0.2%	0.1%	0.1%	0.1%	0.1%	0.17%

The values of AAD% is very high when Abramovic's and Andrade's equations are used, the average values of AAD% are 17.08%, 19.72%, and 12.17% for Abramovic's 1, Abramovic's 2, and Andrade's equations respectively, these high values of AAD% indicate that these equation are not suitable to describe the viscosity - temperature relationship.

The modifications are done on these three equations, the values of AAD% are 5.57%, 4.20%, and 2.07% for modified Abramovic's 1, modified Abramovic's 2, and modified Andrade's equations respectively. These values are still high and these equations did not present a good fit for the viscosity - temperature relationship.

Our proposed equations presented the best fit with the lowest values of AAD%. These values are 0.18%, 0.58%, and 0.17% for our proposed equations.

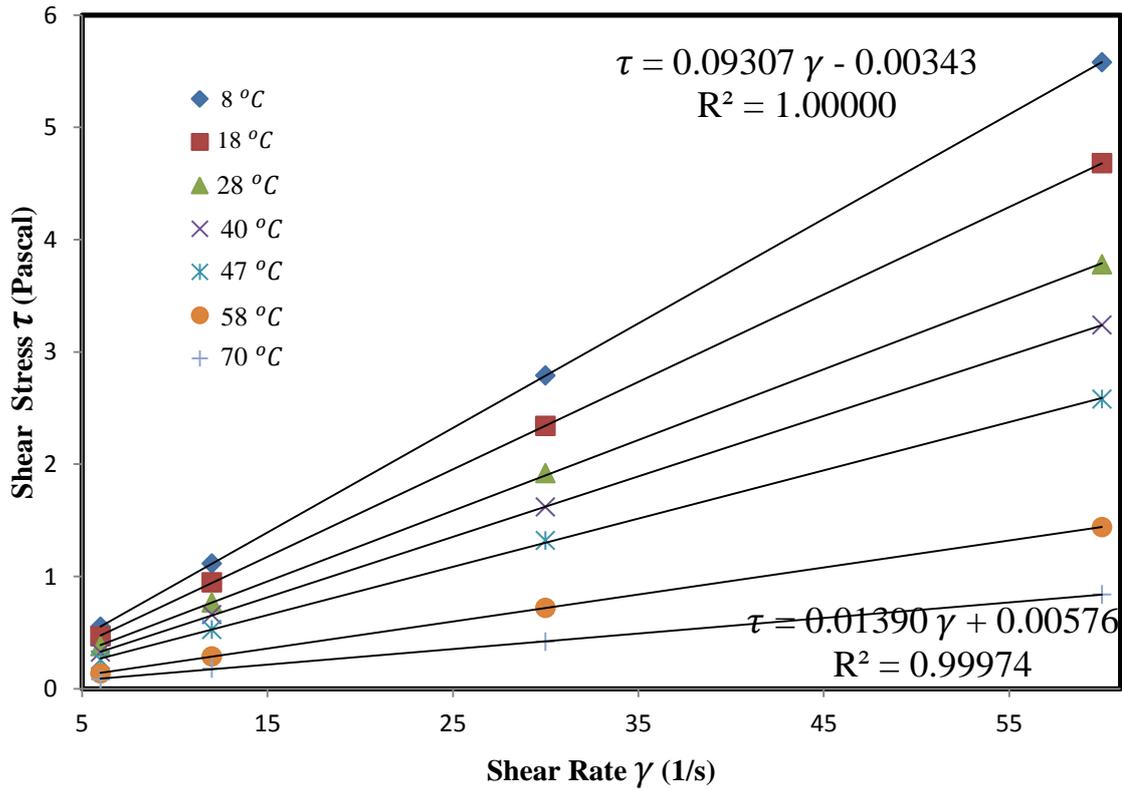
#### **4.6 Shear Stress Versus Shear Rate**

The power law model is used to check whether the behavior of the olive oil samples is Newtonian or Non - Newtonian. A linear fit was done to the experimental results by the power law equation ( $\tau = \eta\dot{\gamma}^n$ ) to find the value of the exponent  $n$ . The results of two samples from L<sub>1</sub> and L<sub>8</sub> are given in Table 4.17.

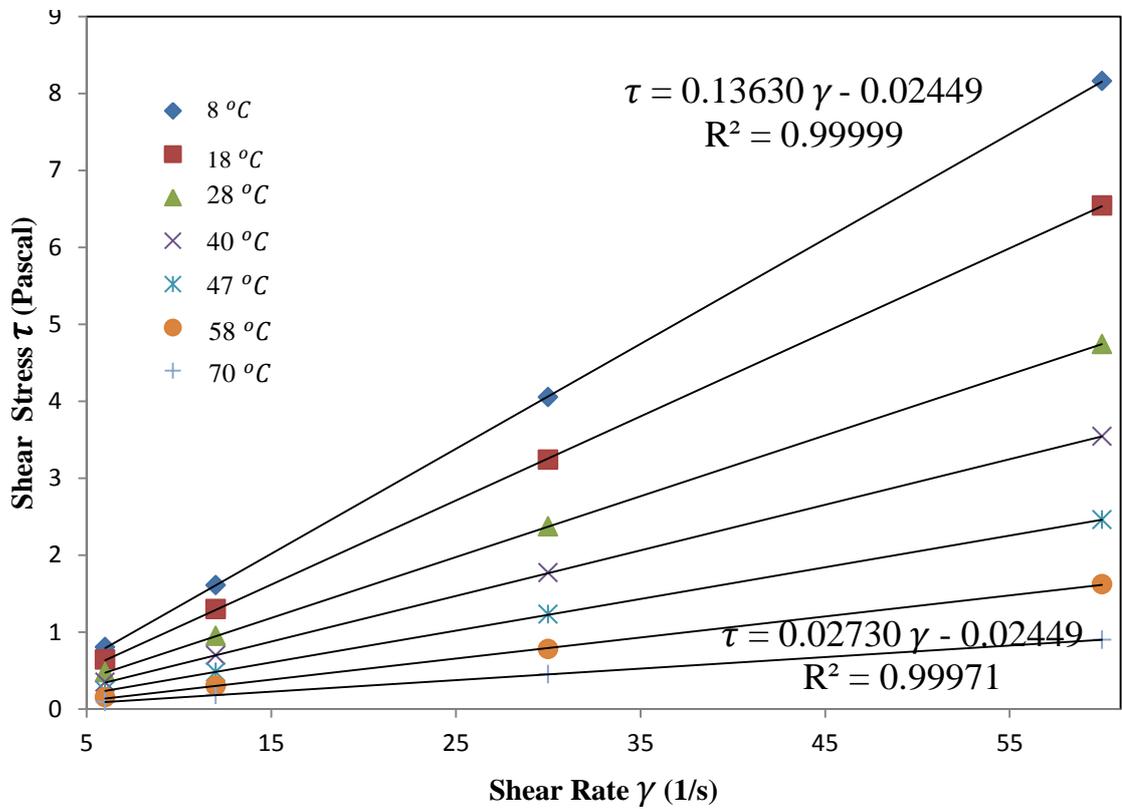
**Table 4.17: The results of power law fit to experimental data from L<sub>1</sub> and L<sub>8</sub>**

t (°C)	L <sub>1</sub>						L <sub>8</sub>					
	Storage age: 2 year		Storage age: 5 years		Storage age: 13 years		Storage age: 3 years		Storage age: 6 years		Storage age: 10 years	
	<i>n</i>	Error	<i>n</i>	Error	<i>n</i>	Error	<i>n</i>	Error	<i>n</i>	Error	<i>n</i>	Error
8	1.00	0.006	1.01	0.008	1.03	0.021	1.01	0.008	1.01	0.010	1.01	0.004
18	1.00	0.006	1.01	0.008	1.00	0.006	1.00	0.006	1.00	0.008	1.00	0.006
28	0.99	0.016	1.00	0.000	0.99	0.010	1.00	0.008	1.00	0.008	1.00	0.000
40	1.01	0.014	1.01	0.006	0.99	0.006	1.02	0.002	1.02	0.002	1.01	0.006
47	0.98	0.012	1.01	0.006	0.98	0.011	1.01	0.010	1.02	0.009	1.00	0.000
58	1.02	0.014	1.05	0.004	1.00	0.003	1.01	0.006	1.04	0.002	1.00	0.004
70	1.02	0.006	1.00	0.000	0.98	0.006	1.05	0.004	1.00	0.004	1.02	0.008

The relationships between shear stress and shear rate for L<sub>1</sub> (2 and 13 years storage age) for different temperatures are shown in Figs. 4.23 and 4.24, respectively.



**Fig.(4.23):** Relationship between shear stress and shear rate for olive oil sample from L<sub>1</sub> (2 year storage age) at different temperatures



**Fig.(4.24):** Relationship between shear stress and shear rate for olive oil sample from L<sub>1</sub> (13 years storage age) at different temperatures

Table 4.17 shows the value of  $n$  is closed to one within an accepted error bars. This means that the olive oil samples are Newtonian fluid.

Figs. 4.23 and 4.24 show that the relationship between shear stress and shear rate for olive oil is always linear at all temperatures. This is another indication of the Newtonian behavior of the olive oil samples as indicated by the simplest equation of Newton's law of viscosity.

## Chapter Five

### Discussion and conclusion

Three samples from  $L_1$  and three samples from  $L_8$  are selected to be analyzed. The reasons of choosing these two regions are:

Firstly: they are far enough from each other.

Secondly: the altitude are different, it is 350 m for  $L_1$ , and 890 m for  $L_8$ .

Thirdly: the quantities of rain are different for both regions, since we have different crops.

#### 5.1 Density

The olive oil density is shown to be decreasing linearly as a function of age. The average value of the measured result of density of olive oil samples is found to be  $0.9180 \text{ gm/cm}^3$ . This result is in a good agreement with Robert and his group value. They found the density of olive oil to be  $0.918 \text{ gm/cm}^3$  (Robert *et al.*, 1979).

#### 5.2 Refractive index

The average value of refractive index is measured to be 1.4708 while the standard value is 1.4677 - 1.4705 (International Olive Council, 2011).

Our values of refractive index are in good agreement with the standard ones. A linear fit showed that the refractive index is decreasing as a function of storage age.

#### 5.3 Acidity

The experimental results of acidity showed that the quality of olive oil samples varies from Extra virgin to Lampante olive oil according to Table (2.3). The samples showed that the acidity increase with storage age. For

example, the acidity was measured to be 0.44% (Extra virgin) for crop 2012 of L<sub>8</sub> and, 1.56% (Virgin) for crop 2008 of L<sub>7</sub>, 3.18% (Ordinary virgin) for crop 2001 of L<sub>6</sub>, and 9.94% (Lampante) for crop 1997 of L<sub>10</sub>.

Falque found that the value of the acidity of the extra - virgin olive oils is 0.39%. (Falque *et al.*, 2007). These values are close to the extra - virgin olive oils value of acidity of crop 2012 of L<sub>8</sub> (0.44%).

The equations from linear fit of density versus storage age, refractive index versus storage age, acidity versus storage age, and refractive index versus density are given in Table 5.1 for different regions.

**Table 5.1: The linear equations by linear fitting for density, refractive index and acidity versus storage age, and refractive index versus density for L<sub>1</sub> and L<sub>8</sub>**

Region		Equation
L <sub>1</sub>	Density as a function of storage age	$\rho = - 0.00009 t + 0.91908$
L <sub>8</sub>		$\rho = - 0.00024 t + 0.91946$
L <sub>1</sub>	Refractive index as a function of storage age	$n = - 0.00008 t + 1.47166$
L <sub>8</sub>		$n = - 0.00017 t + 1.47175$
L <sub>1</sub>	FFA% as a function of storage age	$FFA\% = 0.73817 t - 0.13134$
L <sub>8</sub>		$FFA\% = 0.77305 t - 0.50782$
L <sub>1</sub>	Refractive index as a function of density	$n = 0.75966 \rho + 0.77342$
L <sub>8</sub>		$n = 0.65741 \rho + 0.86723$
L <sub>1</sub>	Viscosity as a function of storage age	$\eta = - 0.55263 t + 76.63158$
L <sub>8</sub>		$\eta = - 0.55465 t + 69.81658$

where  $\rho$  is the density in (gm/cm<sup>3</sup>),  $t$  is the storage age in years, FFA% is the free fatty acid composition, and  $n$  is the refractive index.

The coefficients of the linear equations relating density and storage age, refractive index and storage age, FFA% and storage age, and refractive index and density differ from one region to another as seen from Table 5.1.

#### **5.4 Viscosity**

Many theoretical predictions were suggested to describe the relationship between dynamic viscosity and temperature of olive oil. Three models were tested but failed to describe the relationship because storage age was not taken into consideration. Modifications were done on these equations to improve the theoretical prediction, but still the equations are not proper.

Three new equations were proposed to describe the viscosity - temperature relationship. Our proposed equations presented the best fit for the experimental results.

The results of viscosity measurements showed that the behavior of olive oil is Newtonian since the value of the flow indices ( $n$ ) is very close to one.

Our work is not consistent with the work of Adnan and his group who found the flow index of olive oil to be 0.84, and so olive oil was considered Non - Newtonian (Adnan *et al.*,2009).

## References

- Abramovic H., klofutar G., *"The temperature dependence of dynamic viscosity for some vegetables oils"*, *Acta Chim. Slov.* **45**(1), 69-77 (1998).
- Adolfo, V. F. and Ana, G B., *"Study of the evolution of the physicochemical and structural characteristics of olive and sunflower oil after heating at frying temperatures"*, *Food Chemistry* **98**, 214-219 (2006).
- Ahmad M., Amran A., Giap S.G.E. and Nik W. M., *"The Assessment of Rheological Model Reliability In Lubricating Behavior of Vegetable Oils"*, *Engineering e-Transaction*, **4**(2), 81-89 (2009).
- Adnan Q., Ahmad M., Akhtar N., Farzana K. and Mehmood A., *"Rheological Studies and Characterization of Different Oils"*, *J. Chem. Soc. Pak.*, **31**(2), 201 - 206 (2009).
- Almeida S., Belchiora C., Nascimento M., Vieirab L. and Fleuryb G., *"Performance of a Diesel Generator Fuelled With Palm Oil"*, *FUEL* **81** (16), p.p 2097–2102(2002).
- Andrade E. N. C., *Nature*, **125**, 309-318(1930).
- Angerosa F. , Mostallino R. , Basti C. , Vito R. , *"Influence of malaxation temperature and time on the quality of virgin olive oils"*, *Food Chemistry*, **72** 19-28 (2001).
- Bayrak Y., Iscan M., and Topallar H., *"Kinetics of Autoxidative Polymerization of Sun Flower Seed Oil"*, *Tr. J. of chemistry*, **21**, 118-125 (1997).

- Bird R.B., Armstrong R.C., and Hassager O., "*Dynamics of Polymeric Liquids*", Vol. 1, Wiley, New York, (1987).
- Bruwer J. J., Boshoff B. D., Hugo F. J. CDuPlessis., L. M., Fuls JHawkins., C., Vander Walt A. N., and Engelbert A., "*The Utilization of Sunflower Seed Oil as Renewable Fuel Diesel Engines. In Agricultural Energy*", ASAE, 2,4 -81(1981).
- Chhabra R.P., and Richardson J.F., "*Non - Newtonian Flow in the Process Industries*", *Butterworth Heinemann Publishers*; (1999).
- Clements C., Craig-Schmidt M., Fasina O. O. and Hallman H., "*Predicting temperature - dependence viscosity of vegetable oils from fatty acid composition*", *Journal of the American Oil Chemists' Society*, **83**(10), 899-903 (2006).
- De Guzman J., "*Relation between fluidity and heat of fusion*", *Anales Soc. Espan. Fis. Y. Quim.* **11**, 353-362 (1913).
- Duhne C. R., "*Viscosity - temperature correlations for liquids*", *Chem. Eng.*, **86** (15), 83 (1979).
- Dutt N., Ghosh T. K., Prasad D. H. L., Rani K. Y., and Viswanath D. S., "*Viscosity of Liquids Theory, Estimation, Experiment, and Data*", 444-553 (2007).
- Eromosele C., and Paschal N., "*Characterization and Viscosity Parameters of Seed Oils from Wild Plants*", *Bioresource Technology* **86** 203–205(2003).
- Falque E. and Mendez A. I., "*Effect of storage time and container type on the quality of extra-virgin olive oil*", *Science Direct Food Control*, **18**, 521–529, (2007).

- Farhoosh R., Niazmand R., Rezaei M. and Sarabi M., *"Kinetic Parameter Determination of Vegetable Oil Oxidation Under Rancimat Test Conditions"*, *Eur. J. Lipid Sci. Technol.*, **110**, 587–592(2008).
- Fasina O., Colley Z., *"Viscosity and Specific Heat of Vegetable Oils as a Function of Temperature: 35°C to 180°C"*, *Int. J. Food Prop.*, **11**(4): 738-746. (2008).
- Goering C. E., A. W. Schwab, M. J. Daugherty, E. H. Pryde, and A. J. Heakin, *"Fuel Properties of Eleven Vegetable Oils"*, *ASAE*, 81-3579(1981).
- Hsieh F.H., Rodenbush C.M. and Viswanath D.S., *"Density and Viscosity of Vegetable Oils"*, *JAOCS.*, **76**, 1415–1419 (1999).
- [http://www.aoac.org/iMIS15\\_Prod/AOAC](http://www.aoac.org/iMIS15_Prod/AOAC), AOAC, **Association of Official Agricultural Chemists,1997.**
- <http://www.naooa.org/>, NAOOA , *North American Olive Oil Association*, 2013.
- IOOC, International Olive oil Council **"Trade standards applying to olive oil and olive pomace oil"**, E. /Conv. /Doc. no.1 /16 November 2000. International Olive Oil Council, Madrid, Spain.
- James F., *"Rheological Methods in Food Process Engineering"*, 2<sup>nd</sup> Ed., 1-35 (1996).
- Larson R. G., *"The Structure and Rheology of Complex Fluids"*, Oxford University Press, Oxford and New York, (1999).
- Lupi F.R. , Gabriele D., Facciolo D. , Baldino N. , Seta L. , de Cindio B. , *"Effect of organogelator and fat source on rheological*

*properties of olive oil-based organogels" Food Research*

*International*; **46**(1), 177–184 (2012).

- Martin A. and Bustamante P., "*Physical Pharmacy*", 4<sup>th</sup> Ed, Lippincot, USA, P. 453 (2004)
- Middleman S., "*The Flow of High Polymers*", Wiley Interscience, New York, (1968).
- Munson B.R., Young D.F., and Okiishi T.H., "*Fundamentals of Fluid Mechanics*", Wiley, New York, (1998).
- Natarajan G. and Viswanath D. S., "**Data book on viscosity of liquids**", Hemisphere, New York (1989).
- Nierat T., Musameh S., and Abdelraziq I., "*Temperature and Storage Age (Yearly Basis)-Dependence of Olive Oil Viscosity in Different Locations in Palestine*", J. Mater. Environ. Sci. **5** (1): 245-254 (2014).
- Nierat T., Musameh S., and Abdelraziq I., "*Temperature and Storage Age (Weekly Basis)-Dependence of Olive Oil Viscosity in Different Locations in Palestine*", MSAIJ **9**(11): 445-451 (2013).
- Nierat T., "*Temperature and Storage Age-Dependence of Olive Oil Viscosity in Different Locations in Palestine* ", master thesis, An-Najah National University, (2012).
- Nouredini H., Teoh B.C. and Davis Clements L., "*Viseosities of Vegetable Oils and Fatty Acids*", JAOCS, **69**,12 (1992).
- Patil T., Butala D., Raghunathan T., and Shankar H., "*Thermal Hydrolysis of Vegetable Oils and Fats. 1. Reaction Kinetics*", Ind. Eng. Chem. Res., **27** (5), p 727–735(1988).

- Quinchia L., Delgado M., Franco J., Spikes H. and Gallegos C., "**Low - Temperature Flow Behaviour of Vegetable Oil-Based Lubricants**", *Industrial Crops and Products*, **37** p 383– 388(2012).
- Reid J. F., Hansen A. C., and C. E. Goering, "**Quantifying Diesel Injector Coking with Computer Vision**"., *ASAE*,**32**(5), 1503-1506(1989).
- Remington, "**The Science and Practice of Pharmacy**", Vol. **1**, 21<sup>st</sup> Ed, Lippincot, USA, P. 338 (2006).
- Robert C., "**CRC Handbook of Chemistry and Physics**", David R. Lide; (1979).
- Sims R.E., Raine R., and McLeod R., "**Rapeseed Oil as a Fuel for Diesel Engines**". *SAE-Australia. Paper presented at the National Conference on Fuels from Crops of the Society of Automotive Engineers – Australia* (1981).
- Skelland A.H.P., "**Non - Newtonian Flow and Heat Transfer**", John Wiley and Sons Inc.; (1967).
- Stanciu I., "**A New Viscosity - Temperature Relationship for Vegetable Oil**", *J. Petroleum Technology and Alternative Fuels*, **3**(2), pp. 19-23, (2012).
- The World Bank 2012, West Bank and Gaza Program," **Brief Overview of the Olive and the Olive Oil Sector in the Palestinian Territories**", <http://go.worldbank.org/MBK9GU1TD0>
- Themelis, N. J., "**Transport and chemical rate phenomena**" , Basel : Gordon and Breach 1995

- Toscano G., Riva G., FoppaPedretti E. and Duca D., "*Vegetable Oil and Fat Viscosity Forecast Models Based on Iodine Number and Aponification Number*", *biomass and bioenergy*, **46**p.p 511-516(2012).
- Vogel H., "*Das temperaturabhängigkeitsgesetz der viskosität von flüssigkeiten*", *Physics*. **22**, 645-646 (1921).
- Yalcin H, Toker O.S., and Dogan M., "*Effect of Oil Type and Fatty Acid Composition on Dynamic and Steady Shear Rheology Of Vegetable Oils*", *J Oleo Sci.*, **61**(4):181-7(2012).

## Appendix A

### Density Results

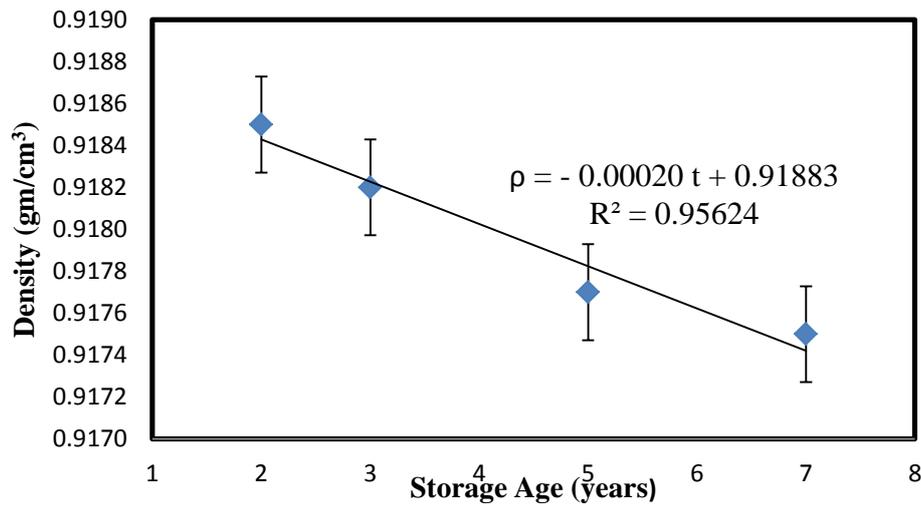
The densities in ( $\text{gm/cm}^3$ ) of the olive oils samples for all regions and for different storage ages are given in Table (A.1).

**Table (A.1): Measured density of olive oil samples in different regions and for different storage ages**

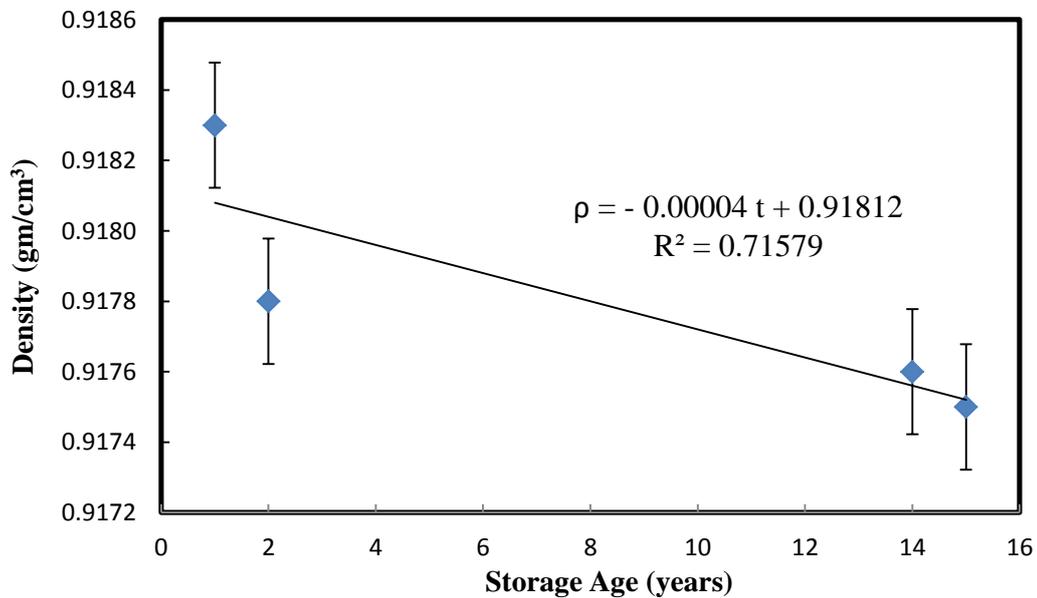
Storage Age (years)	L <sub>2</sub>	L <sub>3</sub>	L <sub>4</sub>	L <sub>5</sub>	L <sub>6</sub>	L <sub>7</sub>	L <sub>9</sub>	L <sub>10</sub>	L <sub>11</sub>	L <sub>12</sub>	L <sub>13</sub>	Average
1	0.9189	0.9183	0.9184	0.9189								0.9187
2	0.9189	0.9178	0.9184				0.9185			0.9185		0.9185
3	0.9185		0.9183	0.9188	0.9183	0.9188	0.9182		0.9185	0.9185	0.9188	0.9185
4			0.9182	0.9186	0.9182	0.9186					0.9185	0.9184
5						0.9185	0.9177					0.9182
6				0.9186		0.9181				0.9180		0.9181
7				0.9182			0.9175					0.9178
8									0.9180			0.9178
12					0.9177						0.9179	0.9178
13					0.9176							0.9178
14	0.9160	0.9176			0.9176			0.9179			0.9179	0.9174
15	0.9155	0.9175			0.9175				0.9174		0.9179	0.9172
16					0.9175			0.9165			0.9175	0.9173
19						0.9174						0.9174
Average	0.9176	0.9178	0.9183	0.9186	0.9178	0.9183	0.9180	0.9172	0.9180	0.9183	0.9181	0.9180

Table (A.1) shows the average density in each region, for each storage age in years.

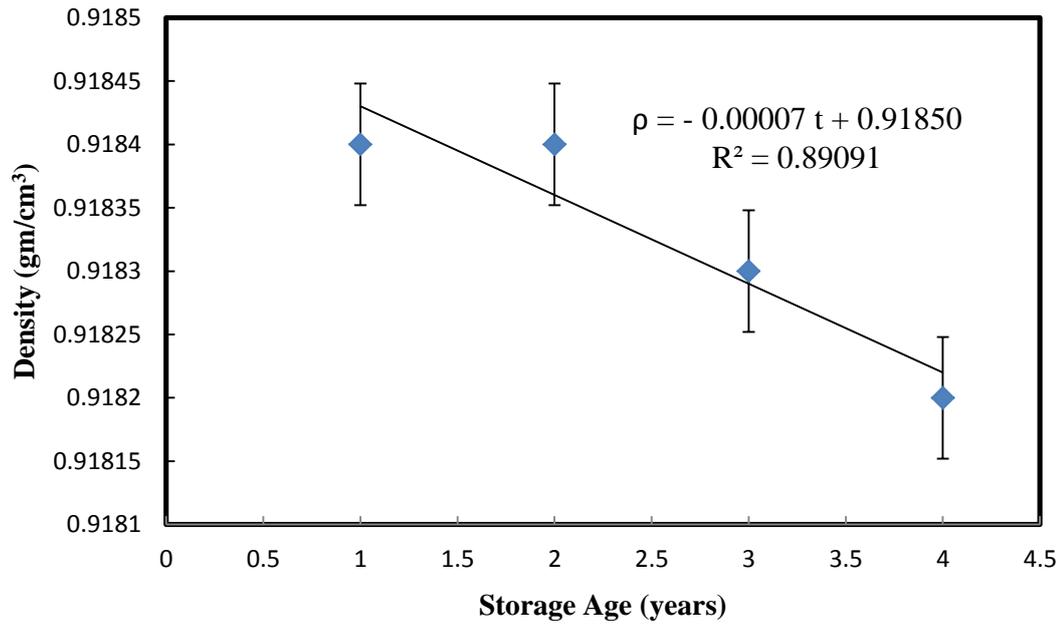
The relationship between density of olive oil samples and storage age are shown in figures (A.1 - A.11).



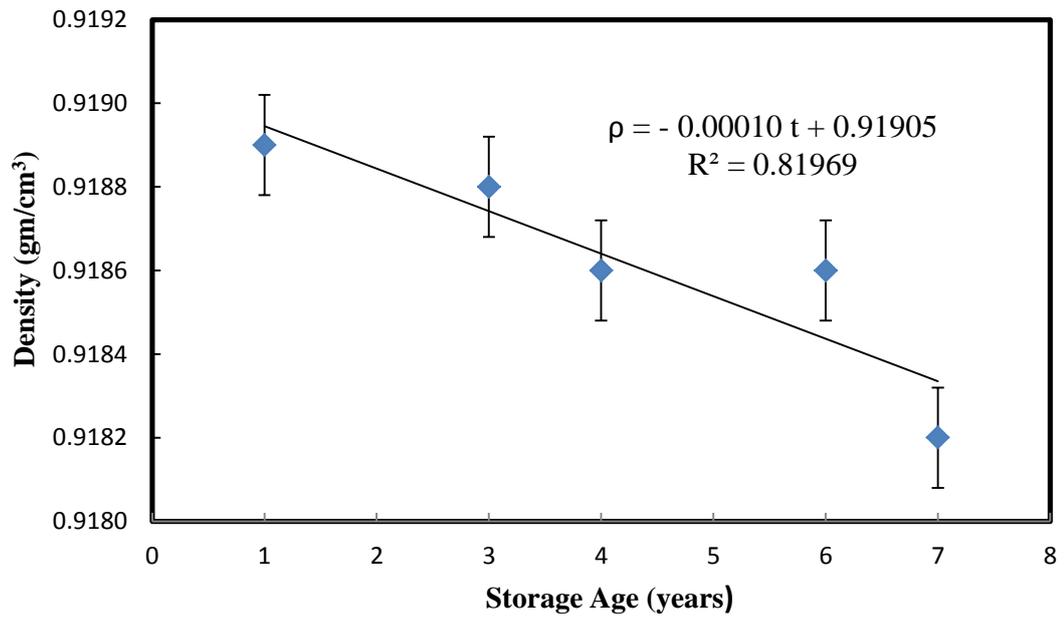
**Fig. A.1:** Measured density versus storage age of olive oil samples of L<sub>2</sub>



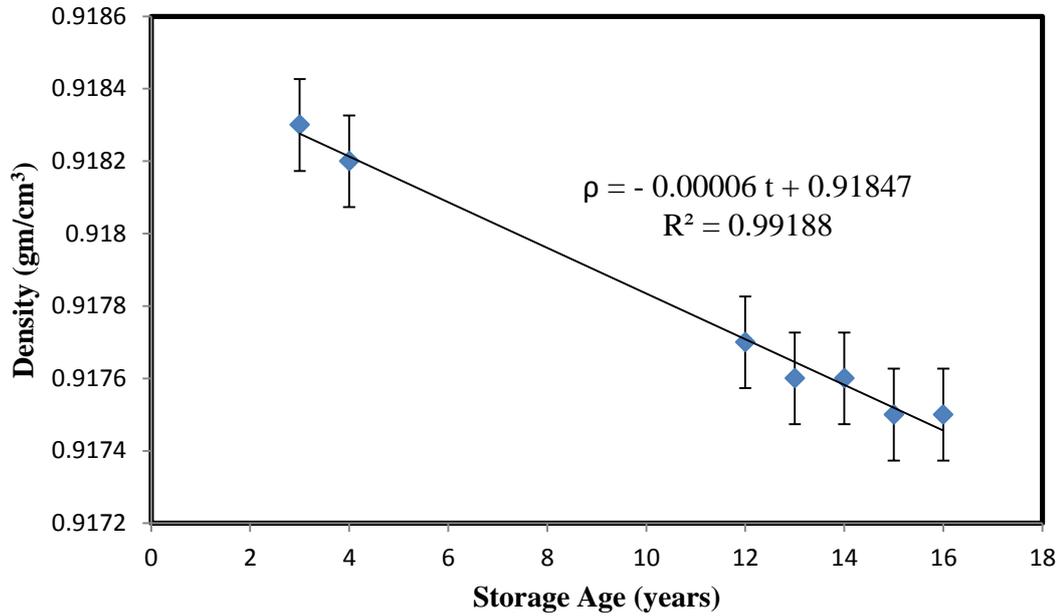
**Fig. A.2:** Measured density versus storage age of olive oil samples of L<sub>3</sub>



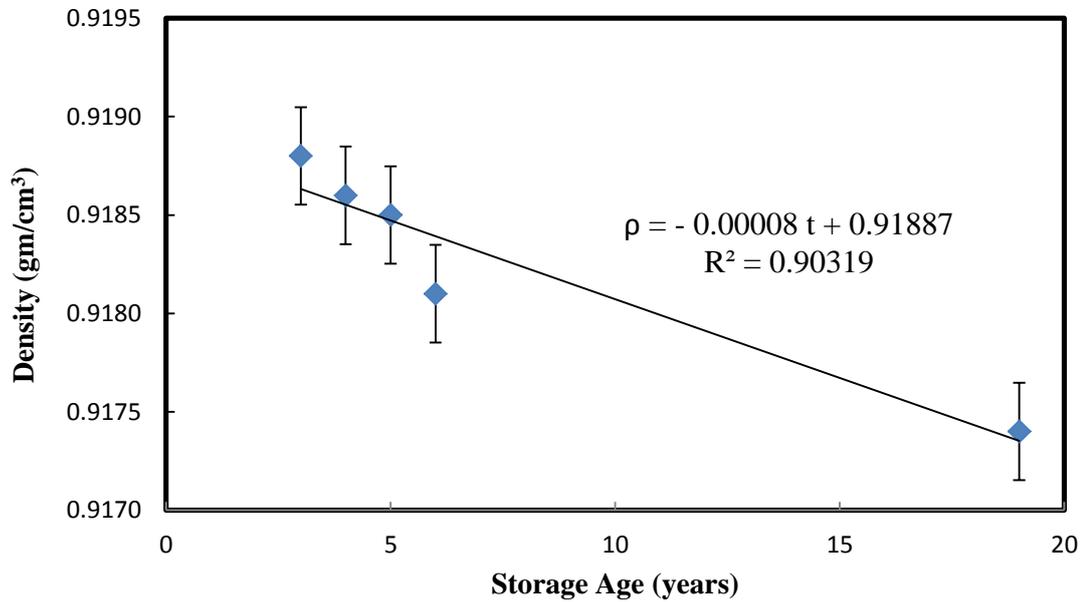
**Fig. A.3:** Measured density versus storage age of olive oil samples of L<sub>4</sub>



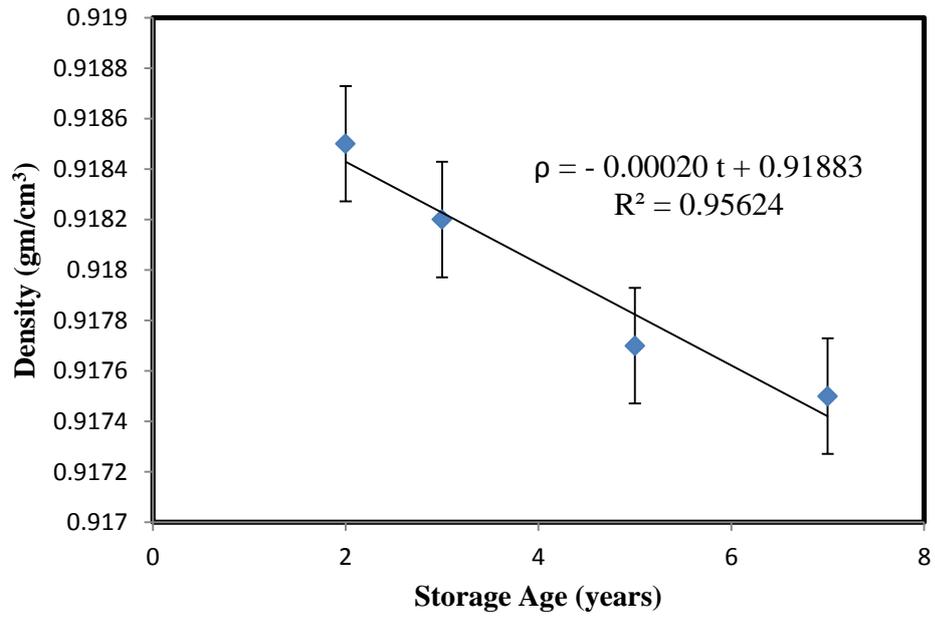
**Fig. A.4:** Measured density versus storage age of olive oil samples of L<sub>5</sub>



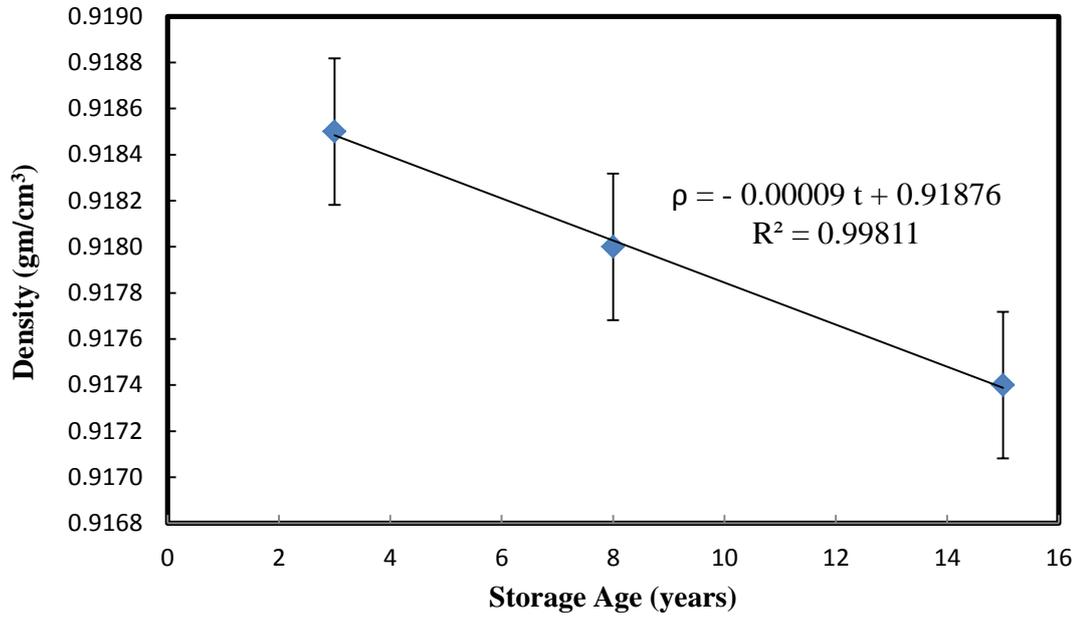
**Fig. A.5:** Measured density versus storage age of olive oil samples of L<sub>6</sub>



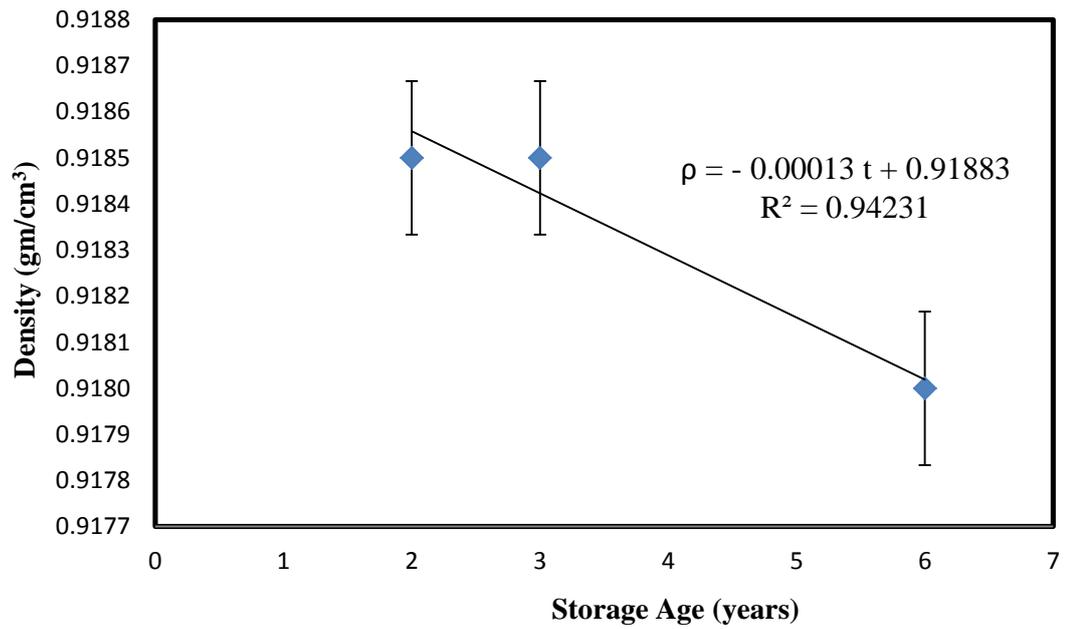
**Fig. A.6:** Measured density versus storage age of olive oil samples of L<sub>7</sub>



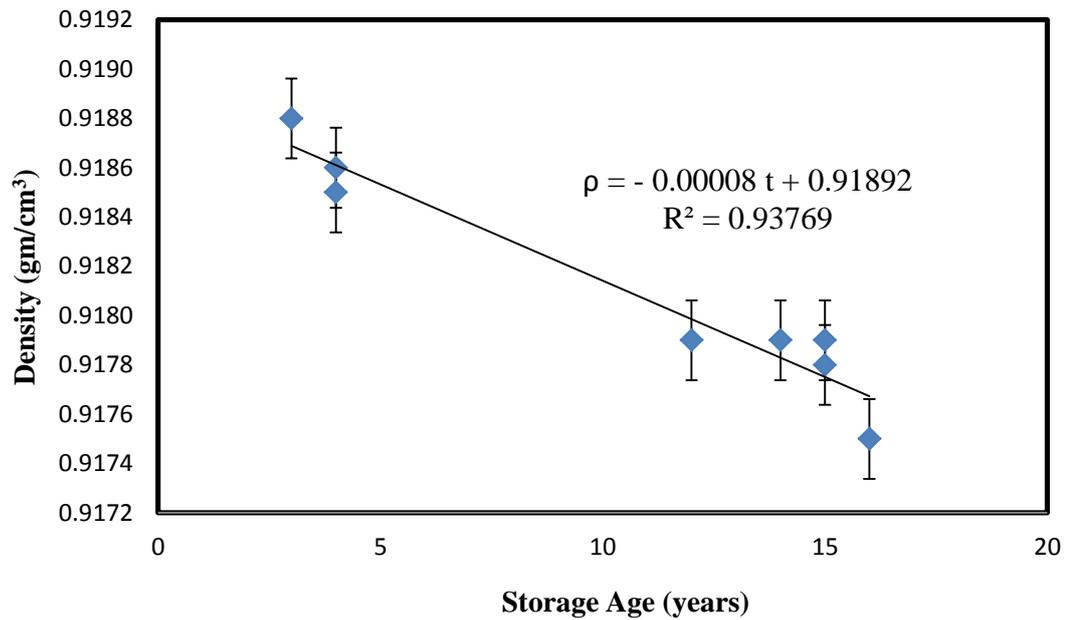
**Fig. A.7:** Measured density versus storage age of olive oil samples of L<sub>9</sub>



**Fig. A.8:** Measured density versus storage age of olive oil samples of L<sub>11</sub>



**Fig. A.9:** Measured density versus storage age of olive oil samples of L<sub>12</sub>



**Fig. A.10:** Measured density versus storage age of olive oil samples of L<sub>13</sub>

## Appendix B

### Refractive Index Results

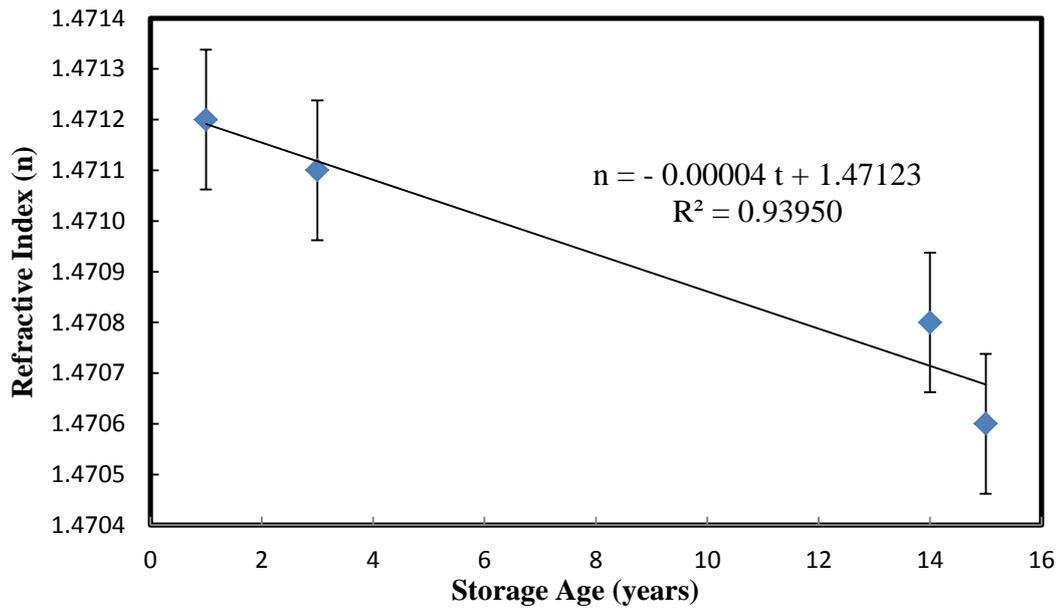
The refractive index of the olive oils samples for all regions and for different storage ages are given in Table (B.1).

**Table (B.1): Measured refractive index of olive oil samples in different region and for different storage ages**

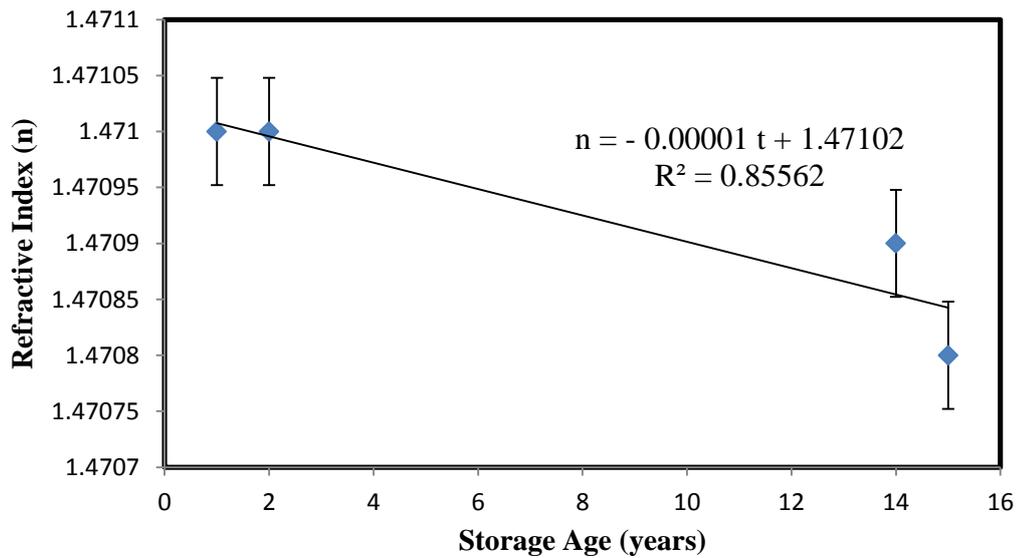
Storage Age (years)	L <sub>2</sub>	L <sub>3</sub>	L <sub>4</sub>	L <sub>5</sub>	L <sub>6</sub>	L <sub>7</sub>	L <sub>9</sub>	L <sub>10</sub>	L <sub>11</sub>	L <sub>12</sub>	L <sub>13</sub>	Average
1	1.4712	1.4710	1.4717	1.4716								1.4715
2		1.4710	1.4716				1.4713			1.4713		1.4713
3	1.4711		1.4711	1.4714	1.4711	1.4715	1.4713		1.4713	1.4712	1.4711	1.4712
4			1.4707	1.4710		1.4714					1.4705	1.4710
5						1.4712	1.4712					1.4711
6				1.4708		1.4712				1.4709		1.4709
7				1.4707			1.4709					1.4708
8				1.4707					1.4711			1.4708
12					1.4705						1.4703	1.4704
13					1.4702							1.4705
14	1.4708	1.4709			1.4701			1.4708			1.4701	1.4706
15	1.4706	1.4708			1.4704			1.4698	1.4710		1.4702	1.4705
16					1.4700			1.4690			1.4700	1.4698
19						1.4711						1.4711
Average	1.4709	1.4709	1.4713	1.4710	1.4704	1.4713	1.4712	1.4699	1.4711	1.4711	1.4704	1.4708

Table (B.1) shows the average refractive index in all regions, for each storage age in years.

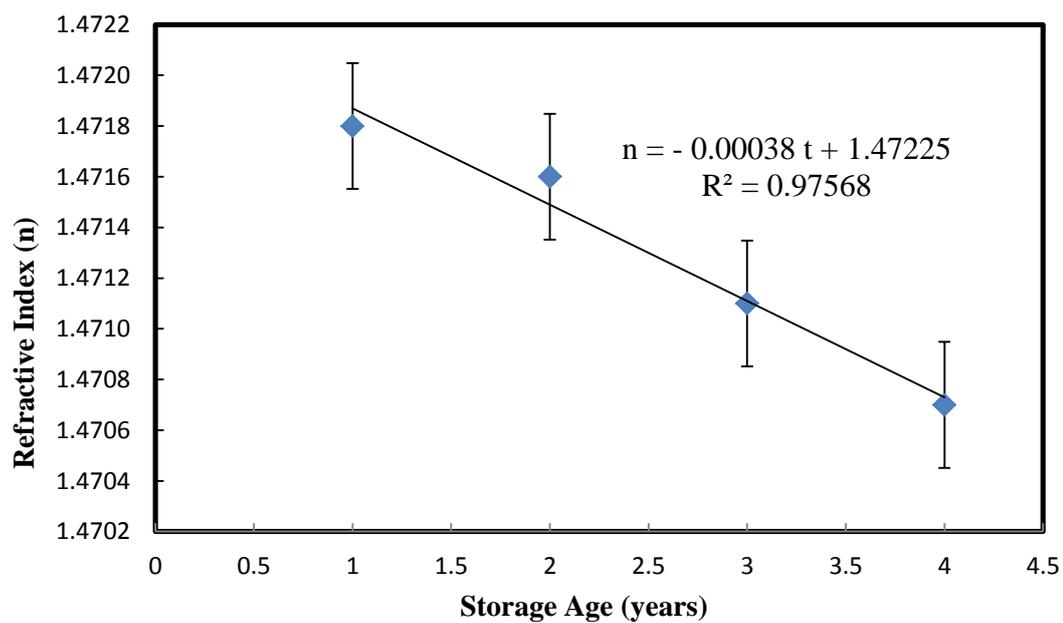
The relationship between refractive index of olive oil samples and storage age is shown in figures (B.1 - B.11).



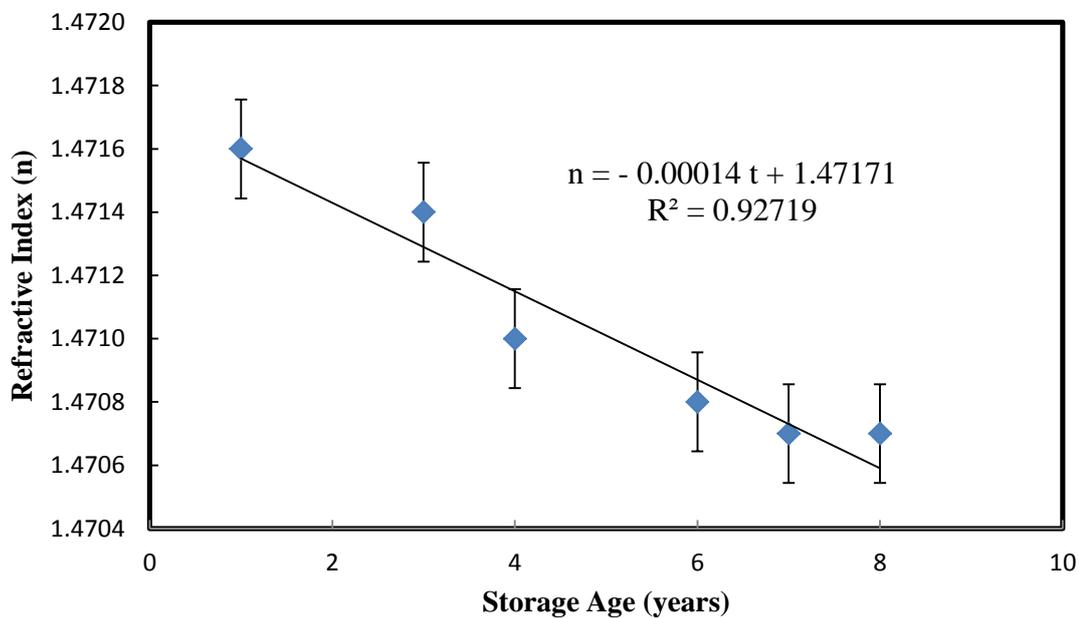
**Fig. B.1:** Measured refractive index versus storage age of olive oil samples of L<sub>2</sub>



**Fig. B.2:** Measured refractive index versus storage age of olive oil samples of L<sub>3</sub>



**Fig. B.3:** Measured refractive index versus storage age of olive oil samples of L<sub>4</sub>



**Fig. B.4:** Measured refractive index versus storage age of olive oil samples of L<sub>5</sub>

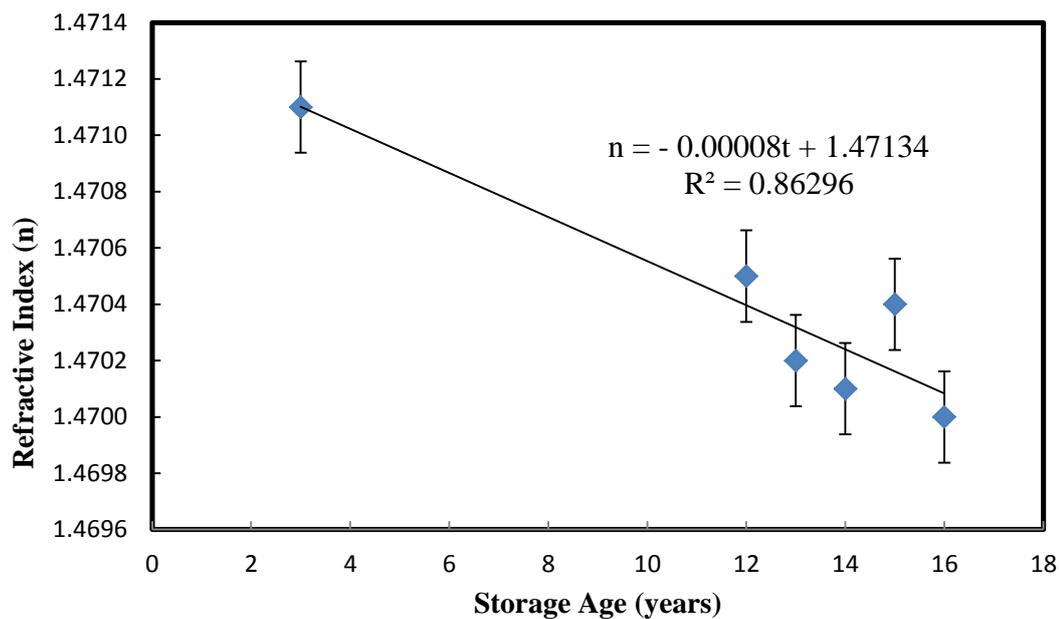


Fig. B.5: Measured refractive index versus storage age of olive oil samples of L<sub>6</sub>

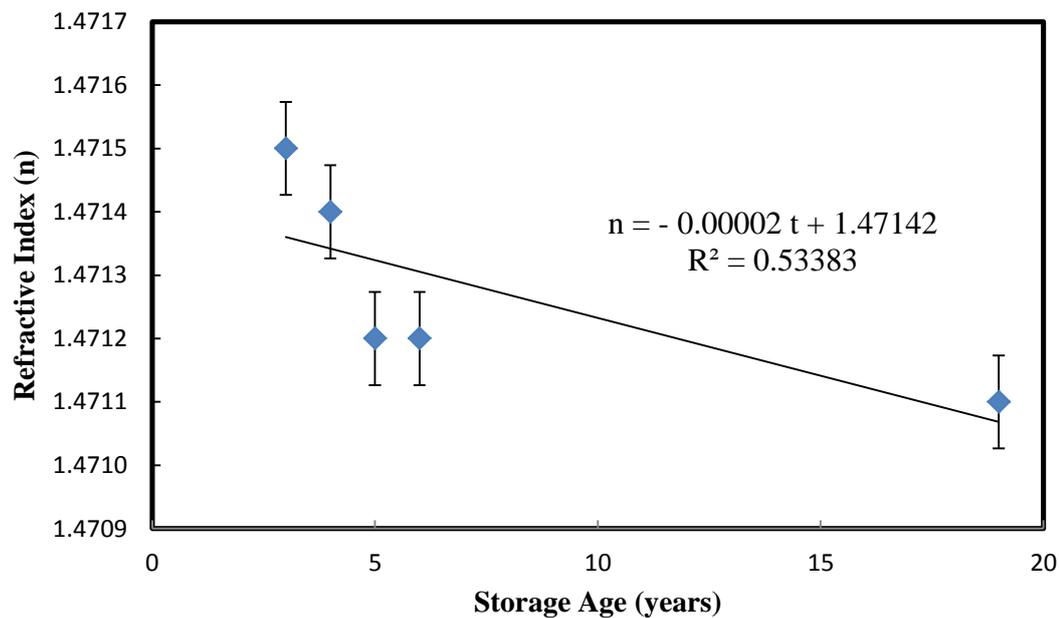


Fig. B.6: Measured refractive index versus storage age of olive oil samples of L<sub>7</sub>

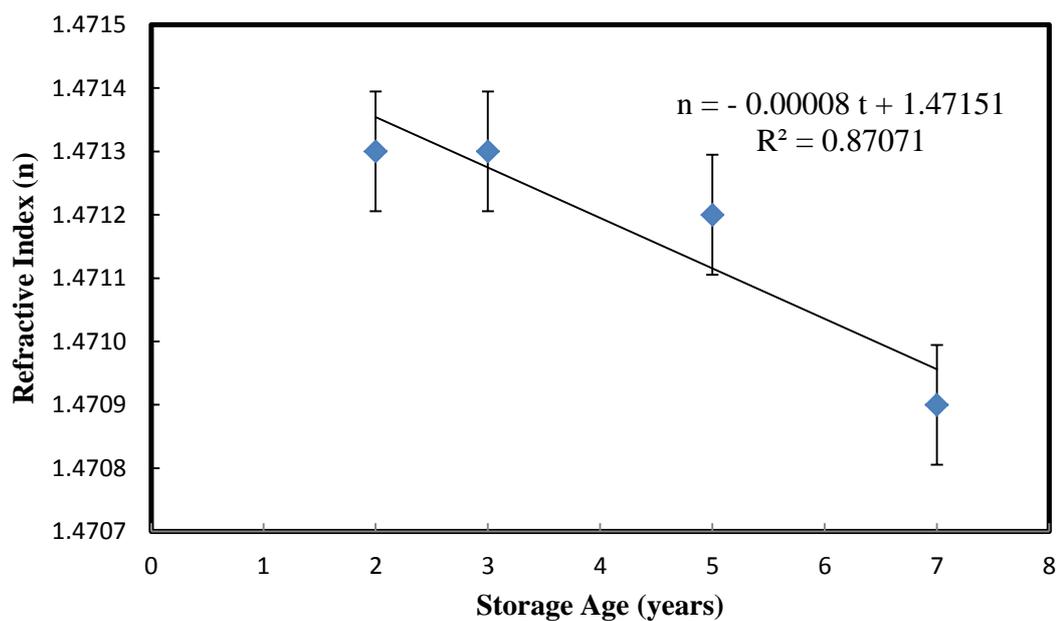


Fig. B.7: Measured refractive index versus storage age of olive oil samples of L<sub>9</sub>

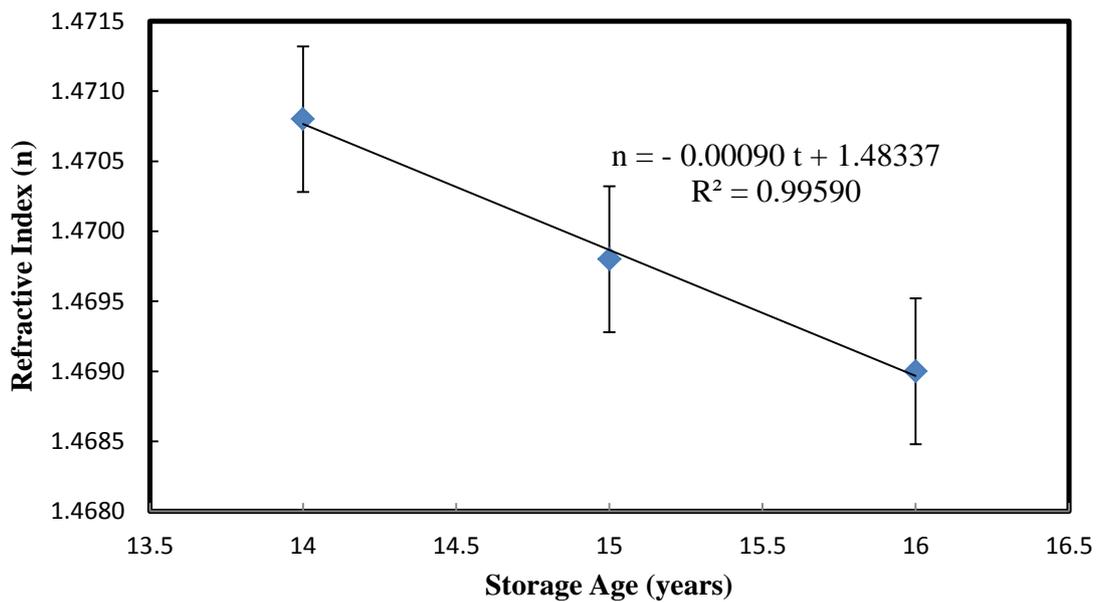
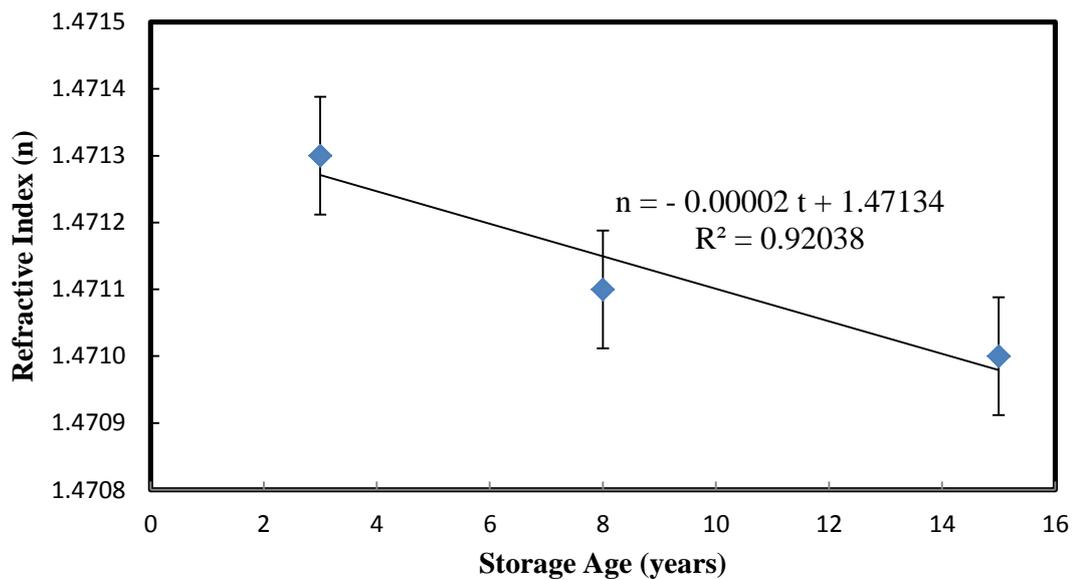
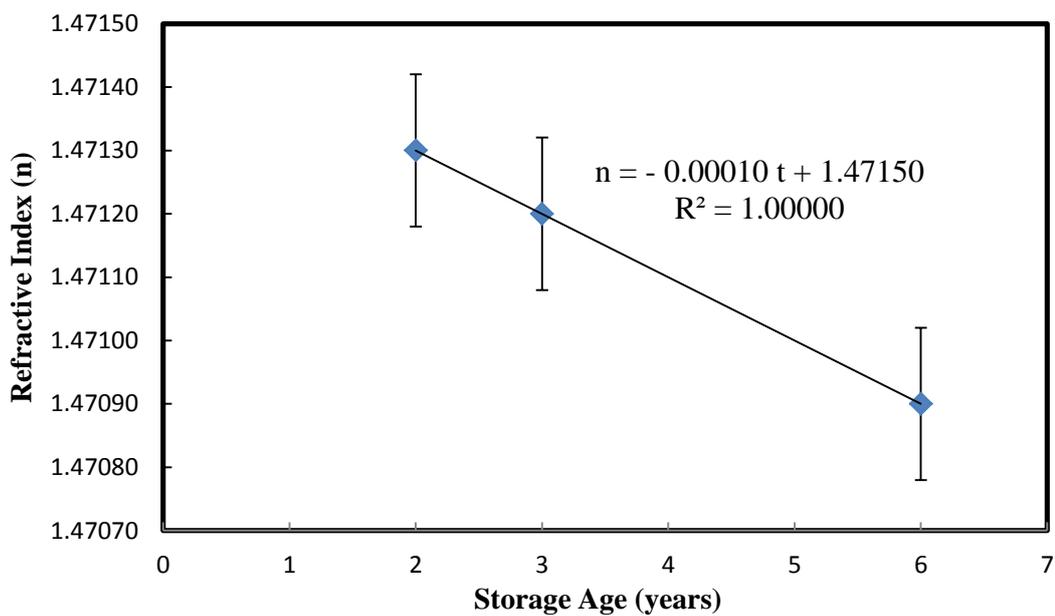


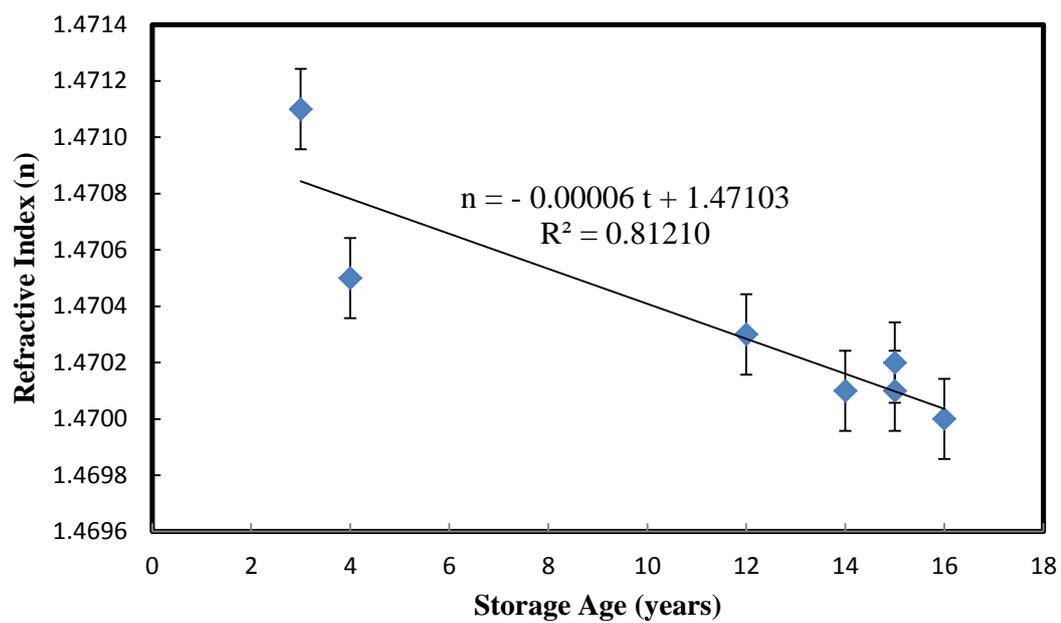
Fig. B.8: Measured refractive index versus storage age of olive oil samples of L<sub>10</sub>



**Fig. B.9:** Measured refractive index versus storage age of olive oil samples of L<sub>11</sub>



**Fig. B.10:** Measured refractive index versus storage age of olive oil samples of L<sub>12</sub>



**Fig. B.11:** Measured refractive index versus storage age of olive oil samples of L<sub>13</sub>

## Appendix C

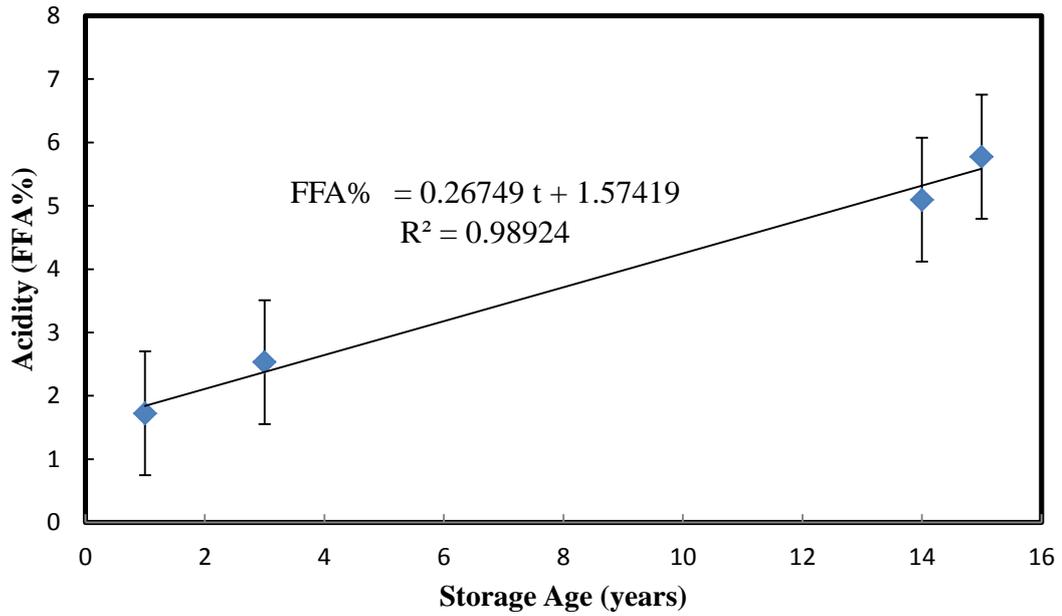
### Acidity Results

The Acidity of the olive oils samples for all regions and for different storage ages are given in Table (C.1).

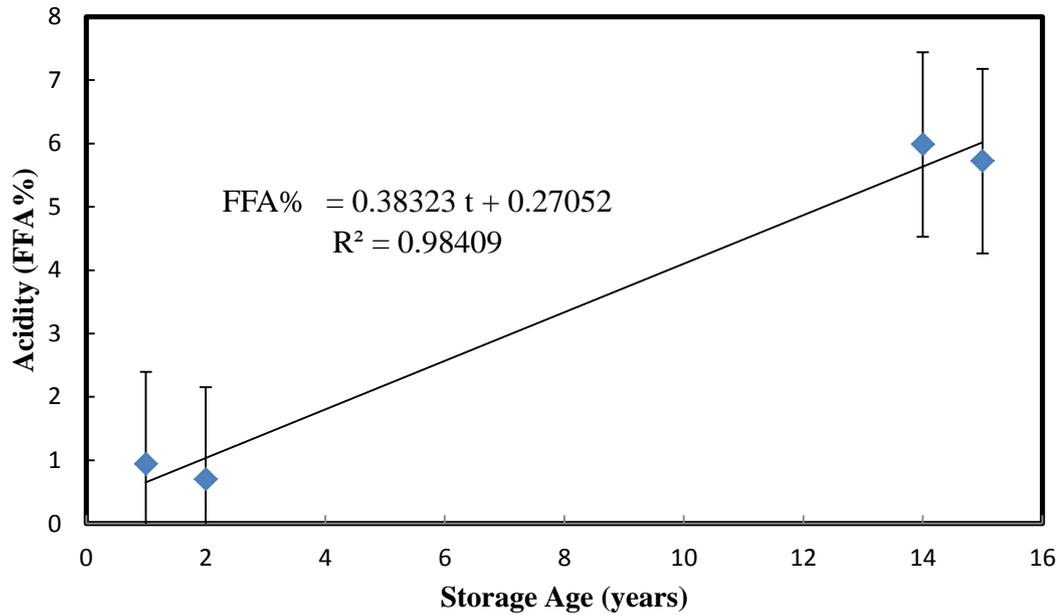
**Table (C.1): Measured Acidity of olive oil samples in different regions and for different storage ages**

Storage Age (years)	L <sub>2</sub>	L <sub>3</sub>	L <sub>4</sub>	L <sub>5</sub>	L <sub>6</sub>	L <sub>7</sub>	L <sub>9</sub>	L <sub>10</sub>	L <sub>11</sub>	L <sub>12</sub>	L <sub>13</sub>
1	1.72	0.94	0.51	0.45							
2		0.70	0.53				0.67			0.77	
3	2.53		0.56	1.49	1.03	0.80	0.96		0.96	1.88	0.56
4			1.96	2.07	1.80	1.12					1.03
5						1.56	1.18				
6				2.40		2.50				2.00	
7				2.88			1.20				
8				2.94					3.92		
12					3.18						2.31
13								4.40			
14	5.09	5.98						5.04			
15	5.77	5.72			3.81				7.07		3.95
16					4.49			9.49			4.60
19					5.02	4.00					5.22

The relationship between Acidity of olive oil samples and storage age is shown in figures (C.1 - C.11).



**Fig. C.1:** Measured acidity versus storage age of olive oil samples of L<sub>2</sub>



**Fig. C.2:** Measured acidity versus storage age of olive oil samples of L<sub>3</sub>

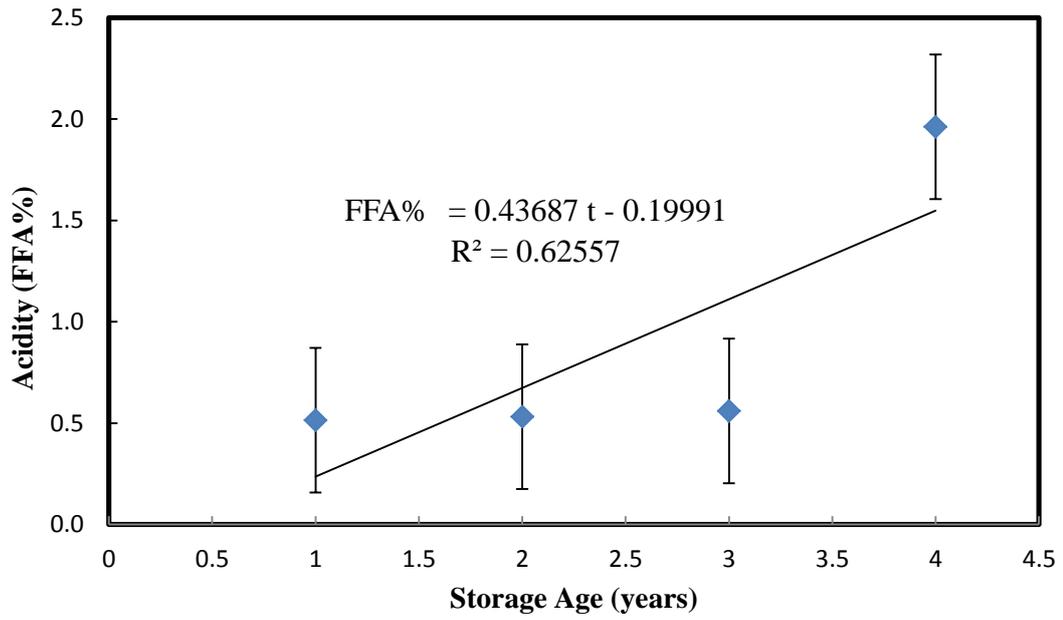


Fig. C.3: Measured acidity versus storage age of olive oil samples of L<sub>4</sub>

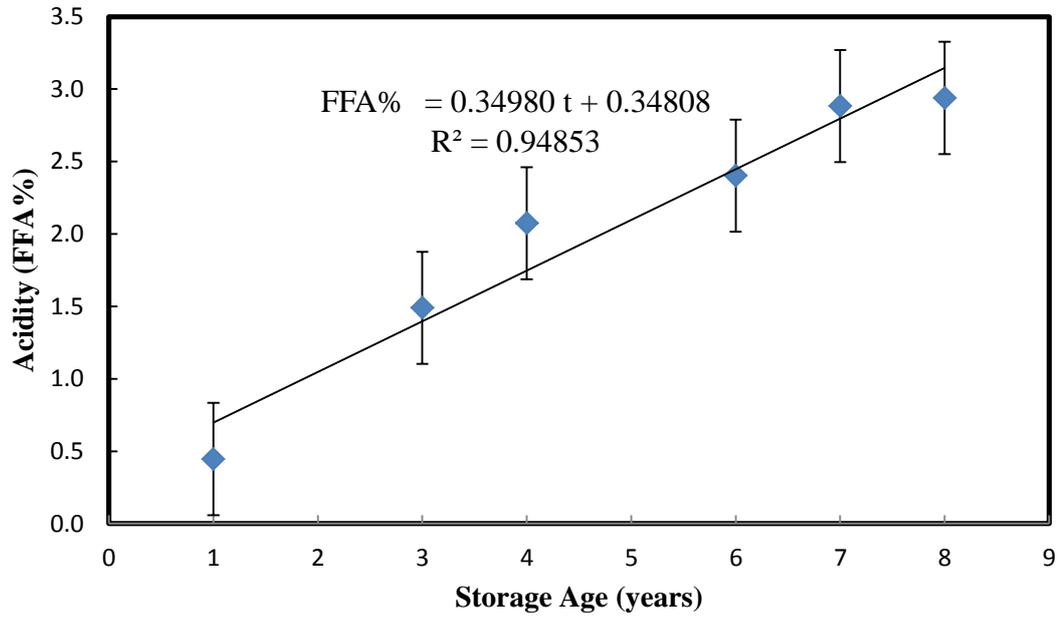
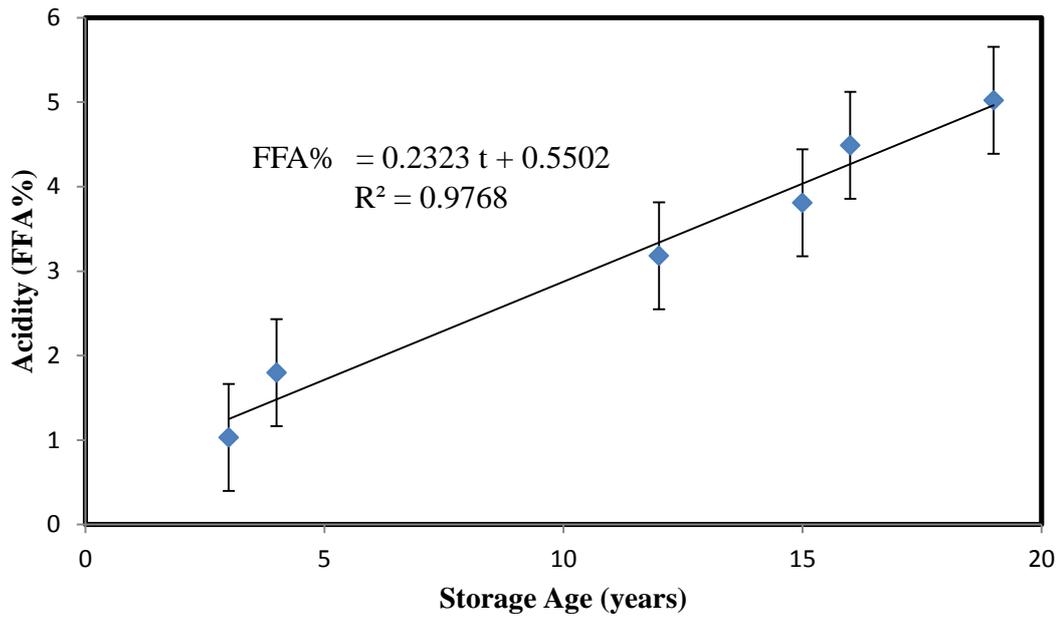
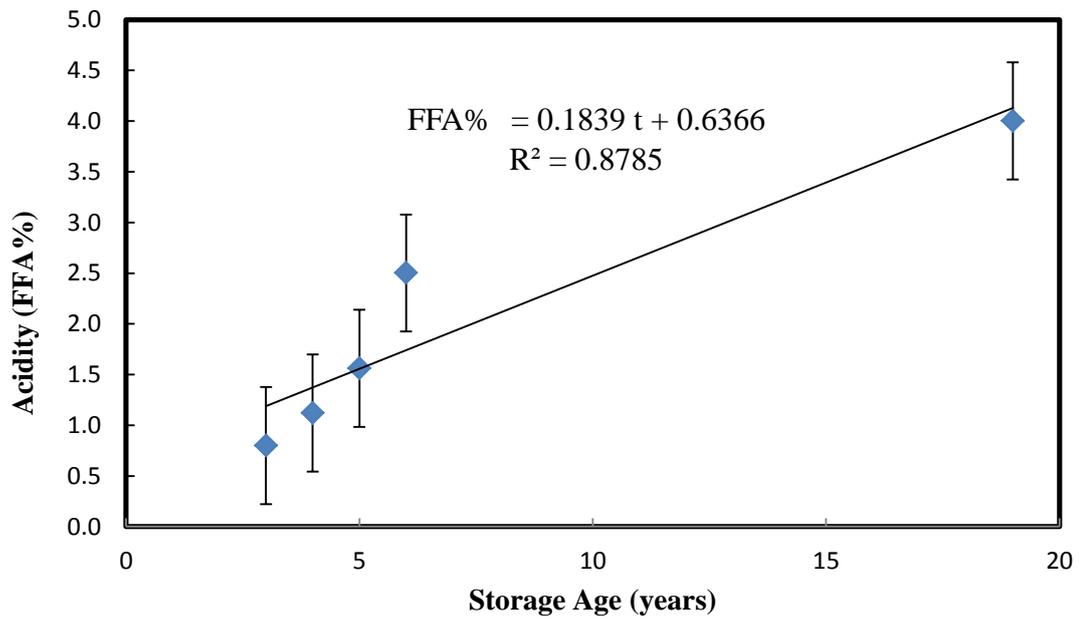


Fig. C.4: Measured acidity versus storage age of olive oil samples of L<sub>5</sub>



**Fig. C.5:** Measured acidity versus storage age of olive oil samples of L<sub>6</sub>



**Fig. C.6:** Measured acidity versus storage age of olive oil samples of L<sub>7</sub>

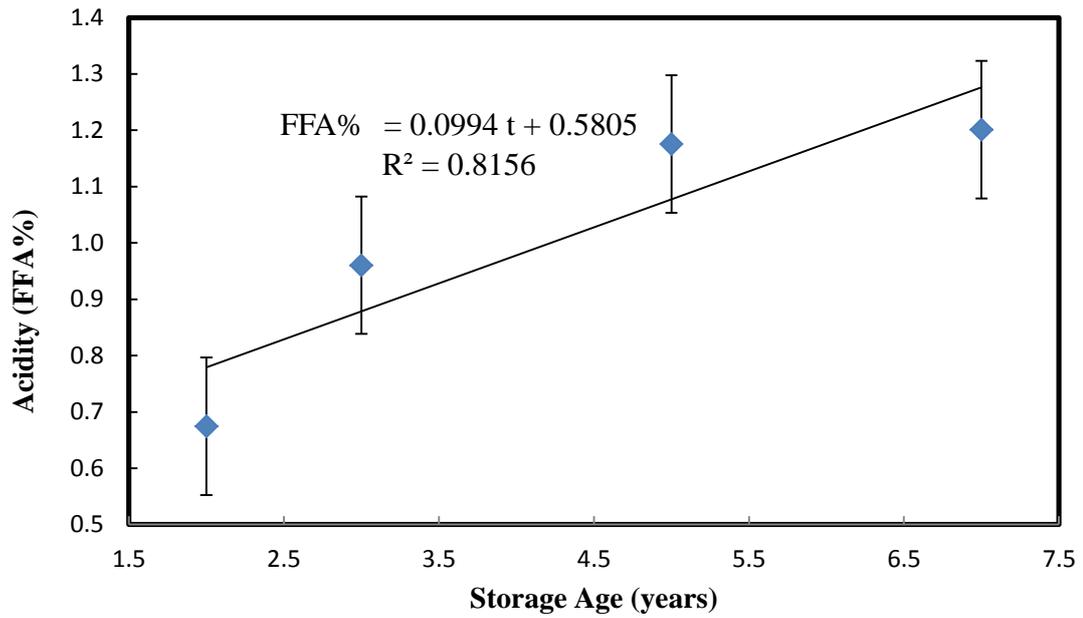


Fig. C.7: Measured acidity versus storage age of olive oil samples of L<sub>9</sub>

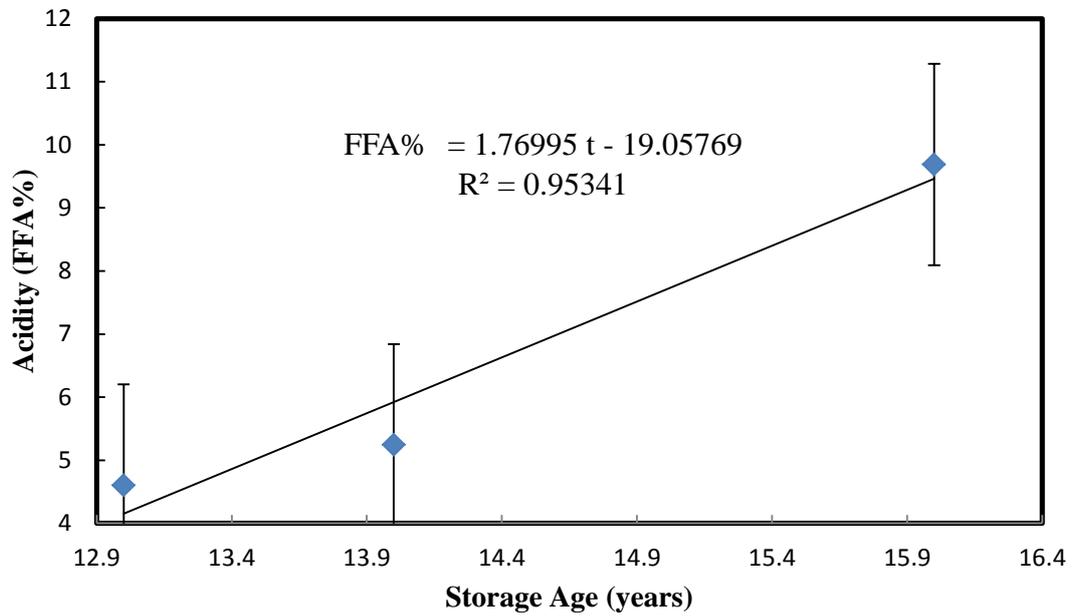
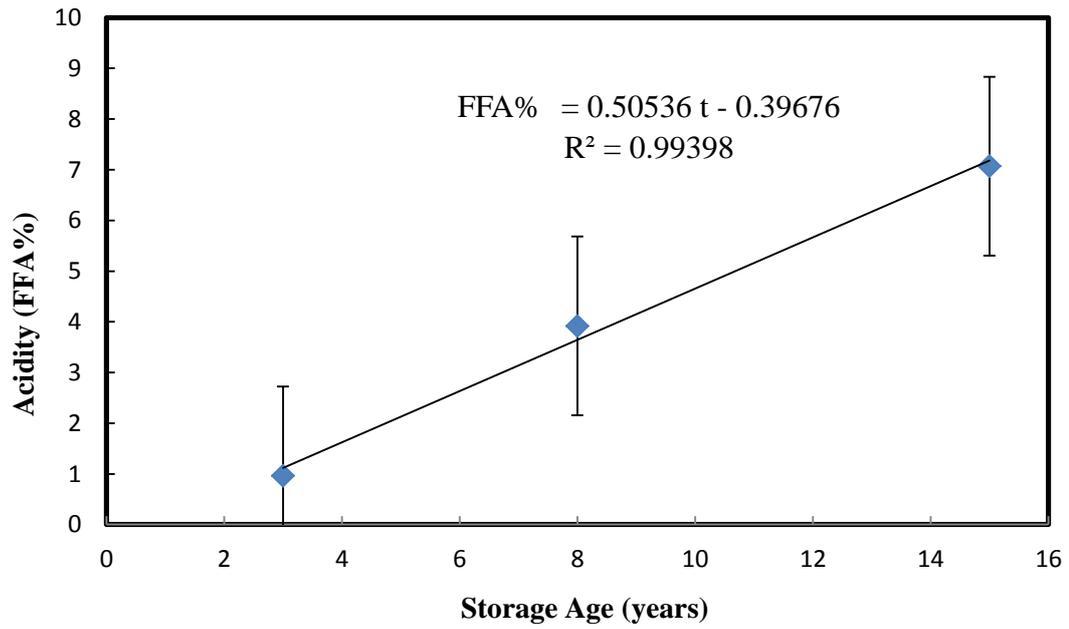
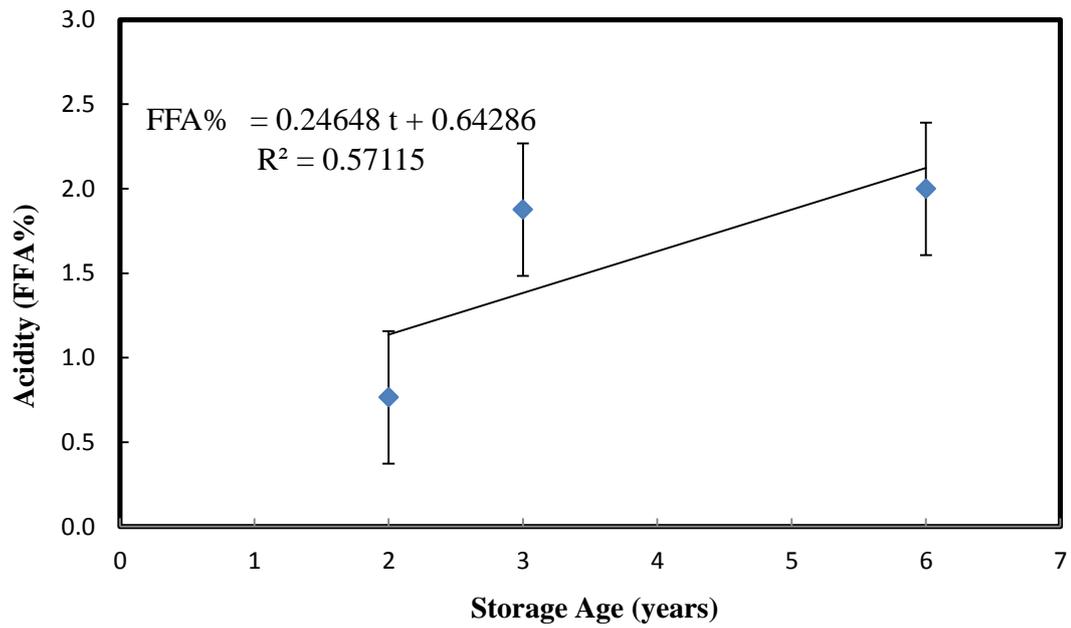


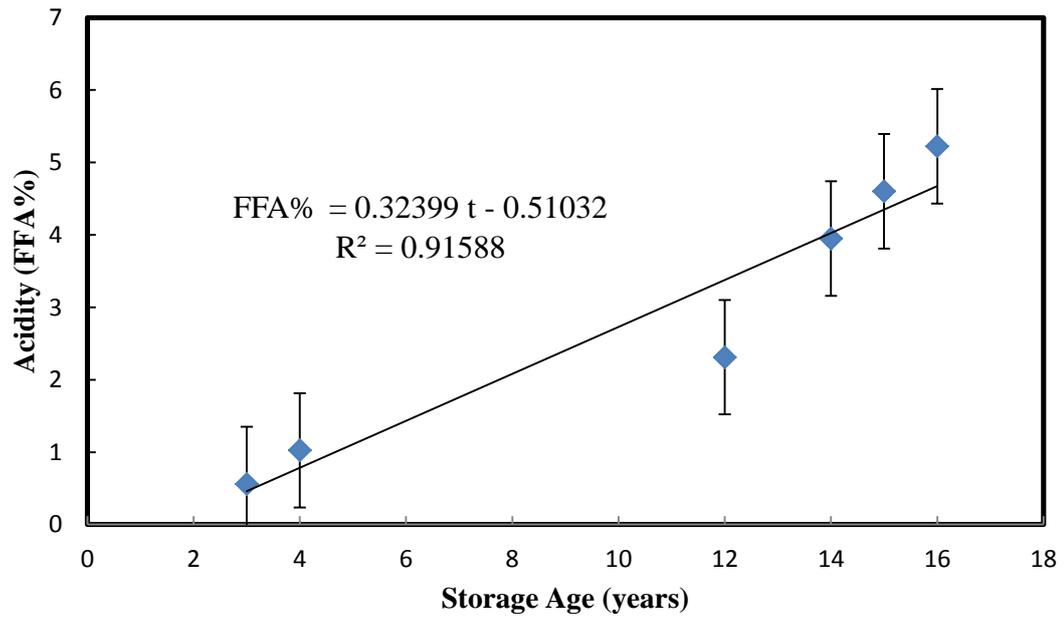
Fig. C.8: Measured acidity versus storage age of olive oil samples of L<sub>10</sub>



**Fig. C.9:** Measured acidity versus storage age of olive oil samples of L<sub>11</sub>



**Fig. C.10:** Measured acidity versus storage age of olive oil samples of L<sub>12</sub>

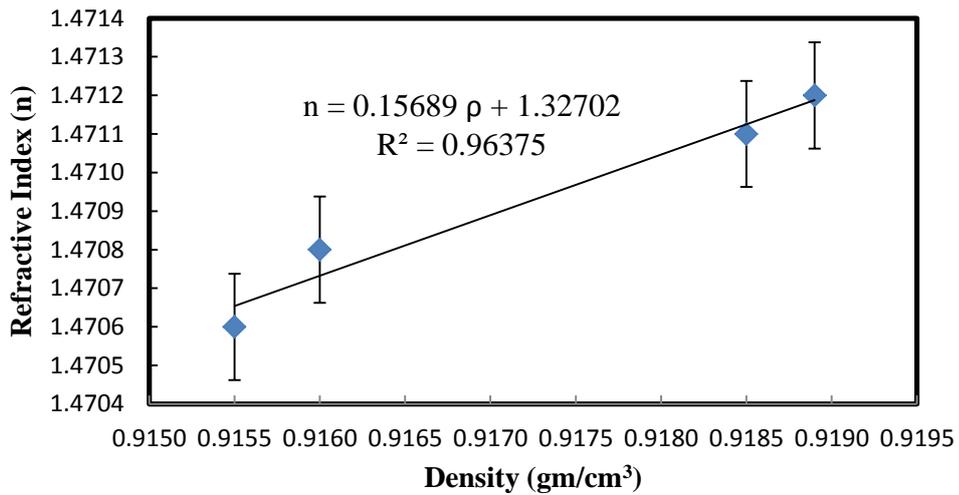


**Fig. C.11:** Measured acidity versus storage age of olive oil samples of L<sub>13</sub>

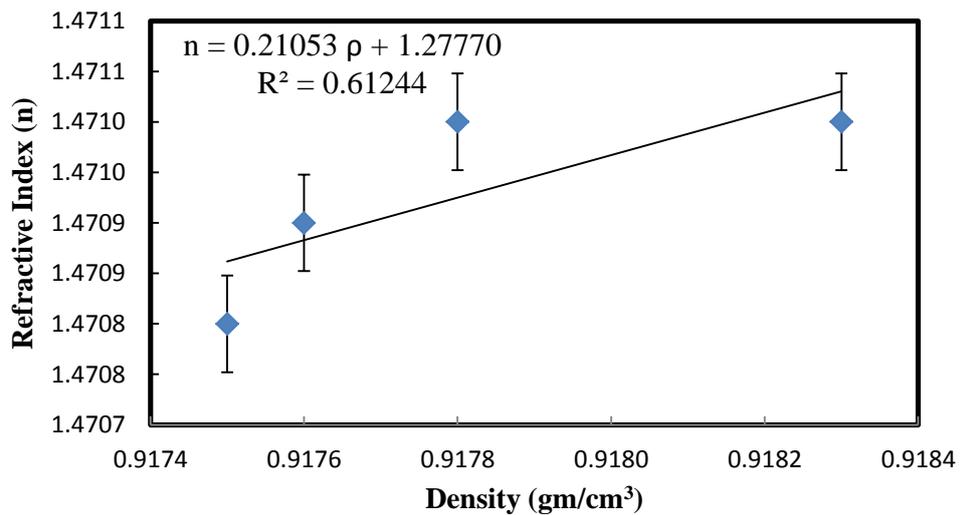
## Appendix D

### Refractive Index as a Function of Density

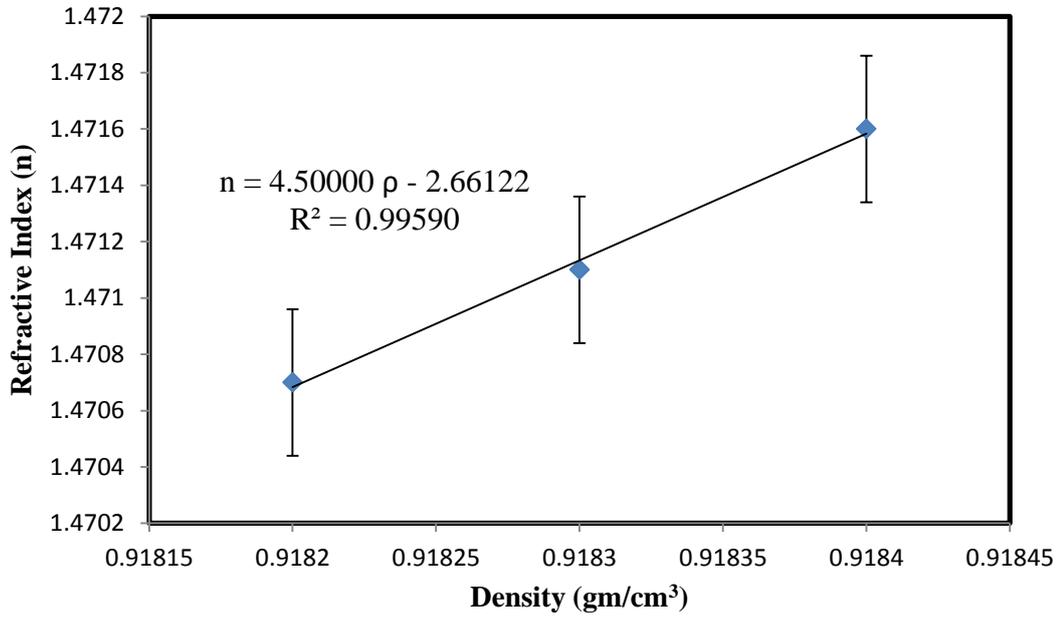
Figs. (D.1 – D.11) show the relationship between measured refractive index and density for all regions and all storage ages.



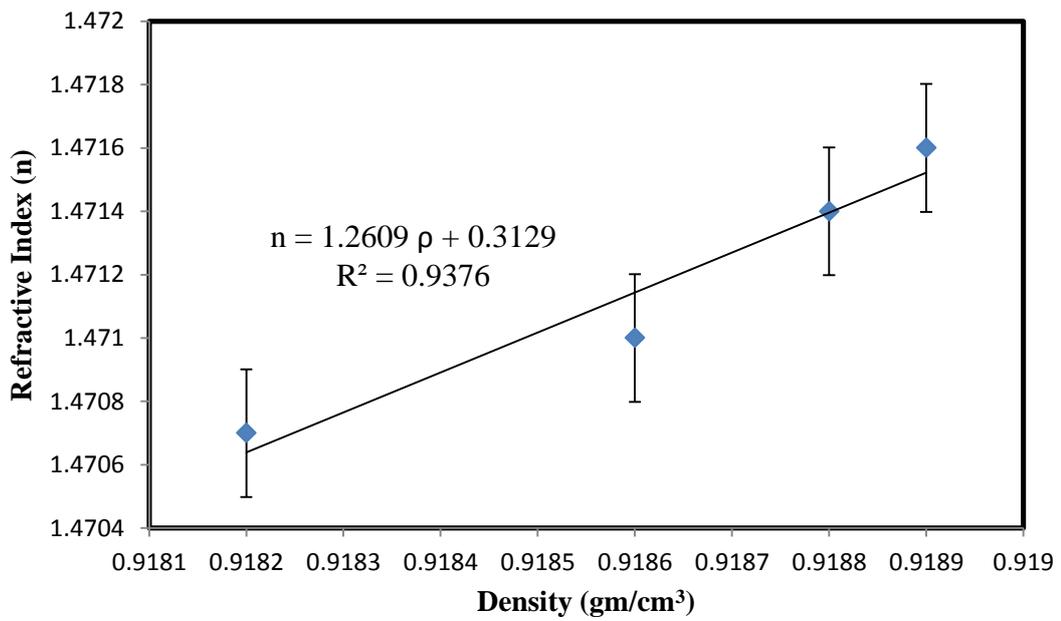
**Fig. D.1:** Measured refractive index versus density of olive oil samples of L<sub>2</sub>



**Fig. D.2:** Measured refractive index versus density of olive oil samples of L<sub>3</sub>



**Fig. D.3:** Measured refractive index versus density of olive oil samples of L<sub>4</sub>



**Fig. D.4:** Measured refractive index versus density of olive oil samples of L<sub>5</sub>

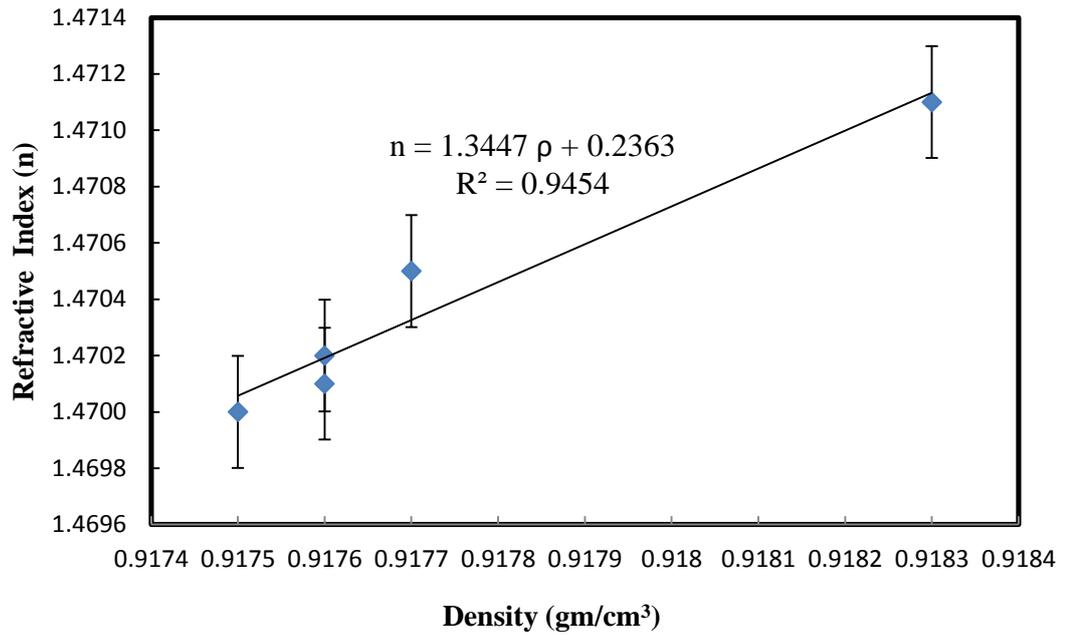


Fig. D.5: Measured refractive index versus density of olive oil samples of L<sub>6</sub>

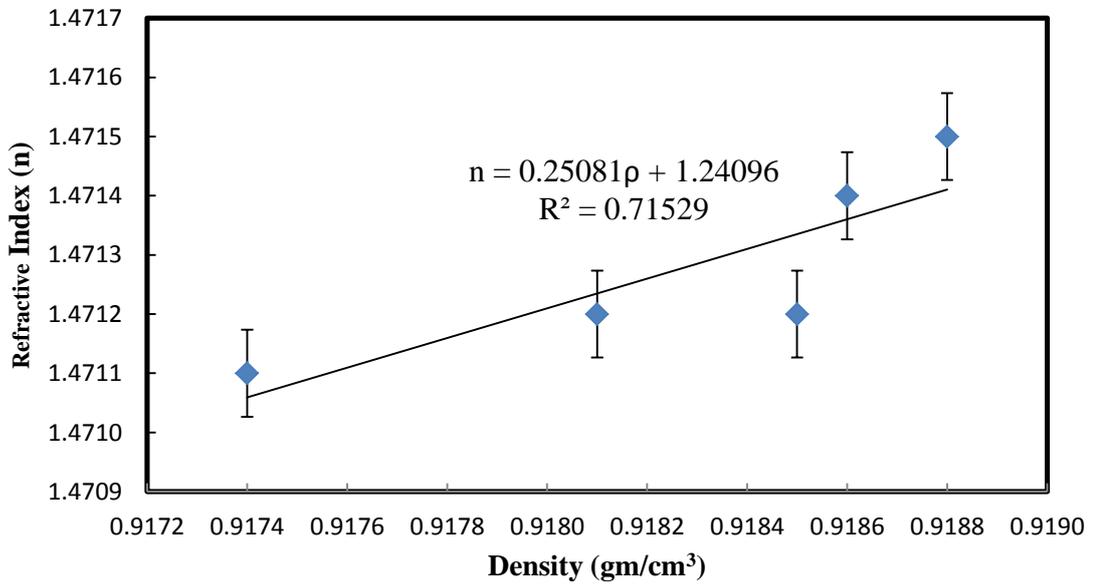
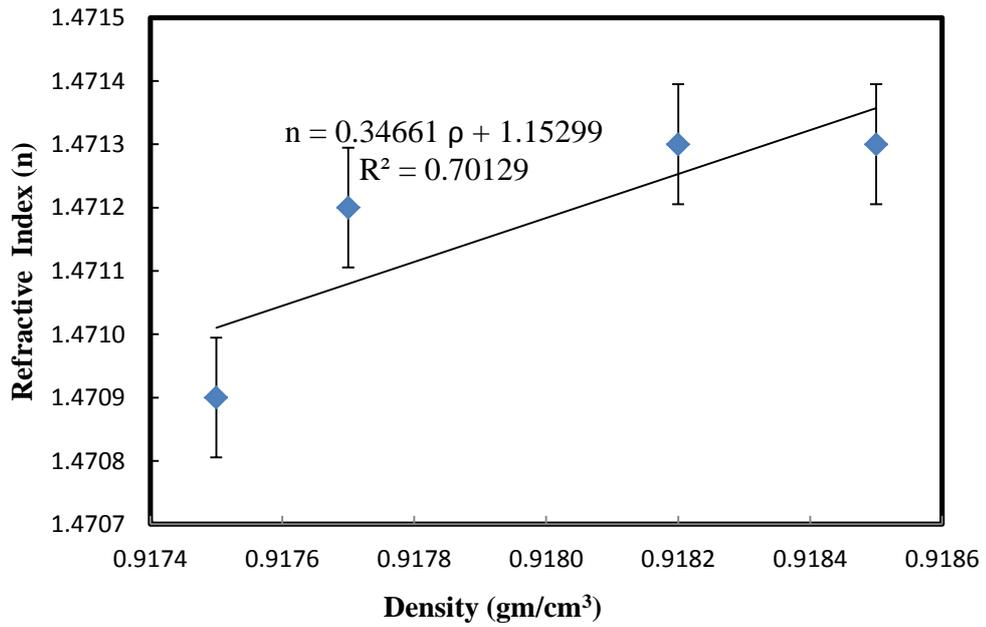
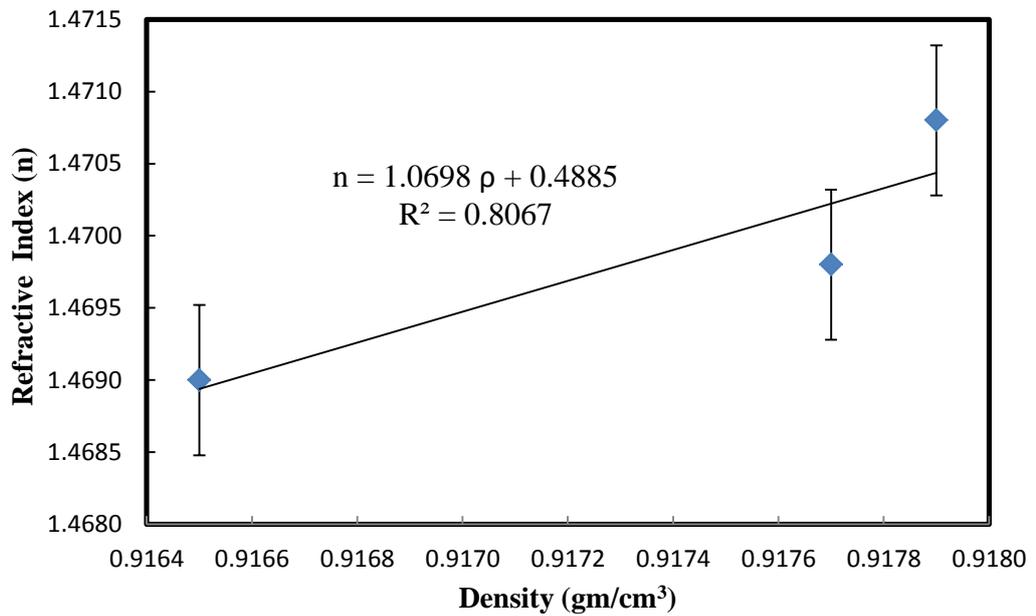


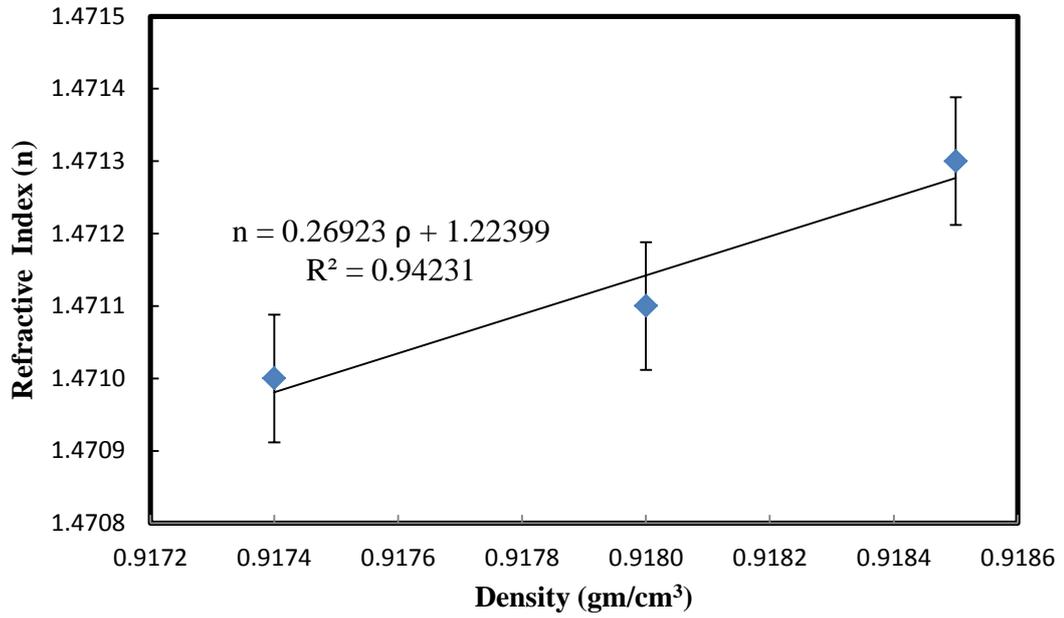
Fig. D.6: Measured refractive index versus density of olive oil samples of L<sub>7</sub>



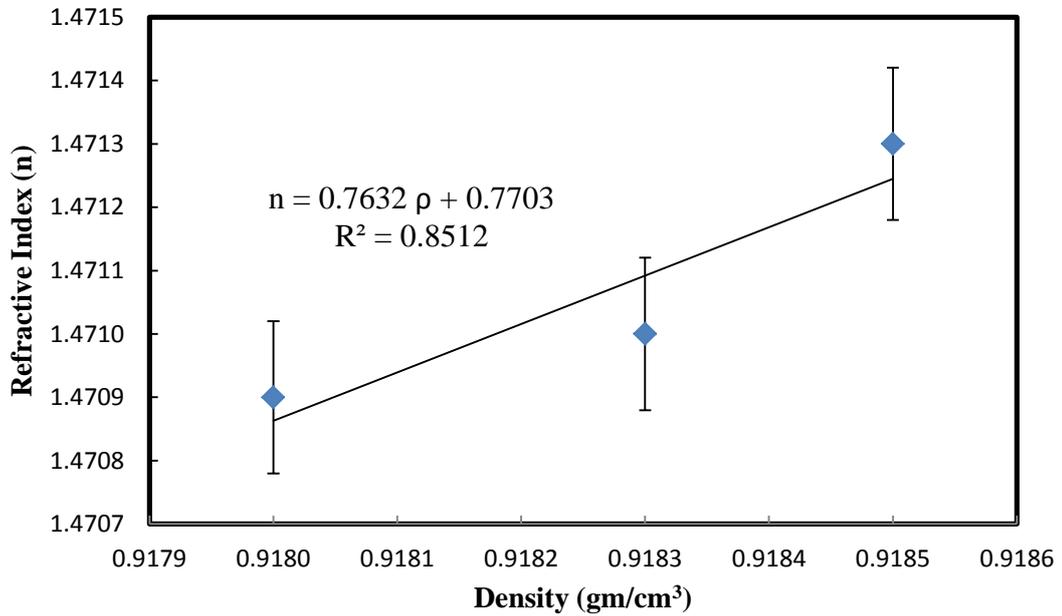
**Fig. D.7:** Measured refractive index versus density of olive oil samples of L<sub>9</sub>



**Fig. D.8:** Measured refractive index versus density of olive oil samples of L<sub>10</sub>



**Fig. D.9:** Measured refractive index versus density of olive oil samples of L<sub>11</sub>

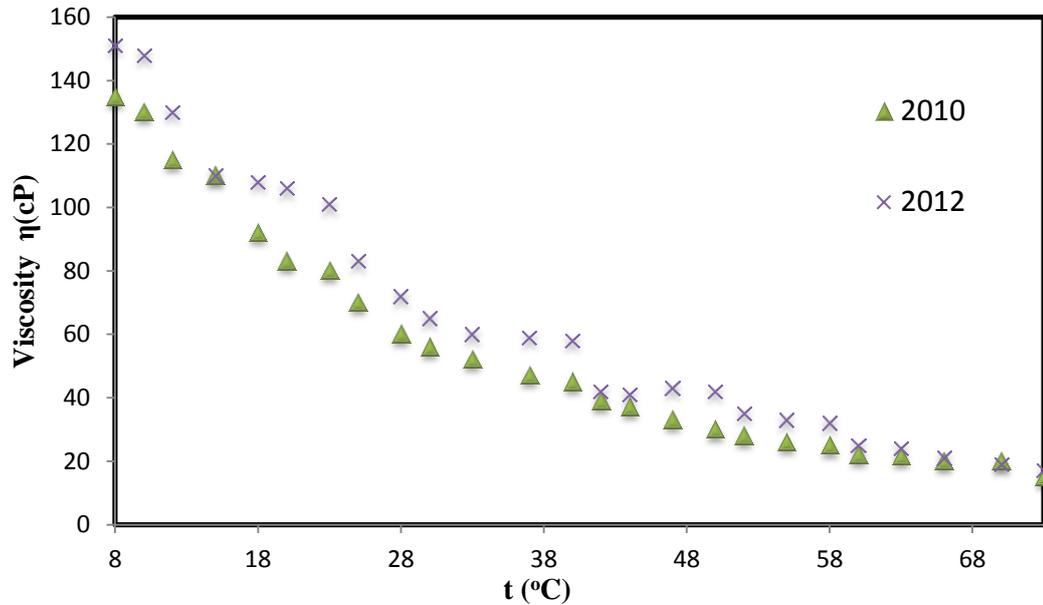


**Fig. D.10:** Measured refractive index versus density of olive oil samples of L<sub>12</sub>

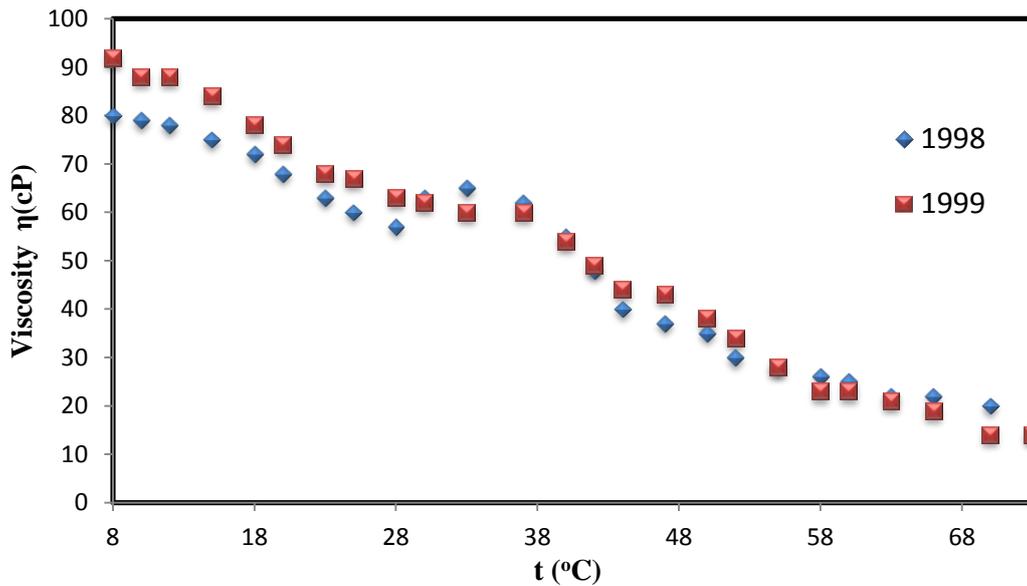
## Appendix E

### Viscosity Results

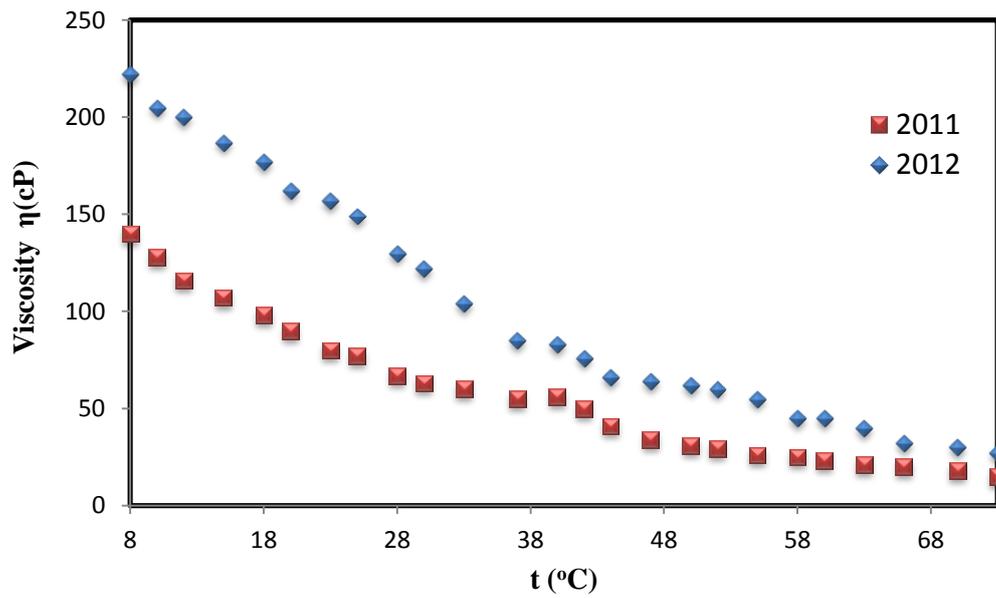
Figs. (E.1 - E.22) show the relationship between measured viscosity and temperature for all regions and all storage ages.



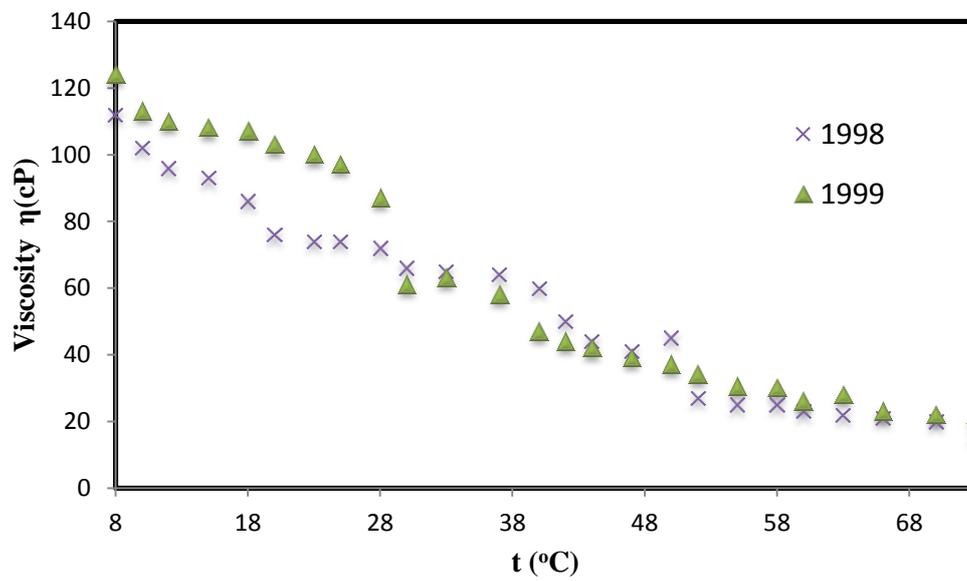
**Fig. E.1:** Measured viscosity versus temperature of olive oil samples of  $L_2$  (1 year and 3 years storage age)



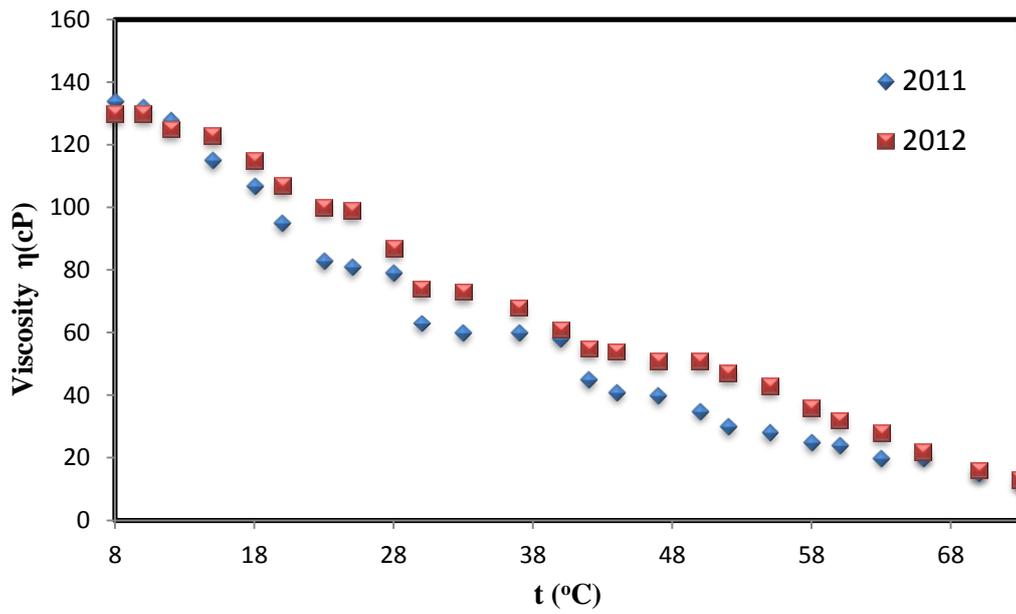
**Fig. E.2:** Measured viscosity versus temperature of olive oil samples of  $L_2$  (14 years and 15 years storage age)



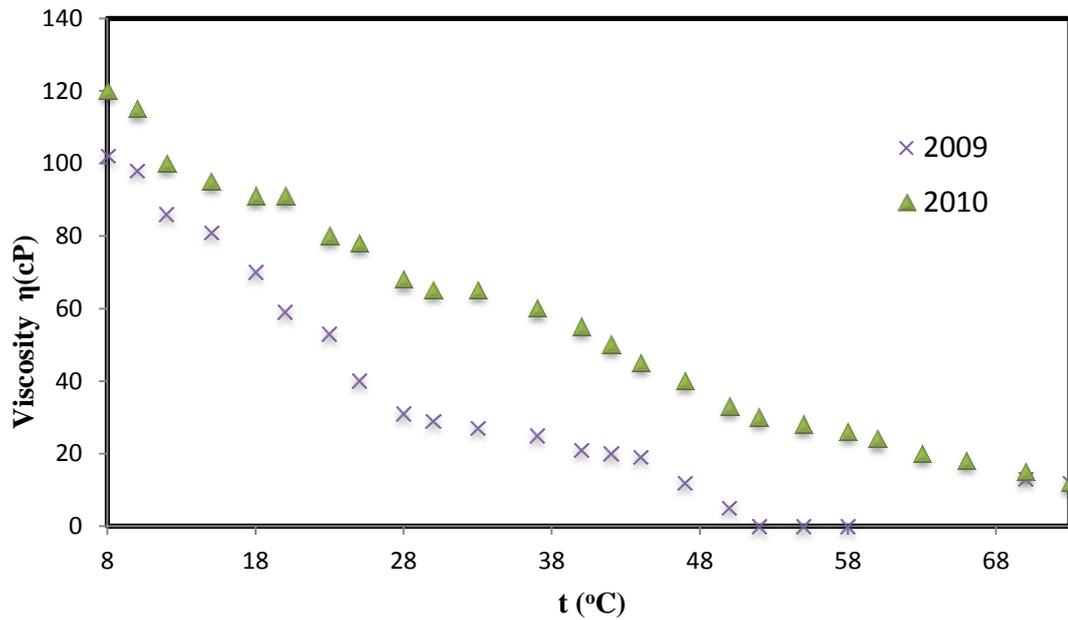
**Fig. E.3:** Measured viscosity versus temperature of olive oil samples of  $L_3$  (1 year and 2 years storage age)



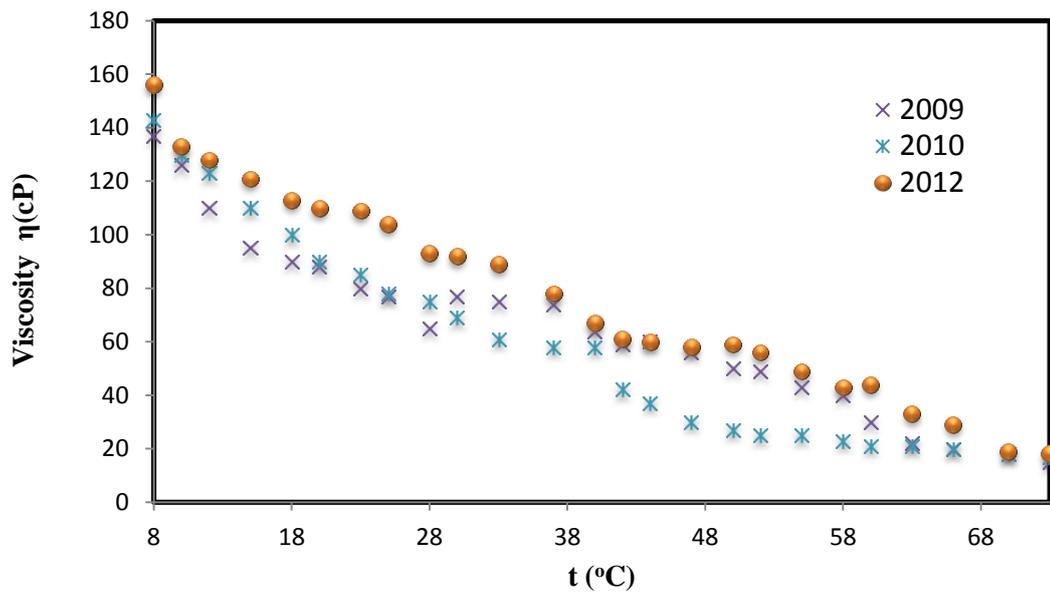
**Fig. E.4:** Measured viscosity versus temperature of olive oil samples of  $L_3$  (14 years and 15 years storage age)



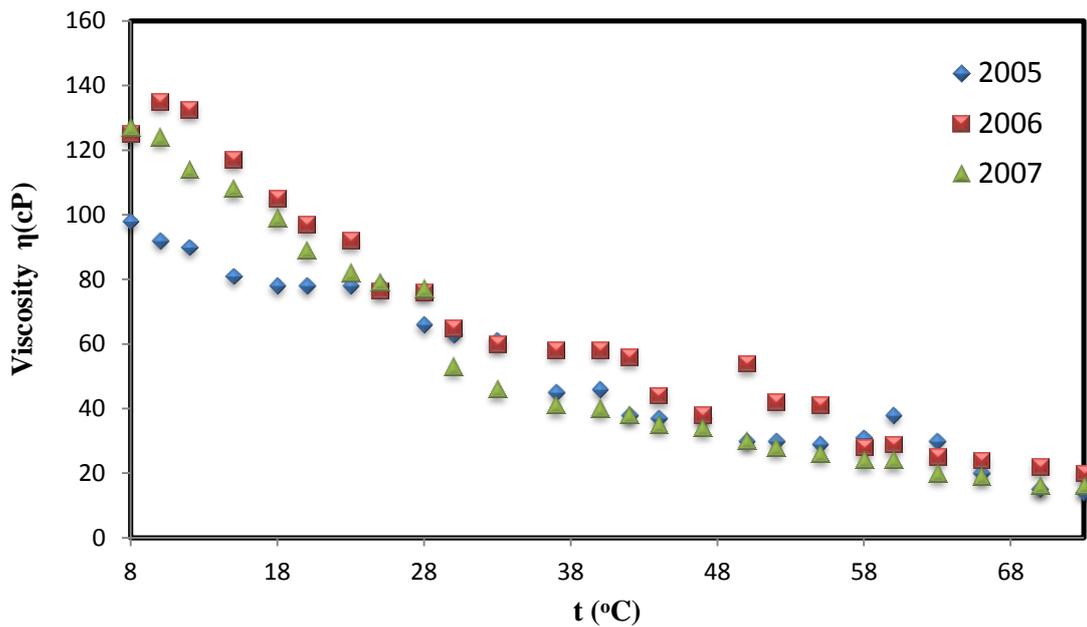
**Fig. E.5:** Measured viscosity versus temperature of olive oil samples of L<sub>4</sub> (1 year and 2 years storage age)



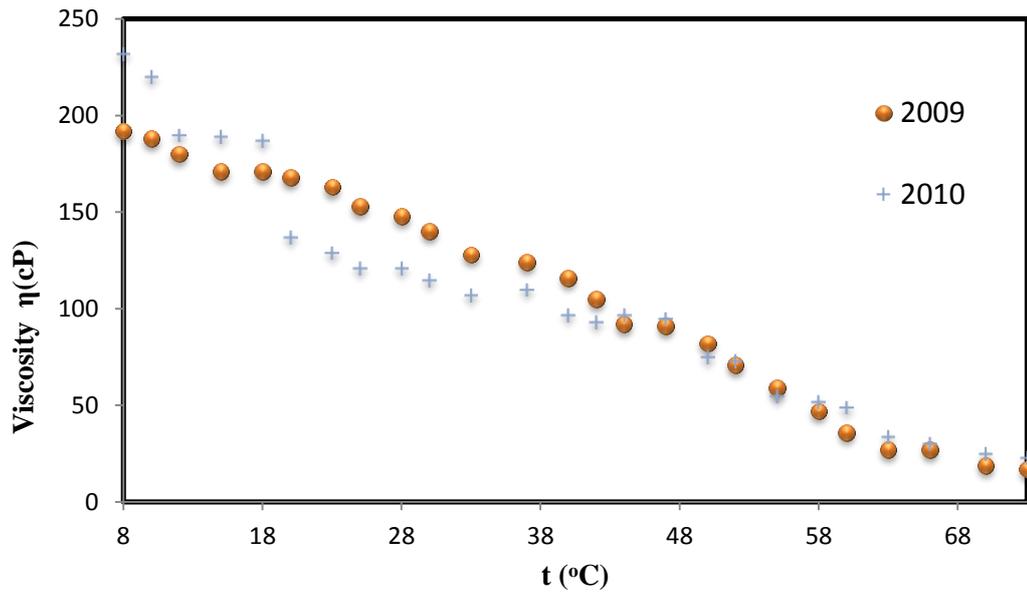
**Fig. E.6:** Measured viscosity versus temperature of olive oil samples of L<sub>4</sub> (3 years and 4 years storage age)



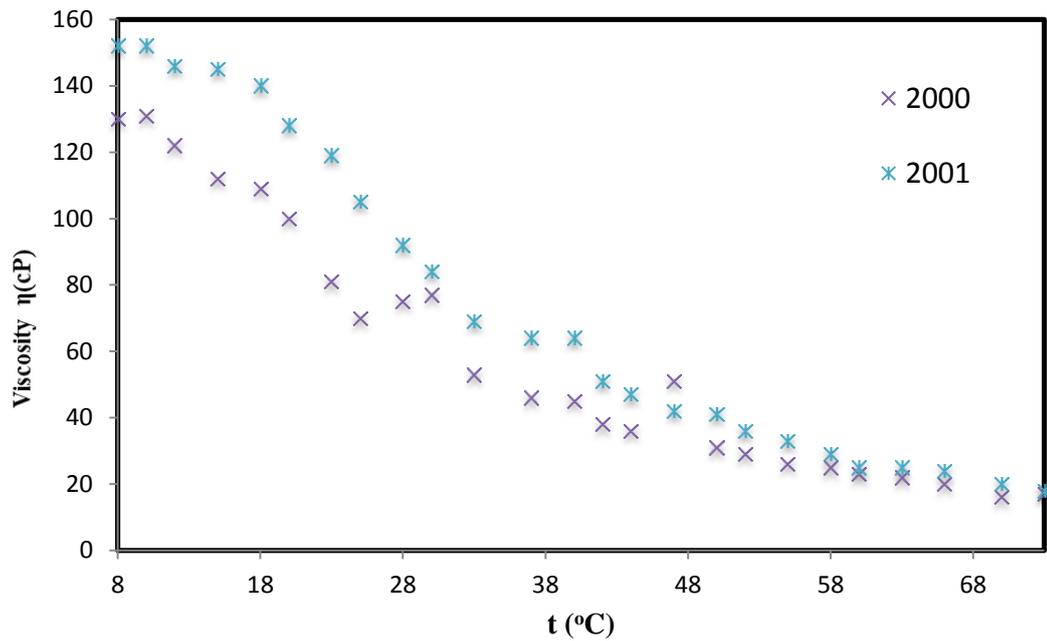
**Fig. E.7:** Measured viscosity versus temperature of olive oil samples of  $L_5$  (1 year, 2 years and 3 years storage age)



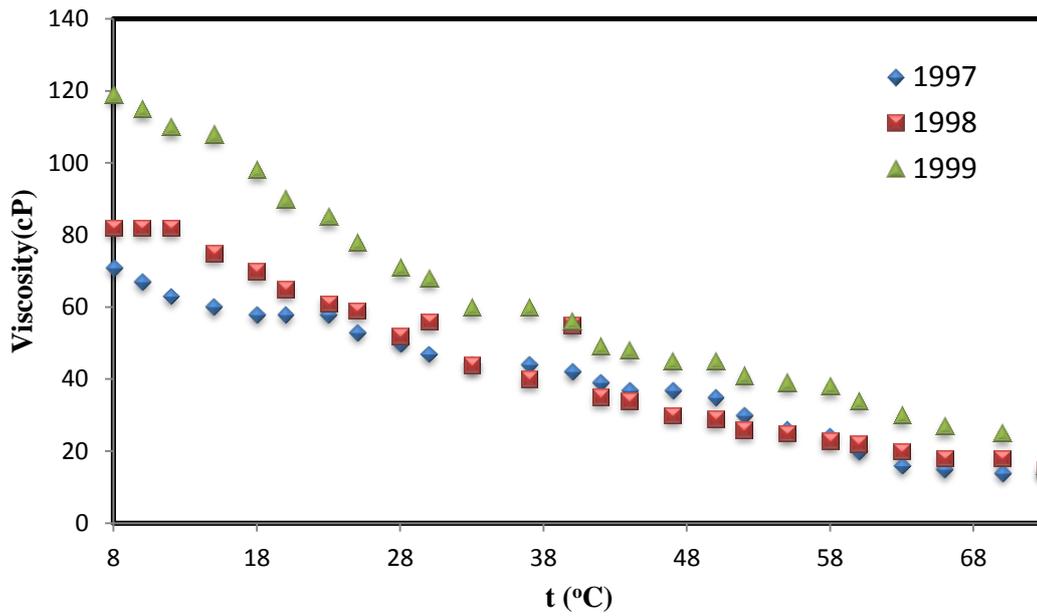
**Fig. E.8:** Measured viscosity versus temperature of olive oil samples of  $L_5$  (6 years, 7 years and 8 years storage age)



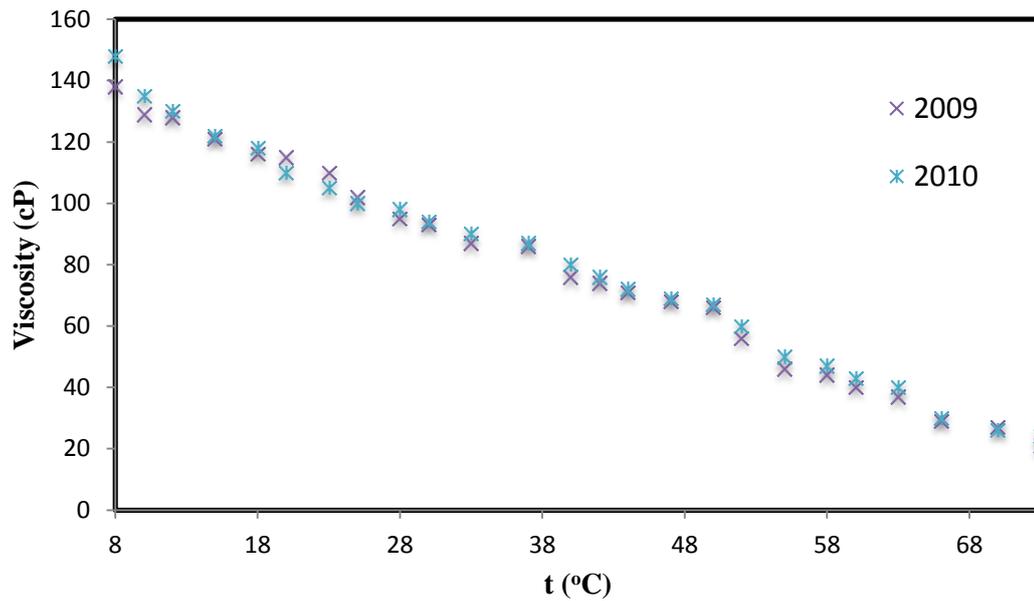
**Fig. E.9:** Measured viscosity versus temperature of olive oil samples of  $L_6$  (3 years and 4 years storage age)



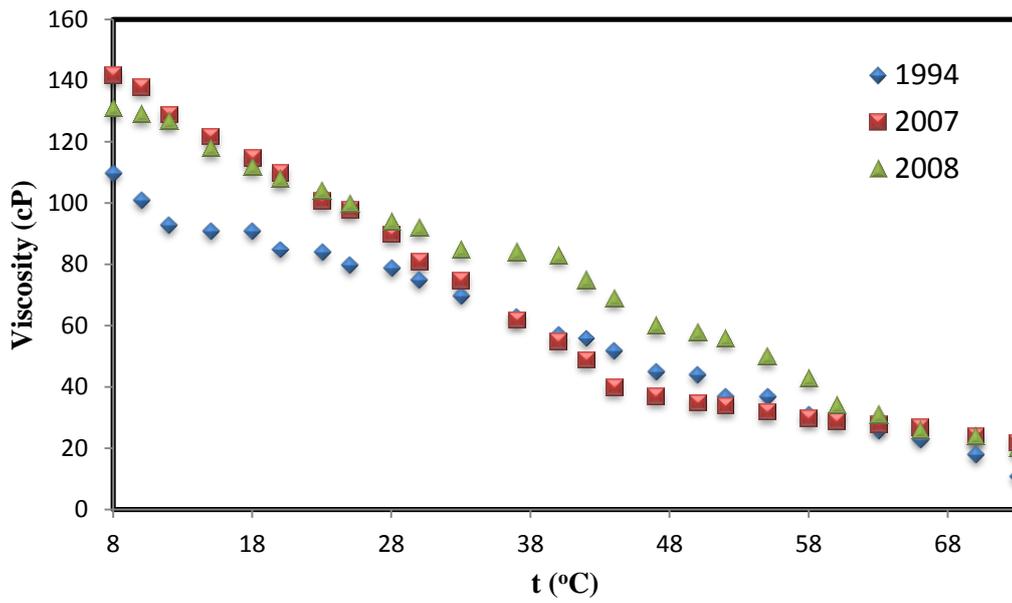
**Fig. E.10:** Measured viscosity versus temperature of olive oil samples of  $L_6$  (12 years and 13 years storage age)



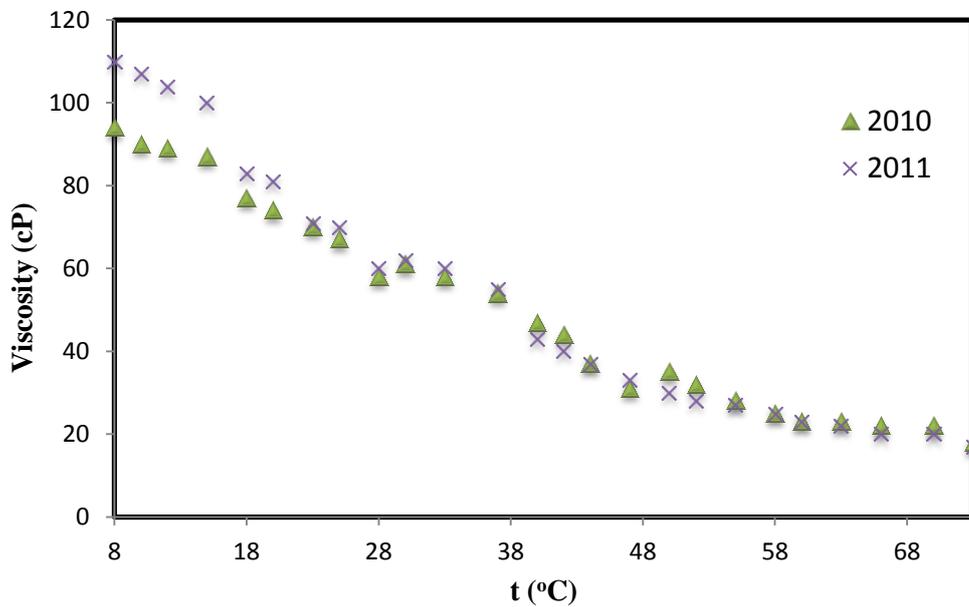
**Fig. E.11:** Measured viscosity versus temperature of olive oil samples of L<sub>6</sub> (14 years, 15 years and 16 years storage age)



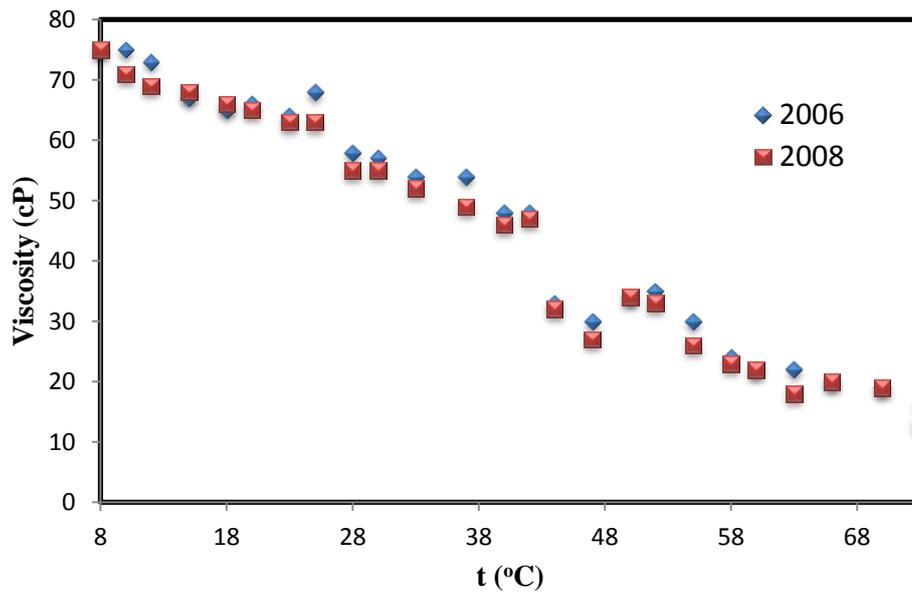
**Fig. E.12:** Measured viscosity versus temperature of olive oil samples of L<sub>7</sub> (3 years and 4 years storage age)



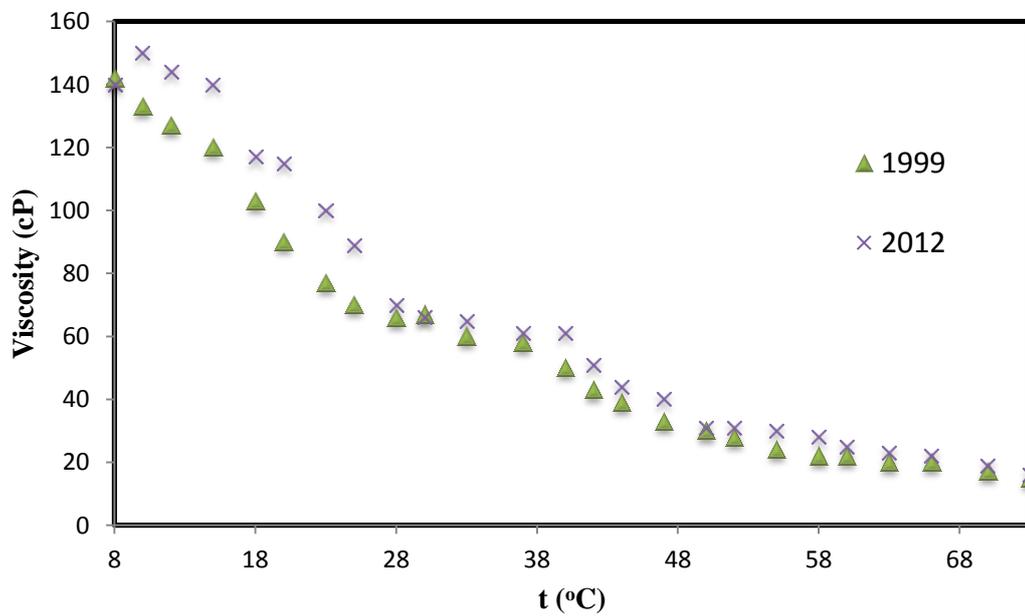
**Fig. E.13:** Measured viscosity versus temperature of olive oil samples of L<sub>7</sub> (5 years, 6 years and 19 years storage age)



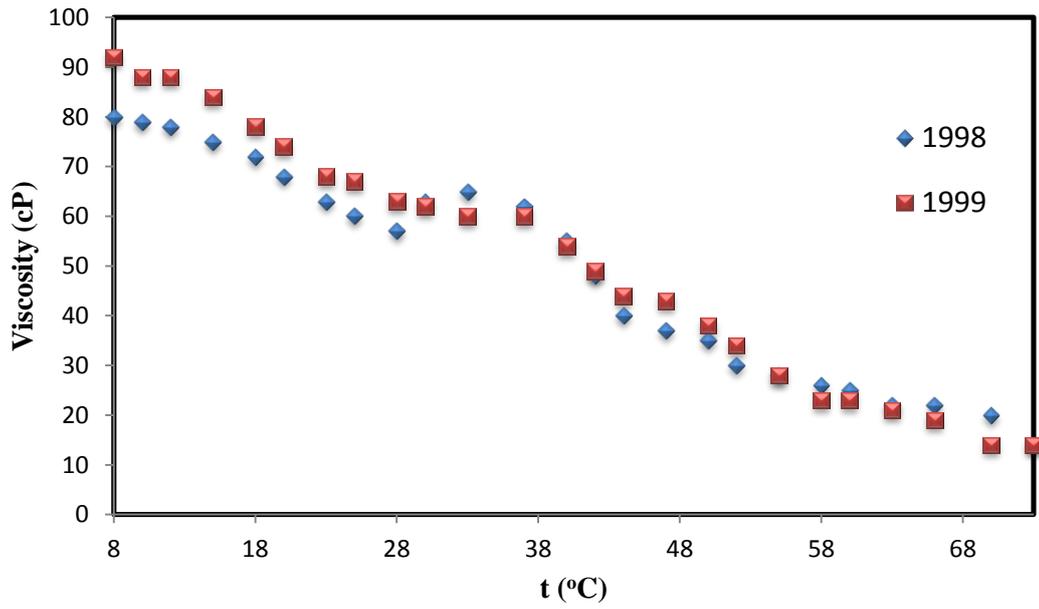
**Fig. E.14:** Measured viscosity versus temperature of olive oil samples of L<sub>9</sub> (2 years and 3 years storage age)



**Fig. E.15:** Measured viscosity versus temperature of olive oil samples of L<sub>9</sub> (5 years and 7 years storage age)



**Fig. E.16:** Measured viscosity versus temperature of olive oil samples of L<sub>10</sub> (1 year and 14 years storage age)



**Fig. E.17:** Measured viscosity versus temperature of olive oil samples of L<sub>10</sub> (14 years and 15 years storage age)

جامعة النجاح الوطنية  
كلية الدراسات العليا

# الخصائص الديناميكية الفيزيائية لزيت الزيتون في فلسطين

إعداد

أحمد مصطفى بهتي

إشراف

أ. د. عصام راشد عبد الرازق

د. شريف محمد مسامح

قدمت هذه الأطروحة استكمالاً لمتطلبات الحصول على درجة الماجستير في الفيزياء بكلية  
الدراسات العليا في جامعة النجاح الوطنية في نابلس، فلسطين.

2014م

ب

## الخصائص الديناميكية الفيزيائية لزيت الزيتون في فلسطين

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### الملخص

تم دراسة عينات من زيت الزيتون في فلسطين ذات أعمار تخزين مختلفة ومن مناطق مختلفة. حيث تم قياس الكثافة ومعامل الانكسار و الحموضة و اللزوجة للعينات. تم دراسة معامل الانكسار لعينات زيت الزيتون كعلاقة مع عمر التخزين و أظهرت النتائج أن معامل الانكسار يقل بزيادة عمر التخزين. أظهرت الدراسة أن حموضة عينات زيت الزيتون من مختلف المحاصيل وفي مناطق مختلفة تزداد بزيادة عمر التخزين. أظهرت النتائج ان معظم عينات زيت الزيتون (عمر التخزين  $\geq 12$  عاما ) لم تتجاوز الحموضة لها معايير الجودة الدولية (  $> 3.3\%$  ). تجدر الإشارة هنا إلى أن زيت الزيتون يمكن تخزينه حتى 12 عاما دون ان تتجاوز الحموضة معايير الجودة العالمية في الظروف المناسبة.

وقد تم دراسة اللزوجة لعينات زيت الزيتون من محصول عام 2012 من مناطق مختلفة، و أظهرت النتائج أن تصنيف معظم عينات زيت الزيتون هو زيت الزيتون البكر. تم اقتراح معادلات تتضمن ثابتين واخرى تتضمن ثلاث ثوابت للحصول على افضل علاقة مناسبة لوصف تغير اللزوجة الديناميكية لزيت الزيتون مع درجة الحرارة. تمت مقارنة النتائج التجريبية للزوجة مع معادلة قانون القوة الخاصة باللزوجة، وقد تبين أن سلوك جميع عينات الزيت هو سلوك نيوتوني.