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Rapid detection of olive oil blends using a paper-based portable microfluidic platform

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Abstract. This paper presents a portable microfluidic platform based on a filter paper on which multi-walled carbon nanotubes were deposited to quickly determine the quality of olive oil by measuring electrical resistance. Three different types of filter paper with different pore sizes and different filtration rates were used in the middle of the microfluidic platform, as a material for soaking a blended olive oil and high-oleic sunflower oil. The rapid prototyping xurographic technique was used to fabricate the complete microfluidic platform. For testing purposes, oil blends in various proportions were deposited through the inlet on the top of the platform. The variation in electrical resistance at room temperature was measured, using the Chemical Impedance Analyzer and different blended oils were successfully detected. Additionally, a prototype of electronic device was developed for acquisition and displaying measured data, based on the created microfluidic platform.

Key words: oil blends, olive oil, microfluidic platform, paper, MWCNT

1. Introduction

Olive oil is obtained from the fruit of olive tree (*Olea europea* L.), the oldest known cultivated tree in history (Tsopelas, Konstantopoulos, & Kakoulidou, 2018). This product is being consumed worldwide, but it is especially important in culinary arts of Mediterranean countries (Filoda et al., 2019). European Union (EU) is considered the biggest producer of olive oil in the world (de la Mata-Espinosa, Bosque-Sendra, Bro, & Cuadros-Rodriguez, 2011). It possesses a higher nutritional value and specific sensorial qualities compared to other types of edible vegetable oils, thus providing a unique taste and many benefits to human health. It is a source of: monounsaturated fatty acids; antioxidant and anti-inflammatory compounds, such as phytosterols, squalene and various types of polyphenols and phenolic acids (Ruiz-Samblás, Marini, Cuadros-Rodríguez, & González-Casado, 2012; Zhang et al., 2017; de la Mata et al., 2012; Carranco, Farrés-Cebrián, Saurina, & Núñez, 2018). Due to the costs associated with oil production, such as olive cultivation, oil harvesting and extraction, the extra virgin olive oil (EVOO) is of the highest quality and price on the market, compared with other types of vegetable oils (Filoda et al., 2019). Due to increasing popularity and higher price, it has always been a target for fraudulent practices, such as substitution with cheaper oils (Jiménez-Carvelo, Osorio, Koidis, González-Casado, & Cuadros-Rodríguez, 2017a). The *Database of Food Ingredient Fraud and Economically Motivated Adulteration* declared that olive oil is the most common target for adulteration, as published in scientific journals, which makes the development of the methods for its quality evaluation an important issue (Zhang et al., 2017). The EU legislation attempts to protect the consumer from possible fraudulent practices, forcing producers to clearly specify the real composition of the product containing olive oil blends (de la Mata et al., 2012). Legal requirements have

been established in *Commission Regulation EU No. 29/2012*, concerning commercialization and labelling of products which contain olive oil and its blends with other edible vegetable oils (Ruiz-Samblás, Marini, Cuadros-Rodríguez, & González-Casado, 2012). This regulation strictly declares that the percentage of olive oil should be clearly shown on the product label (Commision Regulation No. 29, 2012; Jiménez-Carvelo, González-Casado, & Cuadros-Rodríguez, 2017b). Furthermore, in the case where more than 50% of an oil blend consists of olive oil, its presence may be highlighted by images or graphics on the product label. Consequently, there is an urge for developing analytical methods for verifying if the percentage of olive oil in a blend is lower or higher than 50%. Although the field of olive oil blends has been regulated for more than 10 years by EU legislation, it is important to point out that there is still no official method for quantification of olive oil in vegetable oil blends (Monfreda, Gobbi, & Grippa, 2014). An extensive literature data has been published, which discusses the suitability of a wide range of methods for evaluation of quality and detection of adulterants in olive oil (Parker et al., 2014). Generally, two main approaches have been described in last decade: (i) the application of Gas (Rohman, & Che Man, 2012; Ruiz-Samblás, Marini, Cuadros-Rodríguez, & González-Casado, 2012; Monfreda, Gobbi, & Grippa, 2014; Pastor et al, 2019a; Pastor, Ilić, Vujić, Jovanović, & Ačanski, 2019b) and Liquid Chromatography (Jiménez-Carvelo, González-Casado, & Cuadros-Rodríguez, 2017b; Carranco, Farrés-Cebrián, Saurina, & Núñez, 2018; de la Mata-Espinosa, Bosque-Sendra, Bro, & Cuadros-Rodriguez, 2011) with various sources of detection, which is costly, labor-intensive, time consuming, requires sample pre-treatment and the use of organic solvents and reagents, and (ii) the application of various spectroscopic techniques, such as: Fourier-transform infrared spectroscopy (Rohman, & Che Man, 2012; de la Mata et al., 2012; Jiménez-Carvelo, Osorio, Koidis, González-Casado, & Cuadros-Rodríguez, 2017a; Filoda et al., 2019), Raman spectroscopy (Dong, Zhang, Zhang, & Wang, 2012), Fluorescence spectroscopy (Mabood et al., 2016), Nuclear magnetic resonance spectroscopy (Parker et al., 2014) and Mass spectrometry (Liu, Zhu, Wang, Fuchser, & Galvin, 2017), which are usually very costly and hard to be used *on-site* (Tsopelas, Konstantopoulos, & Kakoulidou, 2018). Furthermore, Tsopelas *et al.* (Tsopelas, Konstantopoulos, & Kakoulidou, 2018) proposed the application of electrochemical voltammetric technique for the detection of olive oil adulteration. All these applications are usually combined with the employment of a wide range of supervised and/or unsupervised multivariate chemometric and machine learning algorithms for data treatment, such as Principal component regression, Partial least-squares regression and Support-vector machines (Ruiz-Samblás, Cadenas, Pelta, & Cuadros-Rodríguez, 2014; Horvat, & Horvat, 2016; Horvat, Horvat, & Isic, 2017a; Horvat, Horvat, Rosić, Zindovic, & Kapor, 2017b). Kakani *et al.* (Kakani, Chandu, & Karthikeyan, 2019) reported non-destructive method for estimating up to 50% of adulteration in edible oil applying an open complementary split ring resonator structure and measuring shift in the resonant frequency. Additionally, a gap waveguide cavity resonator was used as a sensing element for detection of different types of oils based on identification of their permittivity (in the range from 2.6 to 4.7) (Alhegazi et al., 2018). Osman *et al.* (Osman, Korostynka, Mason, Cullen, & Al-Shamma'a, 2015) demonstrated the variation of the peak of S_{21} (scattering parameter) from -64.62 to -67.55 dBm, for 100% olive oil to 100% sunflower oil in their blends, respectively. Electrochemical sensor with multi-walled carbon nanotubes (MWCNT) was reported in (dos Santos Moretti et al., 2016), for determination of antioxidants from biodiesel. Additionally, the sensor based on polyaniline-carbon nanotubes was used for phenol detection of phenol from oilfield wastewater (Liu et al., 2019). The pencil-drawn paper-based miniature device was presented in (Dossi et al., 2016), with the aim to detect ortho-diphenols extracted from EVOO, proving the quality of the oil. The indicators of the quality of olive oils are also acidity and peroxide index. Grossi *et al.* (Grossi, Di Lecce, Toschi, & Riccò, 2014) described the method for measuring olive oil acidity by electrochemical impedance spectroscopy. The same group of authors presented the optoelectronic method for determination of peroxide value of olive oil (Grossi, Di Lecce, Arru, Toschi, & Riccò, 2015). Microfluidic *lab-on-a-chip* devices are accurate, rapid and practical, because they integrate many functions - from sample preparation to detection, combined in a

small apparatus. They have been applied in various fields and applications: for the detection of mycotoxins in foods (Guo, Feng, Fang, Xu, & Lu, 2015), detection of biomarkers related to various diseases (Tian et al., 2019), evaluation of cytotoxicity of metal contaminants (Tan et al., 2019), rapid measurements of pH values (Lu et al., 2020) and many others. The microdevice was developed in (Ramos, Contreras, & Macías, 2020) using microfluidic technique for determination of polyphenols from EVOO.

The aim of this study is to fulfil a need for a rapid analytical method to verify the percentage of olive oil declared on the product label. The proposed study focuses on the quantification of EVOO in a simulated blend with a cold-pressed high-oleic sunflower oil (HOSO) using a paper-based microfluidic chip as a rapid screening tool with a considerable potential. The HOSO was chosen as a relatively cheap model adulterant, because its fatty acid composition is very similar to that of olive oil. To the authors' knowledge, there is no literature data reporting the application of this technique in determining adulteration degree of EVOO.

2. Materials and methods

2.1 Fabrication of a portable microfluidic platform

The portable microfluidic platform was manufactured using a rapid and economical xurography technique, which is based on laminating cost-effective polyvinyl chloride (PVC) foils in order to create the multi-layered compact structure. PVC foils were cut in the desired shape by means of the cutter plotter machine. In the middle of the structure, MWCNTs-modified filter paper was embedded in order to create electrically conductive structure. The main fabrication steps are shown in Figure 1.

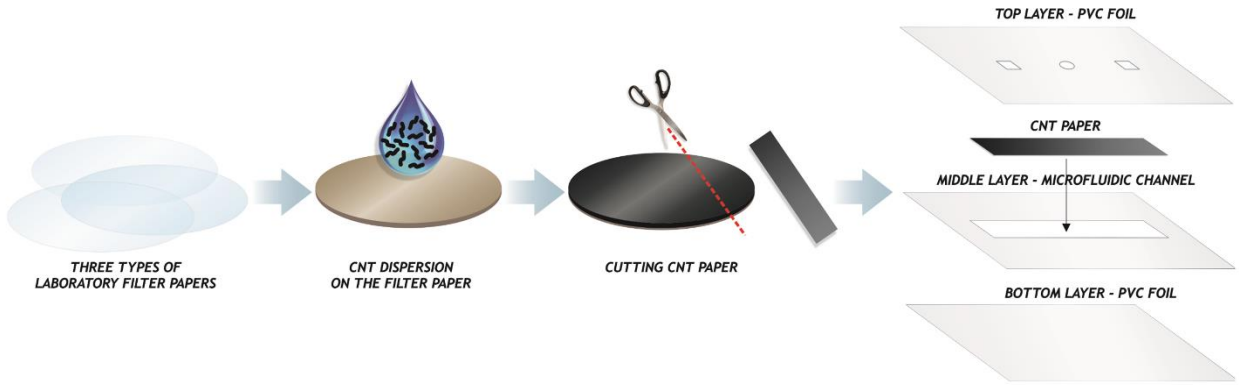

Figure 1. Main fabrications steps in the realization of the microfluidic platform

The bottom layer of the microfluidic platform was 80 μm thick PVC foil. The middle layer was PVC foil with a carved channel in which a piece of filter paper was inserted, before lamination. The top layer is also made of PVC foil with three holes. The middle one was used to inject the oil blend, whereas two outer square holes were used to access the contacts through which the electrical resistance can be measured. Three different types of commercially available filter paper from company Gramed (https://grамed.rs/proizvodi/potrosni-materijal/filtracija/kvantitativni-filter-papiri/) labelled with No. 42, No. 44, and No. 45 were used for realization of the central part of the microfluidic chip. The performances of these papers are presented in Table 1, and it can be seen that they differ in pore size and filtration rate.

Table 1. Characteristics of the three types of paper labelled with No. 42, No. 44 and No. 45

Filter paper	**No. 42**	**No. 44**	**No. 45**
Thickness (mm)	0.2	0.16	0.17
Pore size (μm)	20-25	7-9	2-4
Filtration rate	27	140	195

The above-mentioned filter papers were functionalized by MWCNTs in order to create electrically conductive structure. MWCNTs synthesis was performed for 1 h in a flow of ethylene/nitrogen mixture (1:1) at 700 °C, using an in situ pre-reduced 5% Fe-Co/Al_2O_3. The obtained raw material was boiled under reflux for 6 h in diluted NaOH and 16 h in concentrated HNO_3. The resultant sample was collected on a filter and rinsed with distilled water until a pH neutral followed by drying at 110 °C for 24 h. The obtained functionalized nanotubes (100 mg) were dispersed in 2-propanol and a certain amount of polyvinylpyrrolidone (PVP) was added with the aim to decrease the MWCNTs agglomeration affinity. The MWCNTs dispersion was processed by ultrasonication treatment for 15 min at 5 °C. The MWCNTs dispersion was deposited on three types of filter paper substrates having different pore size, as can be seen in Table 1. The prepared dispersion was deposited on a vertically positioned substrate to ensure that any excess dispersion was drained.

At the top PVC foil, three holes were manufactured of which two square dimensions 3 mm × 3 mm, for access to electrical contacts (the distance between these two holes was 15 mm) and a circular hole in the middle of the structure, 4 mm in diameter as an inlet for oil blends. The three layers of PVC were laminated together at 130 °C to obtain a compact microfluidic platform, as can be seen in Figure 2. The overall dimensions of the microfluidic platform were 6 cm × 3 cm. The weight of the microfluidic platform with inserted filter paper with MWCNTs was equal to 1.32 g.

Figure 2. Fabricated microfluidic platform with MWCNTs-functionalized filter paper in the middle of the structure

2.2 Preparation of oil samples for testing

The oils used in the experiments were a blend of two types of oil, namely, EVOO and cold pressed HOSO. Three different blends were prepared. The first blend contains 20% olive and 80% sunflower oil, the second blend contains 50% olive and 50% sunflower oil, and the third blend contains 70% olive and 30% sunflower oil. The blends were prepared by weighing the exact mass of oil on a technical scale and then homogenized

on a Vortex machine. The amount of oil injected into the microfluidic platform in the experiments was 0.1 ml.

2.3 Characterization methods

For structural analysis of filter paper samples by SEM and AFM methods, the Digitized Scanning Microscope JEOL JSM 6460 LV and NTEGRA Prima atomic force microscope (NT-MDT), respectively, were used. The chemical Impedance Analyzer HIOKI IM3590 was used to measure the electrical parameters of the portable microfluidic platform, and the experimental setup is shown in Figures 3. The electrical resistance was measured in the frequency range from 20 Hz to 200 kHz.

Figure 3. Experimental set-up for measuring electrical parameters of the proposed microfluidic platform

3 Results and discussion

3.1 SEM analysis

To discover internal structure of the analyzed papers before and after MWCNTs deposition, SEM analysis was conducted and micrographs are presented in Figure 4. Differences between morphological characteristics of papers No. 42, 44 and 45 with and without MWCNTs can be noticed and the nanocomposite formation were clearly indicated. Cellulose fibre structure of the paper can be clearly seen.

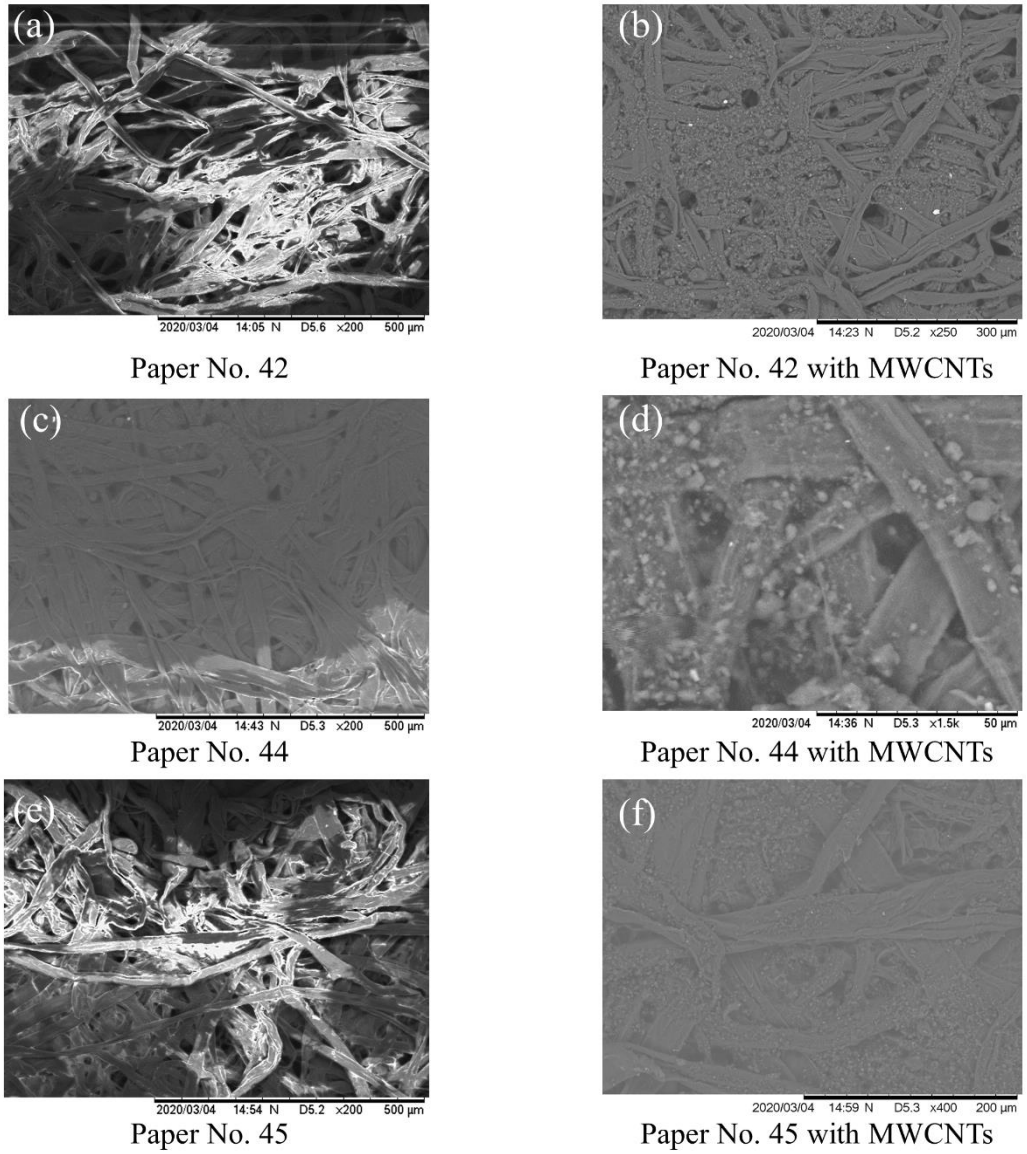

Figure 4. SEM micrographs of the filter papers No. 42, No. 44 and No. 45, before (a, c, e) and after deposition of MWCNTs (b, d, f)

3.2 AFM analysis

Atomic force microscopic analysis was carried out in air using intermittent-contact AFM mode with NT-MDT NSGO1 silicon cantilevers. The rectangular shaped cantilevers (125 µm × 5 µm × 0.5 µm) are fabricated of n-type, Antimony doped, single crystal silicon. Thin Au film was deposited on the reflective side of the cantilever. Nominal resonant frequency of these cantilevers is 150 kHz, while nominal force constant is 5.1 N/m. Image Analysis 2.2.0 (NT-MDT) software was implemented. Comparison of 3D images of the studied filter papers, with and without MWCNTs, was performed using NTEGRA Prima atomic force microscope and the results are presented in Figure 5. The root mean square roughness (R_q) was also calculated and the following results were obtained: (1) Paper No. 42 without MWCNTs, R_q = 492.6 nm, with MWCNTs, R_q = 515.9 nm; (2) Paper No. 44 without MWCNTs, R_q = 271.1 nm, with

MWCNTs, R_q = 544.4 nm; and (3) Paper No. 45 without MWCNTs, R_q = 339.6 nm, with MWCNTs, R_q = 433.0 nm.

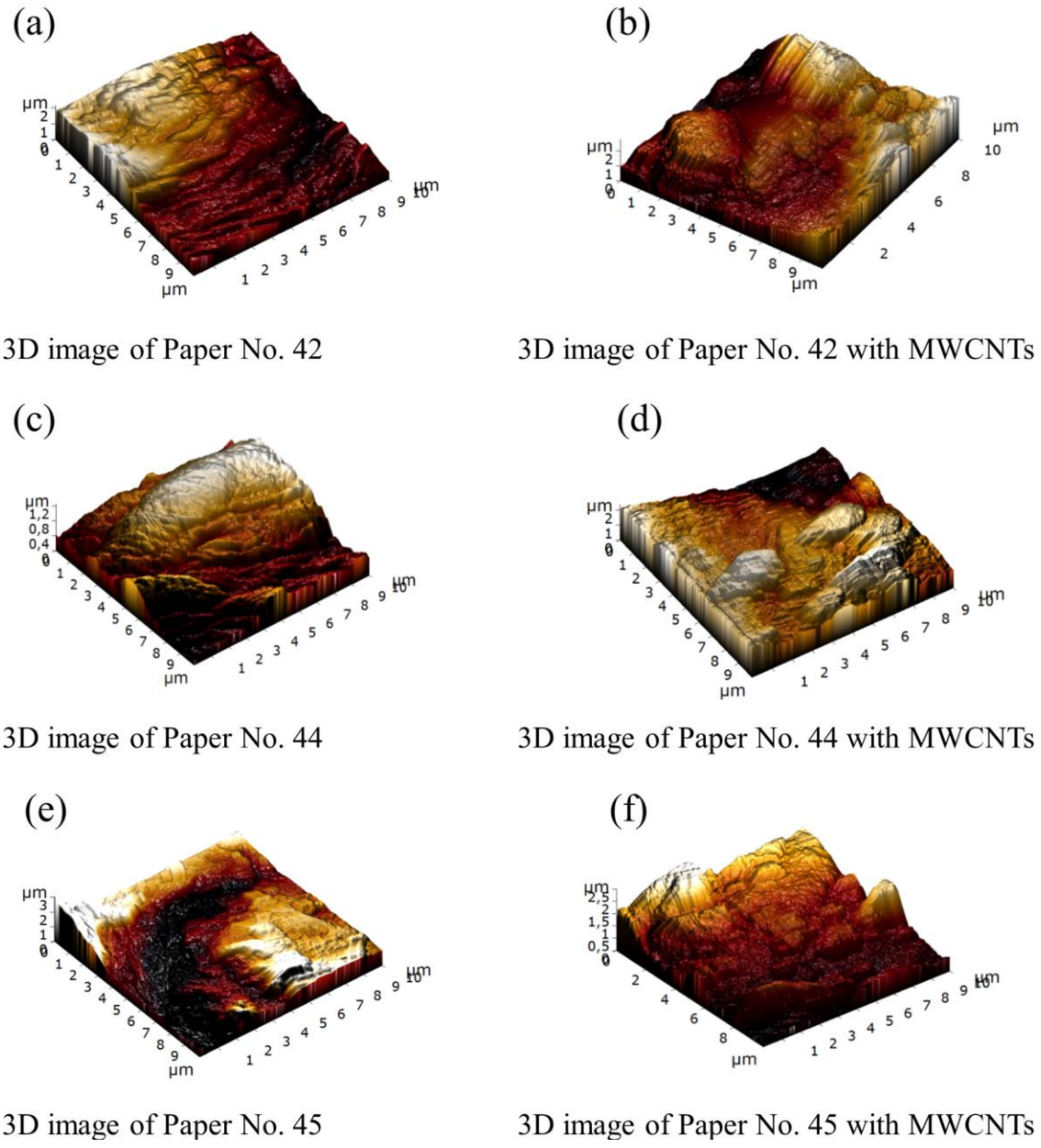

Figure 5. AFM 3D images of the filter papers No. 42, No. 44 and No. 45, before (a, c, e) and after deposition of MWCNTs (b, d, f)

3.3 Impedance spectroscopic analysis

Three types of studied filter papers are electrically non-conductive structures and their resistance were around 10^{10} Ω. In order to create conductive structure and to obtain resistance in MΩ-range these filter papers were functionalized by means of conductive MWCNTs. The rectangular pieces of filter papers

labelled with No. 42, No. 44 and No. 45 with MWCNTs were placed in the middle of the presented microfluidic platform. First, we measured the electrical resistance at two terminals of the structure without any fluid on the filter paper with MWCNTs and the obtained results are depicted in Figure 6. The filter paper No. 42 had the hugest pore sizes (20-25 nm), which created the space for entering MWCNTs in highest percentage, which led to the lowest electrical resistance. Contrary, paper No. 45 demonstrated the highest resistance due to the smallest pore sizes.

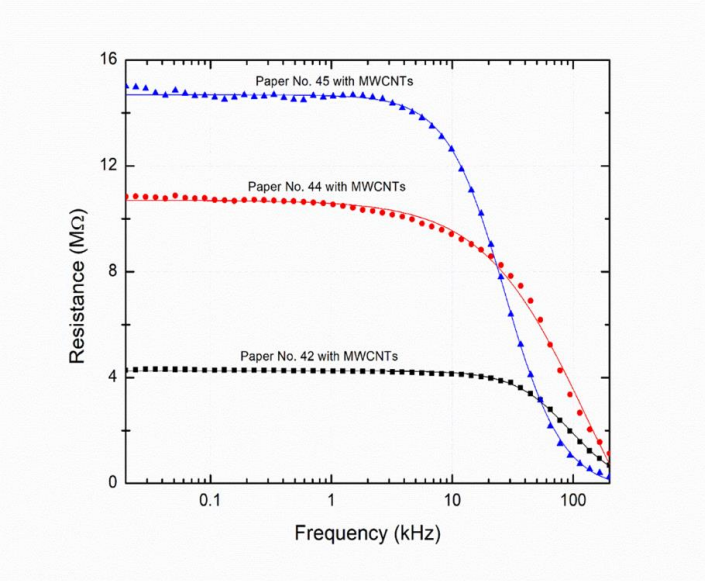

Figure 6. Measured resistance of three types of studied filter papers with deposited MWCNTs

To experimentally verify the developed platform, three different oil blends were prepared and dropped on the middle of the microfluidic platform, through the circular inlet (Figure 2), by single channel electronic pipette. The filter paper with MWCNTs was soaked with the oil blend. The electrical resistance was measured at the terminals of the microfluidic platform as a function of frequency for different oil blends and results are presented in Figure 7.

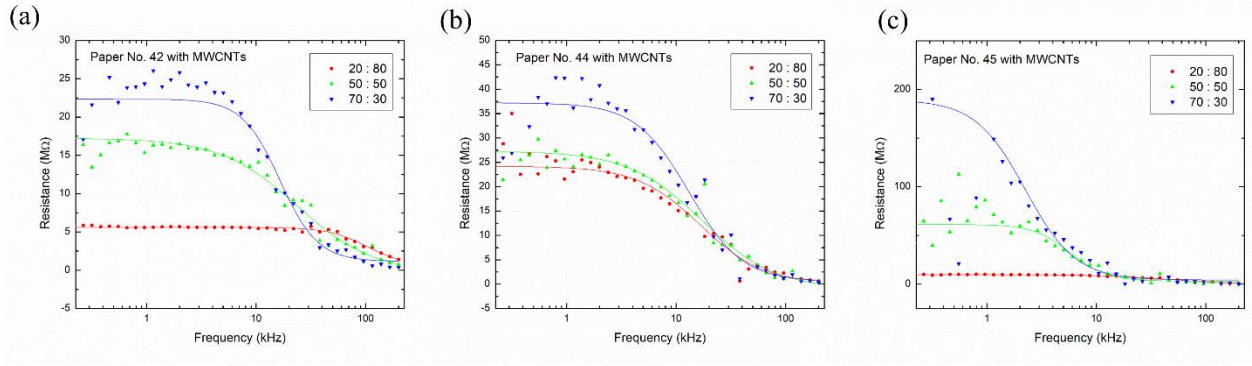

Figure 7. Measured resistance for three types of filter papers with MWCNTs and different oil blends

Figure 7(a) shows the measured resistance as a function of frequency for a microfluidic platform with filter paper No. 42 with MWCNTs. The oil blend with a ratio of 20:80 of olive and sunflower oil has the lowest resistance, which was around 6 MΩ at low frequency range. The oil blend with a ratio of 50:50 has a higher resistance and was about 16 MΩ, but with increasing frequency, this resistance decreases significantly. The highest resistance at frequencies up to 6 kHz has a blended oil with a ratio of 70:30 and it was around 22 MΩ. The same frequency dependence as well as MΩ range of measured impedance was

reported in reference (Dossi et al., 2016) as well as in reference (Prevc, Šegatin, Kralj, Ulrih, & Cigić, 2015) for resistance as a function of frequency for olive flesh and paste. Figure 7(b) displays the variation of resistance as a function of frequency for a microfluidic platform with filter paper No. 44 with MWCNTs. The same trend of resistance was obtained as for paper No. 42. Figure 7(c) depicts the frequency dependence of the resistance for different oil blends, for filter paper No. 45 with MWCNTs. Comparing the graphs in Figure 7, it can be concluded that for the same blended oil highest resistance demonstrated structure with paper No. 45 with MWCNTs, after that paper no. 44 and 42, following the same trend presented in Figure 6. When we compare different oil blends, the highest electrical resistance demonstrated blend with highest ration of olive oil, and this is the same behaviour for all type of analysed filter papers. This is the consequence of higher conductivity of HOSO than EVOO, itself, which can be attributed to higher presence of ions in the sunflower oil than in olive oil. These results are completely in accordance with the reported results in open literature. For example, the conductivity of vegetable oils was measured in (Corach, Sorichetti, & Romano, 2014) as a function of temperature and sunflower oil demonstrated higher conductivity than olive oil in the studied temperature range. It is concluded in reference (Rouabeh, M'barki, Hammami, Jallouli, & Driss, 2019), that sunflower oil is more conductive than olive oil (electrical conductivity for sunflower oil was $\sigma = 333 \cdot 10^{-3}$ S/m, whereas for olive oil $\sigma = 322 \cdot 10^{-3}$ S/m, for transformer applications). It was also reported in (Hassanain et al., 2017), that olive oil had almost the highest value of electrical resistivity comparing with nine other vegetable oil samples. Moreover, according to (Rouabeh, M'barki, Hammami, Jallouli, & Driss, 2019), sunflower oil has lower viscosity than olive oil and decrease in the viscosity can facilitate current flow in oil. Measurement of electrical conductivity or resistivity is important as one of the indicators used in production plants to detect oil contaminants (Corach, Sorichetti, & Romano, 2015) as well as a method for determination of the maturity state of olives (Justicia et al., 2017).

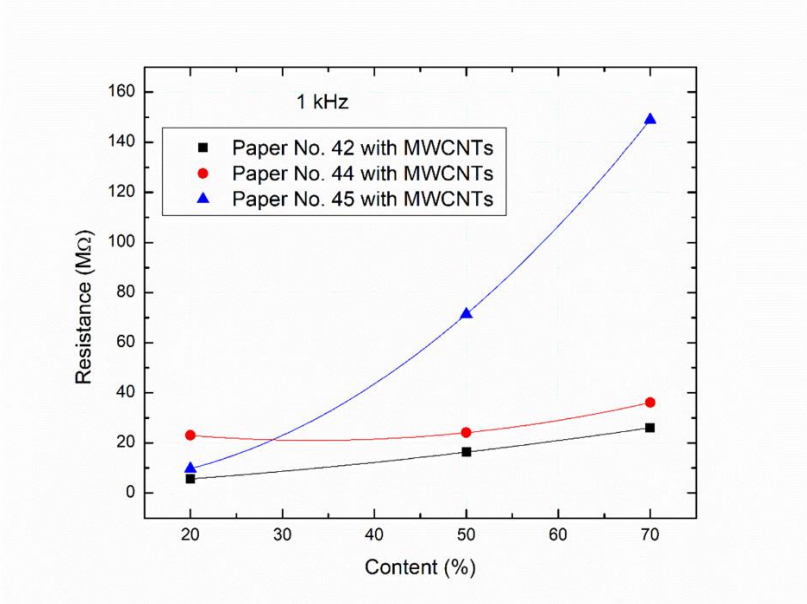

Figure 8. The relationship between the resistance and olive oil content in oil blends at a frequency of 1 kHz, for three filter paper types: No. 42, No. 44 and No. 45 with MWCNTs

Figure 8 shows electrical resistance as a function of the olive oil content in oil blends at a frequency of 1 kHz, using three filter paper types: No. 42, No. 44 and No. 45, with MWCNTs. It can be concluded that the smallest change in resistance was observed using microfluidic platform with filter paper type No. 42 while the largest variation in resistance was exhibited when incorporating filter paper No. 45 in the platform. To

analyse sensing performances of the proposed platform, the sensitivity (defined as $S = \Delta R / \Delta C$, where ΔR is variation of electrical resistance and ΔC changes in olive oil content in %) and linearity (R^2 was taken as a measure of goodness of linear fit) were calculated and presented in Table 2.

Table 2. Sensing characteristics of the microfluidic platform

Filter type	Parameters	
	Sensitivity (S) [MΩ/%]	Linearity (R^2)
Paper No. 42 with MWCNTs	0.41	0.9924
Paper No. 44 with MWCNTs	0.26	0.7128
Paper No. 45 with MWCNTs	2.79	0.9676

As can be seen from Table 2, paper No. 44 with MWCNTs demonstrated the low sensitivity and linearity, whereas the optimal sensing characteristics had the structure with paper No. 45 with MWCNTs with sensitivity 2.79 MΩ per % of the olive oil content in oil blends and with linearity index $R^2 = 0.9676$. This can be attributed to the lowest root mean square roughness of the paper No. 45 with MWCNTs, which can be seen in AFM images comparing the other papers and because of that the oil blends can be evenly distributed through the surface of this paper and good variation of the resistance can be obtained. Thus, it can be concluded that paper No. 45 is the best choice for detection of olive oil content in oil blends using the proposed platform.

The described sensing platform is compact, physically flexible and can be integrated with other electronic components at the circuit board. In order to demonstrate proof of the concept, the complete electronic device was developed, having this platform as an input component, representing actually a variable resistor. The developed prototype consisted of an embedded hardware (Arduino Leonardo), a breadboard with resistor of known value and developed platform (sensing element) which was used as a structure for oil blend injection on the top (Figure 9(a)). These components were connected by wires in a voltage divider form. The sensing element was connected directly to 5V supply through red wire and on the other side it is connected to resistor by blue wire. The same junction spot is connected to analogue input of Arduino Leonardo (A0), which was used for the input voltage measurement. The other side of the resistor was connected directly to the ground (GND wire). Both, 5V and GND were supplied by Arduino outputs. Algorithm for the input signal to voltage conversion and proportional resistance value of sensor was done within programming code. USB was used for power supply as well as for the serial monitor via Arduino IDE. On a small OLED display (Figure 9(b)) the measured value of electrical resistance and percentage of olive oil content in the fluid blend can be shown (based on the look-up tables thanks to calibration curves presented in Figure 8).

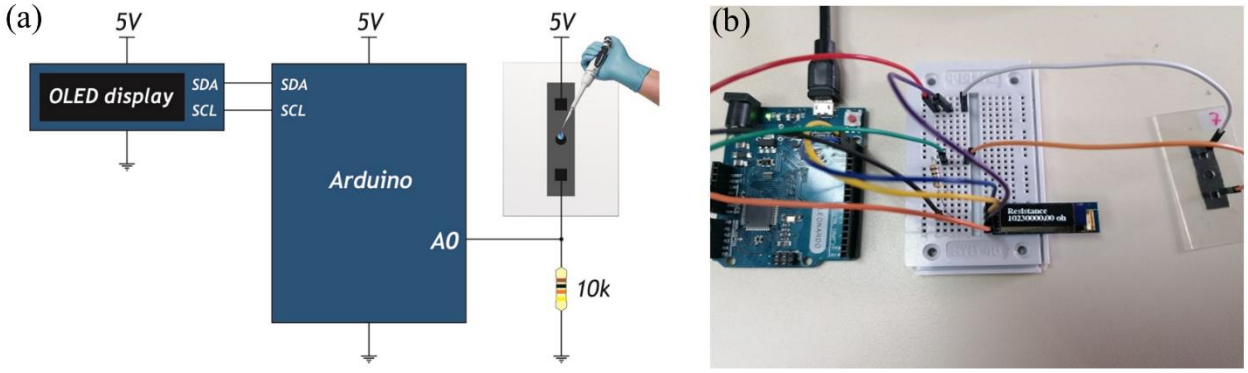

Figure 9. The electronic device for fast measurement of electrical resistance and percentage of olive oil content in oil blends based on proposed microfluidic platform, (a) schematic of electrical circuit, (b) manufactured prototype

With the presented stand-alone and low-cost electronic system, it is possible to detect content of olive oil in oil blends on-site, reducing time and costs of necessary analysis in quality control phases in food industry.

4 Conclusion

This study presented the application of filter paper functionalized with MWCNTs as a central element of the microfluidic platform for detection of olive oil content in oil blends. The proposed microfluidic platform with three different types of filter paper can be used to detect percentage of olive oil in a blend of olive and high-oleic sunflower oil, through measuring electrical resistance at the outer terminals of the platform. SEM and AFM analysis was used for experimental determination of performances of three types of tested filter paper samples with and without MWCNTs. The microfluidic platform with filter paper type No. 42 with MWCNTs demonstrated the lowest resistance, while the microfluidic platform with filter paper No. 45 with MWCNTs had the highest variation of electrical resistance, the highest numerical sensitivity and very good linearity. The proposed platform is compact and lightweight and thanks to these properties very suitable for application in portable and hand-held devices. The prototype of electronic device was created for fast and straightforward determination of the electrical resistance as well as displaying the percentage of olive oil content in the oil blends, based on the developed microfluidic platform. This device has a great potential for application as a sensing element for estimation of percentage of olive oil in the blend with other oil types, which is useful in food industry and quality control of the products.

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