

### **FOURIER TRANSFORM INFRARED (FTIR) SPECTROSCOPY AND PHYSICOCHEMICAL ANALYSIS FOR AUTHENTICATION OF OLIVE OIL ADULTERATED WITH SUNFLOWER OIL**

Naquiah Ahmad Nizar<sup>i</sup>, Siti Aimi Sarah Zainal Abidin<sup>ii</sup>, Amirah Nur Syahirah Abd Ghafur<sup>iii</sup>,  
Aishah Bujang<sup>iv</sup> & Nina Fadhilah Jailani<sup>v</sup>

<sup>i</sup> Malaysia Institute of Transport (MITRANS), Universiti Teknologi MARA, Shah Alam. naquiah@uitm.edu.my

<sup>ii</sup> (*Corresponding author*). Malaysia Institute of Transport (MITRANS), Universiti Teknologi MARA, Shah Alam. sitiaini@uitm.edu.my

<sup>iii</sup> Faculty of Applied Sciences, Universiti Teknologi MARA, Shah Alam. amirahnsyahirah@yahoo.com

<sup>iv</sup> Faculty of Applied Sciences, Universiti Teknologi MARA, Shah Alam. aisah012@uitm.edu.my

<sup>v</sup> Faculty of Applied Sciences, Universiti Teknologi MARA, Shah Alam. fadhi478@uitm.edu.my

#### **Abstract**

*Olive oil has been listed as the top most common food fraud by European Commission and therefore requires constant development of authentication method. This study is conducted to determine the presence of sunflower oil in olive oil at 1, 5, 10 and 20% of adulteration percentage. Fourier Transform Infrared (FTIR) spectroscopy and physicochemical analysis such as specific gravity, refractive index, iodine value, acid value and colour were used to distinguished adulterated samples. The spectral differences between olive oil added with sunflower oil by using FTIR can be seen at frequency region 1402  $\text{cm}^{-1}$  caused by =C-H bending, 1300 – 1000  $\text{cm}^{-1}$  caused by C-O stretching and CH<sub>2</sub>- bending, 965 – 960  $\text{cm}^{-1}$  caused by -HC=CH and 872 – 850  $\text{cm}^{-1}$  caused by =CH<sub>2</sub> wagging. Meanwhile, refractive index can detect differences only at 10% addition level. For the analysis specific gravity, iodine value and acid value were significantly difference ( $p < 0.05$ ) between olive oil and 5% addition of sunflower oil into olive oil which indicated that the 5% addition level can be detected. Colour analysis can determine significant differences at 1% addition. Ensuring olive oil at the highest quality is an importance value in halal supply chain.*

**Keywords:** *Olive Oil, Adulteration, Halal Supply Chain.*

#### **INTRODUCTION**

Olive oil (OO) is oil made from the fruit of *Olea europea* L. (Preedy & Watson, 2010). Olive oil is an economically major product in the Mediterranean countries. The oil is known for its health benefits and gives a fine aroma and a pleasant taste. The quality of olive oil ranges from the low-quality olive-pomace oil (OPO) (or raw residue oil) to high quality extra virgin olive oil (EVOO) (Allam & Hamed, 2007). Water that presents in the olive fruit which are also known as “vegetable” water are up to 70%. The chemical composition of the olive fruit is basically water (50%), carbohydrates (19%), oil (22%), cellulose (5.8%), protein (1.6%) and minerals (ash) (1.5%). There are a few other important constituents including pectin, pigments, organic acids, and glycosides of phenols. Some of the components or the hydrolysis products that are found in the vegetable water which can be squeezed with the oil during processing and are separated by centrifugation (Boskou, 2015).

The high prices of edible oils such as OO or EVOO can cause producers or traders to resort to partial or total substitution of these with lower grade olive oil or other cheaper oils for example sunflower oil (SFO), soybean oil and rapeseed oil. Addition of cheaper, low-

grade or unnecessary substances that can affect the nature and quality of the original food is known as food adulteration. For example, the addition of cheap oil to expensive oils, as in the case of adding refined oils to virgin olive oil (VOO), or the addition of low- to high-quality OO (Abbas & Baeten, 2016). These can cause halal issue which is one of major economic fraud that also give serious consequences for consumer right and health (Poiana et al., 2012). Islam prohibit the act of fraud as mentioned in al-Quran:

﴿وَيْلٌ لِّلْمُطَفِّفِينَ ۝ الَّذِينَ إِذَا أَكْتَالُوا عَلَى النَّاسِ يَسْتَوْفُونَ ۝ وَإِذَا كَالُوهُمْ أَوْ وُزِنُوهُمْ يُخْسِرُونَ ۝ أَلَا يَظُنُّ أُولَٰئِكَ أَنَّهُمْ مَبْعُوثُونَ ۝ لِيَوْمٍ عَظِيمٍ ۝ يَوْمَ يَقُومُ النَّاسُ لِرَبِّ الْعَالَمِينَ ۝﴾

Meaning: “Woe to the defrauders! Those who take full measure ‘when they buy’ from people, but give less when they measure or weigh for buyers. Do such people not think that they will be resurrected, for a tremendous Day, the Day ‘all’ people will stand before the Lord of all worlds?” (al-Quran. al-Mutaffifin 1-6).

Throughout the years, olive oil adulteration has always been a problem and has never been controlled to one region or country only (Torrecilla, 2010). The low production and high price as compared with some other oils making the olive oil (OO) one of the most adulterated vegetable oils. With the intention to improve the control of trade of olive oil, several international institutions such as the International Olive Council (IOC) and the Antifraud Unit of the European Union (OLAF) are focusing on drafting antifraud legislation (Conte et al., 2020). The International Olive Council (IOC) has defined the standard for classifying olive oil according to the chemical, sensory, and organoleptic properties. As for example, extra-virgin olive oil (EVOO) also must have a free acidity percentage of less than 0.8, virgin olive oil (VOO) has a free acidity of less than 1% and OO must have a free acidity of not more than 2% (Gonzalez & Aparicio, 2002).

However, determination of adulteration through physical or chemical properties as mentioned above is not ample. The use of robust technique is necessary. Infrared spectroscopy is a one of spectroscopic technique based on the interaction of infrared radiation with matter. It can be used to identify and quantify compounds which absorb frequencies that are characteristics of their structure such as molecular potential energy surfaces, the masses of the atoms, and the associated vibronic coupling. The infrared spectroscopy (FTIR) has been widely applied in the detection of agricultural products such as wine, olive oil, tea and meat due to the fast and simple operation (Yang et al., 2018).

This study aims to determine the physicochemical differences between olive oil added with sunflower oil at different percentages as well as to determine the spectral differences between olive oil added with sunflower oil by using FTIR. Results presented here will provide information on ability of FTIR to distinguish olive oil (OO) added with sunflower oil (SFO) as there were many research regarding incorporation of extra virgin olive oil into other cheaper oil rather than sunflower oil. The SFO was chosen to be use in present study due to its higher chemical composition similarity with OO such as palmitic acid. By adding different concentration of SFO to the OO, the spectral region can be determined to distinguish between addition samples and control OO.

## METHODOLOGY

### Preparation of Sample

A set of four samples containing olive oil and sunflower oil were mixed together in accurately weighted proportions of 1%, 5%, 10% and 20% v/v (Table 1) and shaken vigorously to ensure the total homogenization. One commercial olive oil was used as standard and the samples containing sunflower oil were assigned as adulterated. Meanwhile, 100% sunflower oil used as negative control. The mixed samples were proceeded with physicochemical analysis and FTIR reading

Name	Addition percentage (%)	Olive oil content (g)	Sunflower oil content (g)
Olive oil (positive control)	0	10	0
Sample 1	1	9.9	0.1
Sample 2	5	9.5	0.5
Sample 3	10	9	1
Sample 4	20	8	2
Sunflower oil (negative control)	0	0	10

Table 1: Addition percentage which assume for every 10mL of olive oil

### FTIR Analysis

FTIR spectra of samples was measured using FTIR Spectrometer (Spectrum One, Perkin Elmer, USA). A Perkin-Elmer Spectrum one FTIR spectrophotometer equipped with a deuterated triglycerine sulphate (DTGS) detector was used to collect FTIR spectra with a resolution of  $4\text{ cm}^{-1}$  at 20 scans. A small quantity ( $2\ \mu\text{L}$ ) of the sample was deposited using a Pasteur pipette between two well-polished KBr disks, creating a thin film. All spectra were recorded and processed with the computer software program Spectrum for Windows (Perkin-Elmer). Each sample was measured in triplicates (Selaimia et al., 2017)

### Specific gravity (SG).

Based on AOCS Method Cc 10a-25 (AOAC, 1984), the specific gravity (SG) of the oil samples were determined starting by the empty SG bottle will be weighed. Then, the SG bottle was filled with the oil, the bottle was closed with stopper which has a capillary bore. The filled SG bottle was cooled at  $7\text{ }^{\circ}\text{C}$  for 24 hours. The next day, the SG bottle was warmed at  $25\text{ }^{\circ}\text{C}$  until the expansion has ceased. The SG bottle was wiped and cleaned on the outside and weighed again. Each sample was measured in triplicates. The SG of the oil sample was calculated using formula:

$$\text{Specific gravity} = \frac{\text{Density of oil (}^g/mL\text{)}}{\text{Density of water (}^g/mL\text{)}}$$

### Refractive Index (RI).

The determination of refractive index (RI) was measured by a Digital ABBE refractometer (AR2008, Kruss Optronic, Germany). The temperature used was  $20\text{ }^{\circ}\text{C}$  for oils. The secondary prism will be opened and 2-3 drops oil was placed at the centre of the main prism. The oil sample was made sure evenly distributed and no presence of air bubble. The secondary prism was closed with caution. While looking through the eyepiece, the measurement knob turned slowly until the boundary line can be observed (this line may not be clear). The measurement knob was turned until the view changes from dark to light and the colour compensator knob was turned to remove the colour of the boundary line. After that, a clear boundary line can be seen. The measurement knob was turned again to coincide the boundary line with the crossed line. The RI as recorded. Each sample was measured in triplicates (Khaled et al., 2018).

### Iodine value.

According to AOCS Official Method Tg 1-64 (AOAC, 1984), approximately 0.15 g of oil sample was weighed into a dry and clean conical flask. About 20 mL of cyclohexane was added to dissolve the oil. Next, 25 mL of Wij's solution was added, the stopper was inserted and the solution was shaken gently. Then, the flask was placed in the dark for 1 hour. After standing, 20 mL of 15% potassium iodide solution and 100 mL of distilled water was added. One mL of starch indicator was added. The solution was titrated with 0.1 M sodium thiosulphate solution until the blue colour disappeared after vigorous shaking. Each sample was

measured in triplicates. The volume of sodium thiosulphate used was recorded and calculated using formula:

$$\text{Iodine value} = \frac{12.69 M(V_b - V_s)}{W}$$

M – Molarity of the sodium thiosulphate solution used  
 V<sub>b</sub> – Volume in mL of the sodium thiosulphate solution used for the blank test  
 V<sub>s</sub> – Volume in mL of the sodium thiosulphate solution used for the sample test  
 W – Weight in gram of the sample

### Acid Value

As stated by AOCS Official Method Cd 3d-63 (AOAC, 1984), 25 mL diethyl ether was mixed with 25 mL ethanol and 1 mL of 1% phenolphthalein solution. Approximately 10 g of fresh oil sample was dissolved in the mixed neutral solvent. The solution was titrated with aqueous 0.1 M NaOH until pink colour persists for 15 seconds obtained. The titre was made sure to not exceed 10 mL, if not two phases may be formed. Each sample was measured in triplicates. The volume of NaOH used was recorded and calculated using formula:

$$\text{Acid value} = \frac{V_t (\text{mL}) \times 0.0256 (\text{g})}{\text{Weight of sample (g)}} \times 100$$

V<sub>t</sub> – Volume in mL of NaOH solution used

### Viscosity

According to Sahasrabudhe *et al.* (2017), the viscosity of the olive oil samples was determined by using Brookfield viscometer (LVT, Brookfield Engineering Laboratories Inc, USA). The spindle selected was spindle number 1 and attached to the shaft. Then, the viscometer was levelled. The oil samples were placed in 250 mL beaker. The spindle was immersed into the oil sample up to the indentation at the neck of the spindle. A suitable speed of 60 rpm will be selected. Next, the viscometer was turned on the readings was collected after the viscometer gave three consistent reading about 1 minutes apart. For the oil samples, the valid dial readings taken between 10 until 90. Each sample was measured in triplicates. The reading taken was expressed as centipoise (cP) and calculated by using the following formula:

$$\text{Viscosity (centipoise)} = \text{Dial reading} \times \text{Factor}$$

### Colour

The colour of the oil samples was determined by using Chroma meter (CR-400, Minolta, Japan). The oil sample was placed in a petri dish and positioned directly on the light. The colour parameter value that measured was lightness (L\*), redness (a\*) and yellowness (b\*). Three reading were taken from each sample. L\* value measure lightness from black to white (1-100); +a\* values represent redness and -a\* value represent greenness; +b\* values represent yellow and -b\* values represent blue. The colour analysis was performed in triplicate for each olive oil, mixed olive oil and sunflower oil (Almeida *et al.*, 2017).

### Statistical Analysis

All analysis were done in triplicate (n=3) and the data was expressed as the mean and standard deviation (SD). All the data were subjected to one way analysis of variance (ANOVA) by using SPSS software. Mean values were compared at p<0.05 significant level by Duncan's multiple range test.

## RESULTS AND DISCUSSION

### FTIR Analysis

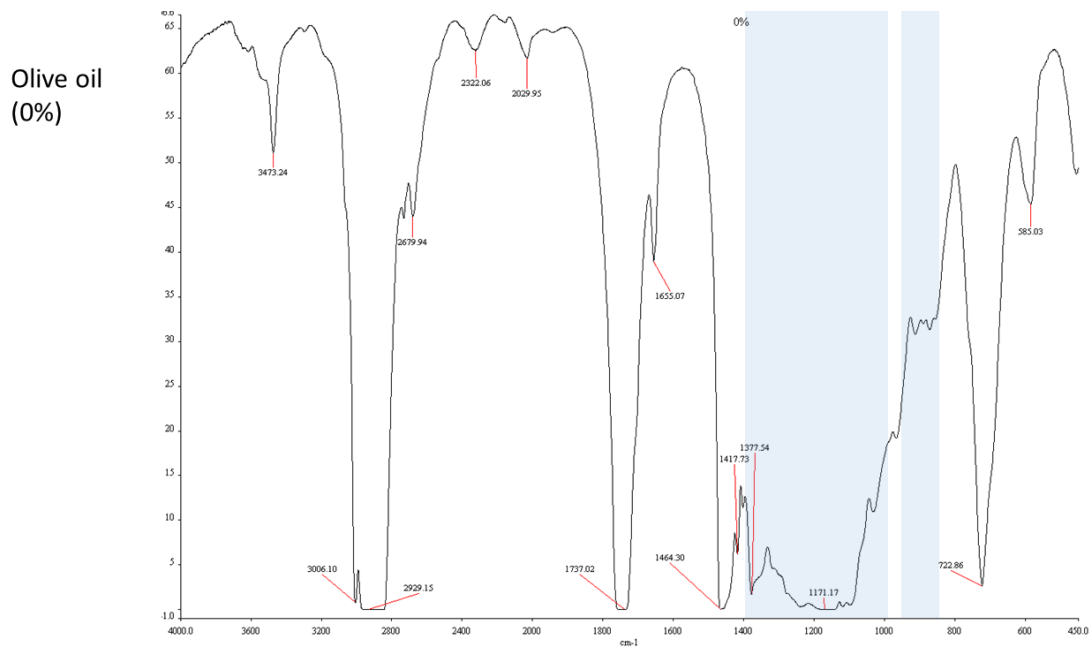
Triglycerides were the principal component in fats and oils and, consequently, dominate FTIR spectra of fats and oils. Based on the spectra, the differences can be seen at the frequency regions of 1402  $\text{cm}^{-1}$ , 965 – 960  $\text{cm}^{-1}$ , 872 – 850  $\text{cm}^{-1}$  and 1300 – 1000  $\text{cm}^{-1}$ . Rohman and Che Man [19] stated that, there was a difference in the spectra which can be seen at frequency of 1402  $\text{cm}^{-1}$  caused by =C-H bending. A study conducted by Rohman et al. (2010) reported that the difference between the spectrums can be seen in the frequency region of 965 – 960  $\text{cm}^{-1}$  that showed the chemical bonds of –HC=CH. According to Rohman and Che Man (2010) there were difference in the spectrum at 872 – 850  $\text{cm}^{-1}$  due to =CH<sub>2</sub> bonds. The differences of the spectra for these three regions were mainly due to the double bond. As the addition percentage of the sunflower oil into the olive oil increases, the amount of linoleic acid increases thus the increases of the amount of double bond present. According to Allam and Hamed (2007), there were visual differences in the peak at 1300 – 1000  $\text{cm}^{-1}$  and it was very useful to detect olive oil adulteration as it showed C–O stretching and CH<sub>2</sub>–bending. This was due to increases the addition percentage will decrease the amount of carbonyl and alkane group. Below showed the overlay spectra of olive oil, mixture of olive oil and sunflower oil. The summary of the frequency region and chemical bonds that showed difference in olive oil, mixture of olive oil and sunflower oil can be seen in the table below.

Frequency Region ( $\text{cm}^{-1}$ )	Chemical Bonds	Reference
1402	=C-H bending	Rohman & Che Man (2010)
965 – 960	–HC=CH	Rohman et al. (2010)
872 – 850	=CH <sub>2</sub> wagging	Rohman et al. (2010)
1300 – 1000	C–O stretching CH <sub>2</sub> – bending	Allam & Hamed (2007)

Table 2: The summary of the frequency region and chemical bonds of olive oil, mixture of olive oil and sunflower oil

*Specific gravity (SG).* The specific gravity was the ratio between the density of an object, and a reference substance. Result for specific gravity analysis with different percentage of adulteration was shown in Table 3 below. Mean within each column with different superscript are significantly different at  $p < 0.05$ . From this study, it was found that specific gravity for control olive oil is lower (0.9087) than sunflower oil with 0.9475. These could be due to the specific gravity of unsaturated glycerides was higher than the corresponding saturated ones, sunflower oil had higher unsaturated fatty acid as compared to olive oil. When comparing these results to Codex Alimentarius Commission [9], the specific gravity for olive oil should be 0.910-0.916 while sunflower oil should be 0.918-0.923 which were different form the study. However, by comparing the results from Fakhri (2011), specific gravity for olive oil was nearly the same with 0.8974. As the percentage of addition increases, the specific gravity also increases linearly with 0.9096, 0.9151, 0.9174 and 0.9385. Increases of the specific gravity may be attributed by the different fatty-acid composition, different total solid content and different degree of saturation (O'Brien, 2009). As sunflower oil was added to the olive oil causing the changes in the amount of linoleic acid in the oil thus resulting in the increases of specific gravity. Although the specific gravity increases, there was no significant difference between the control olive oil with 1% of addition.

a. Olive Oil (Positive Control)



b. Sunflower Oil (Negative Control)

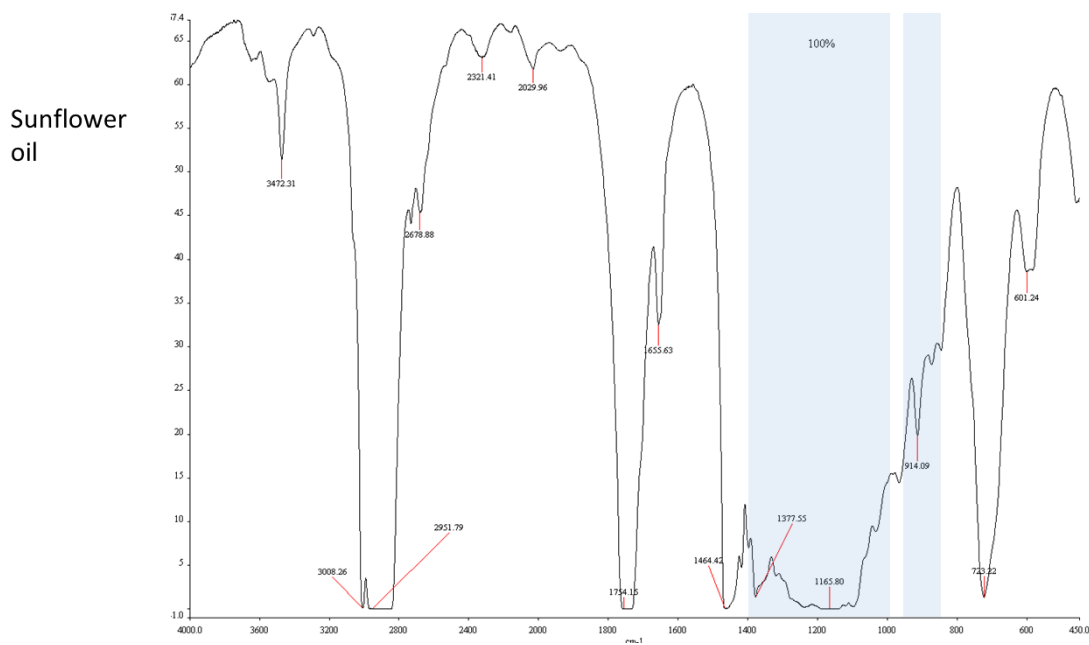


Figure 1: FTIR spectra of a) Olive oil 0% and b) Sunflower oil 0%. The highlighted regions indicate the observed spectra differences as summarised in Table 2.

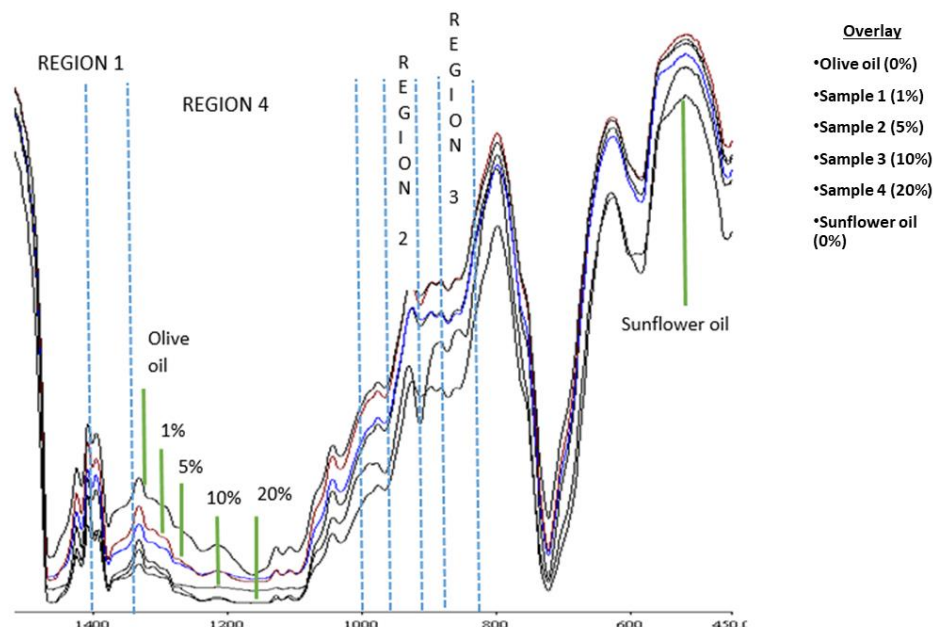


Figure 2: FTIR spectra of Olive oil, samples adulterated with sunflower oil (1, 5, 10 and 20%) and Sunflower oil. The dotted line indicates the observed spectra differences as summarised in Table 2.

**Refractive index (RI).** Refractive index known as the ratio of the velocity of light in a vacuum to its velocity in a specified medium. The refractive index for olive oil was 1.4676 while sunflower oil was 1.4723. This is consistent with what has been found in previous study conducted by Mengistie et al. (2018), the refractive index for sunflower oil was 1.475 at 30 °C while another study conducted by Fakhri (2011) stated that the refractive index for olive oil was 1.4670 at 30°C. According to Table 3, the results for refractive index for addition percentage of 1%, 5%, 10% and 20% were 1.4682, 1.4678, 1.4676 and 1.4687. There was no significant difference between control olive oil and addition percentage with 1% and 5%. The significant difference only can be seen at addition percentage of 10% and above. Refractive index of fats and oils was a basic value that relates to molecular weight, fatty acid, chain length, the degree of unsaturation and degree of conjugation. Sunflower (876.16 g/mol) had higher molecular weight than olive oil (282.468 g/mol). This is consistent with what has been found in previous study by Silla *et al.* (2014) which stated there was a general tendency for the refractive index to increase as the molecular weight of liquid increases. From the result, refractive index cannot be used to determine the addition of sunflower oil in olive oil at low concentration only 10% and above.

**Iodine value (IV).** Iodine value also called iodine number was known to measure the degree of unsaturation of an oil or fat. From the results, it is clear that the iodine value for the olive oil (77.4742) was lower as compared to sunflower oil (101.8631). According to Codex Alimentarius Commission [9], the iodine value for olive oil should be 75-94 while for sunflower oil was 118-141. The iodine value for olive oil (77.4742) was nearly the same as a study conducted by Abdel-Razek *et al.* (2011) which had the iodine value of 83. Contrary to the findings of Roiaini *et al.* (2015) with the iodine value of 94 which may due to differences in cultivar and purities of olive oil sources. In line with previous studies by Konuskan et al. (2018), the iodine value for sunflower oil was 102.02 which was quite the same as the result 101.8631.

Other than that, the iodine value increased to 79.0240, 80.9428, 84.2054 and 91.4883 as the percentage of addition increased. These was due to the amount of double bonds present which react with iodine compounds. The higher the iodine value, the more C=C bonds were present in the oil. In line with previous studies done by Siddique et al.

(2010), the increment of iodine value was due to the fact that after the oils were adulterated together, their degree of unsaturation changed leading to changed iodine value. Although the iodine value increased, there was no significant difference between the control olive oil with 1% of addition percentage.

**Acid value (AV).** The acid value (AV) was a common parameter in the specification of fats and oils. Table 3 showed the results of acid value with different percentage of adulteration of sunflower oil to olive oil. The results demonstrated that olive oil (0.1788) had a significantly lower acid value, and the acid value for sunflower oil was much higher (0.8172). The findings were directly in line with previous findings by Fakhri (2011), which reported that the acid value for one of the sunflower oils used was 0.802. As for olive oil, Ashif (2017) reported in their study that the acid value was 0.24 which was slightly higher than from the result. The result also showed an increase of acid value with the increasing addition percentage. These could be due to the amount of free fatty acid present in the adulterated oil as sunflower had higher acid value than olive oil. Low acid value in oil indicates that the oil will be stable over a long period of time and protect against rancidity and peroxidation. This could be attributed to presence of natural antioxidants in the seeds such as vitamins C and A as well as other possible phytochemical like flavonoids (Aremu et al., 2015). Even when the acid value increases, there was no significant difference between the control olive oil with 1% addition percentage. Overall, the analysis of acid value can only be used to determine the addition of sunflower oil in olive oil above 1%.

**Viscosity.** Viscosity is resistance of a fluid (liquid or gas) to a change in shape. This analysis was done to know which oil was more viscous. From this finding, it was suggested that the viscosity for control olive oil was higher with 130.00 cP and sunflower oil was lower with 110.00 cP. Viscosity was closely correlated with the structural parameters of the fluid particles and the value increases with increasing degree of saturation. The study done by Fakhri (2011) reported the value of 69.998 cP for olive oil and 59.254 cP for sunflower oil at 25 °C. The vast difference could be due to the differences in the equipment used and the spindle used. Table 3 showed that olive oil was more viscous as compared to sunflower oil. The viscosity decreased as the addition percentage increased. According to Rubalya Valantina et al. (2013), the viscosity of olive oil decreases with the addition of sunflower oil due to factors like density, molecular weight and unsaturation. In this case, the reason was the content of large amount of unsaturated fatty acid in sunflower oil. Although the viscosity decreased, there was no significant difference between the control olive oil with 1% addition level.

Table 3: The results of physicochemical properties of olive oil, mixture of olive oil & sunflower oil

Sample (with percentage addition of SFO)	Specific gravity (SG).	Refractive index (RI).	Iodine value (IV).	Acid value (AV).	Viscosity.
Olive oil (0%)	0.9087 ± 0.0004 <sup>e</sup>	1.4676 ± 0.0003 <sup>d</sup>	77.4742 ± 0.8504 <sup>e</sup>	0.1788 ± 0.0003 <sup>e</sup>	130.00 ± 2.500 <sup>a</sup>
Sample 1 (1%)	0.9096 ± 0.0013 <sup>e</sup>	1.4682 ± 0.0002 <sup>d</sup>	79.0240 ± 0.3596 <sup>e</sup>	0.1785 ± 0.0006 <sup>e</sup>	128.17 ± 1.4434 <sup>a</sup>
Sample 2 (5%)	0.9151 ± 0.0010 <sup>d</sup>	1.4678 ± 0.0002 <sup>d</sup>	80.9428 ± 0.1880 <sup>d</sup>	0.2087 ± 0.0006 <sup>d</sup>	120.83 ± 1.4434 <sup>b</sup>
Sample 3 (10%)	0.9174 ± 0.0003 <sup>c</sup>	1.4676 ± 0.0001 <sup>c</sup>	84.2054 ± 0.3067 <sup>c</sup>	0.2308 ± 0.0003 <sup>c</sup>	119.17 ± 1.4434 <sup>b</sup>
Sample 4 (20%)	0.9385 ± 0.0005 <sup>b</sup>	1.4687 ± 0.0001 <sup>b</sup>	91.4883 ± 0.4882 <sup>b</sup>	0.2784 ± 0.0151 <sup>b</sup>	115.83 ± 1.4434 <sup>c</sup>
Sunflower oil (0%)	0.9475 ± 0.0018 <sup>a</sup>	1.4723 ± 0.0011 <sup>a</sup>	101.8631 ± 1.8789 <sup>a</sup>	0.8172 ± 0.0094 <sup>a</sup>	110.00 ± 2.500 <sup>d</sup>

Mean within each column with different superscript are significantly different at p<0.05

*Colour.* The value of L\* for the lightness from black (0) to white (100), a\* from green (-) to red (+), and b\* from blue (-) to yellow (+). From Table 4, the L\* value for olive oil was  $24.84 \pm 0.1960$ , the a\* value was  $-0.63 \pm 0.0265$  and the b\* value was  $3.19 \pm 0.0361$ . Sunflower oil obtained L\* value of  $25.49 \pm 0.0100$ , a\* with the value of  $-0.12 \pm 0.0611$  and b\* value of  $1.94 \pm 0.0322$ . These indicated that the olive oil was darker, yellowish and more greenish in colour while the sunflower oil was lighter, less greenish and less yellowish. In line with international food standard by Codex Alimentarius Commission (2013), the colour of olive oil should be yellow to green.

Table 4: Colour of olive oil, mixture of olive oil and sunflower oil

Sample (with percentage addition of SFO)	L*	a*	b*
Olive oil (0%)	$24.84 \pm 0.1960^c$	$-0.63 \pm 0.0265^a$	$3.19 \pm 0.0361^a$
Sample 1 (1%)	$25.04 \pm 0.0058^b$	$-0.63 \pm 0.0153^a$	$2.93 \pm 0.0208^b$
Sample 2 (5%)	$23.89 \pm 0.0100^d$	$-0.62 \pm 0.0100^a$	$2.93 \pm 0.0351^b$
Sample 3 (10%)	$25.07 \pm 0.0116^b$	$-0.62 \pm 0.0116^a$	$2.67 \pm 0.0200^c$
Sample 4 (20%)	$25.63 \pm 0.0000^a$	$-0.62 \pm 0.0264^a$	$2.68 \pm 0.0764^c$
Sunflower oil (0%)	$25.49 \pm 0.0100^a$	$-0.12 \pm 0.0611^b$	$1.94 \pm 0.0322^d$

Mean within each column with different superscript are significantly different at  $p < 0.05$

The results describe that there were significant differences for L\* and b\* as the addition percentage increased. The value for a\* did not have any significant difference. The L\* value increases due to sunflower oil was much lighter colour as compared to the olive oil. According to Escolar et al. (2007) and Gonzalez et al. (2007), some olive oils also had low b\* values due to effect of their pigment contents, which were high enough in some olive oils to give coordinates falling within the zone for most virgin or extra virgin oils. A degraded or adulterated sample can exhibit a carotenoid profile which decreased the b\* value.

The findings above describes the used of FTIR, specific gravity, refractive index, iodine value, acid value and viscosity in determining the authenticity of EVOO. As the name Extra Virgin is indicated, such marketed olive oil must be in high purity. Any attempt to substitute, mixed or combined with cheaper version (Olive Oil) is considered as non-authentic.

Halal authenticity is an issue of major concern in the food industry. Many cases were reported worldwide involving adulteration of haram or mushbooh ingredients in foods productions. A lot of cases related to olive oil happened globally, and might affect Malaysia as the importer of this commodity. For instance, in April 2008, an "Operation Golden Oil" conducted by 400 Italian police officers confiscated seven olive oil plants that arrested 40 people in nine provinces of northern and southern Italy because they added chlorophyll to sunflower and soybean oil to make it more green and selling it as EVOO, both in Italy and abroad. 25,000 litres of the fake oil were seized and prevented from being exported. As mention by Butleron in February 14, 2012, in the article form Olive Oil Times there was alleged international olive oil scam in which palm, avocado, sunflower and other cheaper oils were passed off as OO. The oils were blended in an industrial biodiesel plant and adulterated in a way to hide markers that would have revealed their true nature.

Therefore it is hope that the method that is being developed in this study will help authoritative body to discriminate or authenticate the products in the market in order to safeguard Muslim customers while selecting food goods for consumption. Additionally, the task of halal verification requires competence from other associated technical domains, such as food science and technology, chemistry, and veterinary science, and cannot be only based on knowledge from the shariah. Halal verification now requires the use of cutting-edge, high-tech analytical instrumentation in addition to physical inspection and documentation.

## CONCLUSION

Analysis of specific gravity, refractive index, iodine value, acid value and viscosity can be used to determine difference at the addition percentage of 5%. Refractive index only can detect differences at 10% and above. While, colour analysis can detect differences at 1% addition level. The spectral differences between olive oil added with sunflower oil by using FTIR can be seen at frequency region 1402  $\text{cm}^{-1}$  caused by =C-H bending, 1300 - 1000  $\text{cm}^{-1}$  caused by C-O stretching and CH<sub>2</sub>- bending, 965 - 960  $\text{cm}^{-1}$  caused by -HC=CH and 872 - 850  $\text{cm}^{-1}$  caused by =CH<sub>2</sub> wagging. Further study on physicochemical analysis should be carried out such as fatty acid composition. Chemometrics analysis, including quantification using partial least squares (PLS) and principal component regression (PCR) calibrations and discriminant analyses can be done to obtain more specific and detail results for FTIR analysis.

## ACKNOWLEDGEMENT

Authors would like to thank Vanguard Research Grant, MITRANS for funding this publication and Food Processing Laboratory and Food Analysis Laboratory, Faculty of Applied Sciences, Universiti Teknologi MARA for the access given.

## REFERENCES

### Book

- Association of Official Analytical Chemists. 1984. *Official method of analysis of the Association of official Analytical chemist (AOAC)*. Washington D.C, USA.
- Boskou, D. 2015. *Olive and olive oil bioactive constituents*. Illinois, United States: AOCS Press.
- O'Brien, R. D. 2009. *Fats and Oils*. CRC Press. Taylor and Francis Group, 3<sup>rd</sup> Ed., 2-52.
- Preedy, V. & Watson, R. 2010. *Olives and olive oil in health and disease prevention*. Amsterdam: Academic Press, 1st Ed.
- Silla, E., Arnau A. & Tuñón, I. 2014. *Fundamental Principles Governing Solvents Use*. In: Handbook of Solvents, 11-72.

### Journal

- Abbas, O., & Baeten, V. 2016. *Advances in the identification of adulterated vegetable oils*. *Advances in Food Authenticity Testing*, 519-542. <https://doi.org/10.1016/B978-0-08-100220-9.00019-9>
- Abdel-Razek, A. G., El-Shami, S. M., El-Mallah, M. H. & Hassanien, M. M. M. 2011. *Blending of virgin olive oil with less stable edible oils to strengthen their antioxidative potencies*. *Australian Journal of Basic and Applied Sciences*. 5(10), 312-318.
- Allam, M. A. & Hamed, S. F. 2007. *Application of FTIR spectroscopy in the assessment of olive oil adulteration*. *Journal of Applied Sciences Research*. 3(2), 102-108.
- Almeida, D. T., Curvelo, F. M., Costa, M. M., Viana, T. V. & Lima, P. C. 2017. *Oxidative stability of crude palm oil after deep frying akara Fried Bean Paste*. *Food Science and Technology*. <https://doi.org/10.1590/1678-457x.02217>
- Aremu, M., Ibrahim, H. & Bamidele, T. 2015. *Physicochemical characteristics of the oils extracted from some Nigerian plant foods: A Review*. *Chemical and Process Engineering Research*. 32, 36-52.
- Ashif S. 2017. *Physicochemical Analysis and Fatty Acid Composition of Oils Extracted from Two Exotic Cultivars of Olive (Olea Europaea L.) Cultivated In Balochistan, Pakistan*. *Indo American Journal of Pharmaceutical Sciences*, 4(10). DOI: 10.5281/zenodo.1039702

- Conte, L., Bendini, A., Valli, E., Lucci, P., Moret, S., Maquet, A., Lacoste, F., Brereton, P., García-González, D. L., Moreda, W., & Toschi, T.G. 2020. *Olive oil quality and authenticity: A review of current EU legislation, standards, relevant methods of analyses, their drawbacks and recommendations for the future*. Trends in Food Science & Technology, 105, 483-493.
- Fakhri, N. 2011. *Studies on Various Physico-Chemical Characteristics of Some Vegetable Oils*. Journal of Environmental Engineering and Science, 5.
- Gonzalez, D., Haro, M. R., & Ayuso, J. 2007. *The Color Space of Foods: Virgin Olive Oil*. Journal of Agricultural and Food Chemistry, 55(6), 2085-2093. <https://doi.org/10.1021/jf062899v>
- Gonzalez, D. R., Aparicio, R. 2002. *Detection of defective virgin oils by metal oxide sensors*. Journal of Agricultural and Food Chemistry (Eur Food Res Technol). 215, 118-123. <https://doi.org/10.1007/s00217-002-0527-9>
- Khaled, R., Souhail, N. & Amina, I. 2018. *Orange peel fixed oil (citrus sinensis "valencia"), physicochemical properties, fatty acids profile, potential uses and the effect of environmental factors on it*. Bulgarian Journal of Agricultural Science, 24(1), 91-98.
- Konuskan, D. B., Arslan, M., & Oksuz, A. 2018. *Physicochemical properties of cold pressed sunflower, peanut, rapeseed, mustard and olive oils grown in the Eastern Mediterranean region*. Saudi Journal of Biological Sciences, 26 (2), 340-344. <https://doi.org/10.1016/j.sjbs.2018.04.005>
- Mengistie, T., Alemu, A. & Mekonnen, A. 2018. *Comparison of physicochemical properties of edible vegetable oils commercially available in Bahir Dar, Ethiopia*. Chemistry International 4(2), 130-135.
- Poiana, M. A., Mousdis, G., Alexa, E., Moigradean, D., Negrea, M., & Mateescu, C. 2012. *Application of FT-IR spectroscopy to assess the olive oil adulteration*. Journal of Agroalimentary Processes and Technologies, 18(4), 277-282.
- Rohman, A., & Che Man, Y. B. 2010. *Fourier transform infrared (FTIR spectroscopy for analysis of extra virgin olive oil adulterated with palm oil*. Food Research International, 43(3), 886-892. <https://doi.org/10.1016/j.foodres.2009.12.006>
- Rohman, A., Che Man, Y. B., Ismail, A., & Hashim, P. 2010. *Application of FTIR spectroscopy for the determination of virgin coconut oil in binary mixtures with olive oil and palm oil*. Journal of the American Oil Chemists' Society, 87(6), 601-606. <https://doi.org/10.1007/s11746-009-1536-7>
- Roiaini, M., Ardiannie, T. & Norhayati, H. 2015. *Physicochemical properties of canola oil, olive oil and palm olein blends*. International Food Research Journal 22(3), 1227-1233.
- Rubalya Valentina, S., Chandiramouli, R., & Neelamegam, P. 2013. *Detection of adulteration in olive oil using rheological and ultrasonic parameters*. International Food Research Journal, 20(6), 3197-3202.
- Sahasrabudhe, S. N., Rodriguez-Martinez, V., O'Meara, M., & Farkas, B. E. 2017. *Density, viscosity, and surface tension of five vegetable oils at elevated temperatures: Measurement and modeling*. International Journal of Food Properties, 1-17. <https://doi.org/10.1080/10942912.2017.1360905>
- Selaimia, R., Oumeddour, R., & Nigri, S. 2017. *The chemometrics approach applied to FTIR spectral data for the oxidation study of Algerian extra virgin olive oil*. International Food Research Journal, 24(3), 1301-1307.
- Siddique, B. M., Ahmad, A., Ibrahim, M. H., Hena, S., Rafatullah, M. & Mohd Omar, A. K. 2010. *Physicochemical properties of blends of palm olein with other vegetable oils*. Journal of Grasas Y Aceites, 61(4), 423-429. DOI: <https://doi.org/10.3989/gya.010710>
- Torrecilla, J.S., Rojo, E., Domínguez, J.C., & Rodríguez, F. 2010. *A novel method to quantify the adulteration of extra virgin olive oil with low-grade olive oils by*

- UV-vis. Journal of Agricultural and Food Chemistry, 87-90.  
<https://doi.org/10.1021/jf903308u>
- Yang, L., Wu, T., Liu, Y., Zou, J., Huang, Y., Babu, S. V., & Lin, L. 2018. *Rapid identification of pork adulterated in the beef and mutton by infrared spectroscopy.* Journal of Spectroscopy, 81-90.  
<https://doi.org/10.1155/2018/2413874>

#### **Document**

- Codex Alimentarius Commission. 2013. *Standard for Olive Oils and Olive Pomace Oils (Codex Stan 33-1981)*. Rome: World Health Organization : Food and Agriculture Organization of the United Nations.

#### **Internet**

- Butleron, J. 2012. Spanish police say palm, avocado, sunflower was passed off as olive oil. Olive Oli Times.  
<https://www.oliveoiltimes.com/business/europe/operation-lucerna-olive-oil-fraud/24815> (accessed on 06 February 2022)
- Moore, M. 2010. Italian police crackdown on olive oil fraud. The Daily Telegraph.  
<https://www.telegraph.co.uk/news/worldnews/1580807/Italian-police-crack-down-on-olive-oil-fraud.html> . (accessed on 26 February 2022)

#### **Disclaimer**

*Opinions expressed in this article are the opinions of the author(s). Al-Qanatir: International Journal of Islamic Studies shall not be responsible or answerable for any loss, damage or liability etc. caused in relation to/arising out of the use of the content.*