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# Silicon-Based Optoelectronic Tongue for Label-Free and Nonspecific Recognition of Vegetable Oils

Bohdan V. Oliinyk, Karyna Isaieva, Anton I. Manilov,\* Tetyana Nychporuk, Alain Geloën, Florent Joffre, Valeriy A. Skryshevsky, Sergii V. Litvinenko, and Vladimir Lysenko



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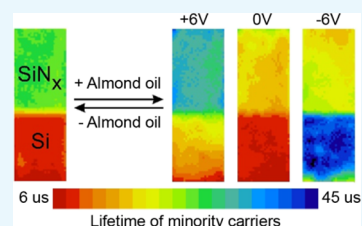


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Supporting Information

**ABSTRACT:** A special electronic tongue system based on photoelectric measurements on Si–Si/SiN<sub>x</sub> sensitive structures is reported. The sensing approach is based on measuring of minority carrier lifetime in silicon-based substrates using microwave-detected photoconductance decay. This inexpensive and environmentally friendly combinatorial electronic sensing platform is able to create characteristic electronic fingerprints of liquids, detect, and recognize them. In particular, an application of the optoelectronic tongue for recognition of vegetable oils and their mixtures is described.



## INTRODUCTION

Development and improvement of analytical methods for assuring quality and authenticity of vegetable oils are very important research axes in the food field. In particular, the objectives of empowering detection and fostering prevention of oil fraud are considerably supported by European Union's research and innovation programs.<sup>1</sup> On the basis of the expanding market for vegetable oils, their authenticity has become an important subject from both commercial and health point of view.<sup>2,3</sup> Among all vegetable oils, the extra virgin olive oil is known to have the highest economical value.<sup>4</sup> At the same time, rapeseed, soyabean, and palm oils are the cheapest ones available in the market and can be used to 'bulk-out' more expensive products. Thus, the analytic methods resolving the problem of oil adulteration as well as determining origin, quality, and oxidative stability of oils are continuously under intensive development.

The main instrumental approaches of oil analysis include different methods of chromatography and mass spectrometry, as well as their combination, such as: high-resolution gas chromatography, high-performance liquid chromatography, supercritical fluid chromatography, stable isotope ratio mass spectrometry, chemical ionization mass spectrometry, and so forth.<sup>5–7</sup> In addition, nuclear magnetic resonance spectroscopy,<sup>8</sup> differential scanning calorimetry,<sup>9</sup> Fourier transform infrared spectroscopy, Raman spectroscopy,<sup>10–12</sup> fluorescence spectroscopy,<sup>13,14</sup> and DNA-analysis instruments<sup>12,15</sup> are also widely applied for investigation of vegetable oils.

Most of the conventional methods cited above consist of time-consuming, expensive, and complex set-ups and technological procedures. Some of them use toxic chemicals and solvents. Therefore, development of innovative, cheap, and efficient sensor-based concepts is of primary importance.<sup>16</sup> In order to improve efficiency of the oil detection, novel sensing components are investigated, for instance, microwave sensing

interdigitated electrodes<sup>17</sup> or a fiber optic long period grating-based sensor.<sup>18</sup> Nevertheless, the most promising results are obtained with multisensor platforms exploiting the concept of electronic nose and tongue. For example, electronic noses based on metal oxide semiconductors,<sup>19–21</sup> conducting polymers,<sup>22–24</sup> quartz crystal microbalances,<sup>25,26</sup> and detectors of surface acoustic wave<sup>27,28</sup> were applied for differentiation of vegetable oils. As for the electronic tongues, they are mainly based on voltammetric measurements in electrochemical cells<sup>29–32</sup> or potentiometric sensor arrays.<sup>33–37</sup>

Our research team has recently developed a special electronic tongue system based on optoelectronic measurements with use of silicon wafers.<sup>38</sup> In particular, we have shown its successful application for the recognition of water and spirits. This kind of electronic tongue is an inexpensive and environmental friendly combinatorial electronic sensing platform that is able to record characteristic electronic fingerprints of liquids resulted from their interaction with either naturally aged or specifically treated silicon substrates. In this letter, we report an application of the silicon-based optoelectronic tongue for recognition of vegetable oils and their mixtures.

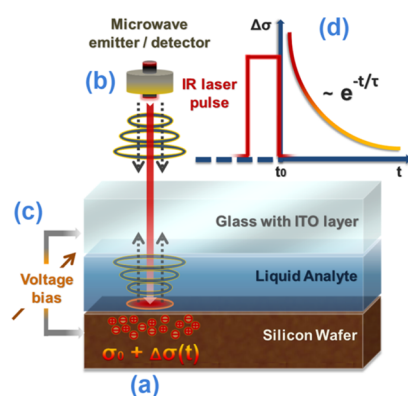
## RESULTS AND DISCUSSION

Our sensing approach is based on measuring of minority carrier lifetime in silicon by using a microwave-detected photoconductance decay as shown in Figure 1. A pulse of infrared (IR) laser (904 nm) generates free electron–hole

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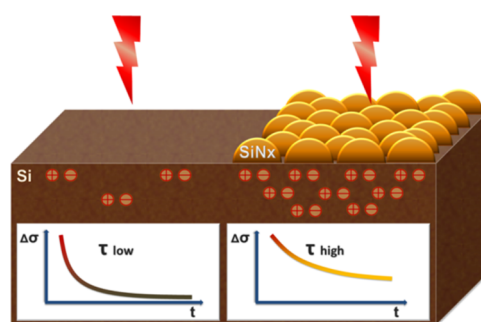


**Figure 1.** Basic physical principles used in the optoelectronic tongue: (a) generation of free electron–hole pairs near the silicon surface under absorption of IR laser pulse; (b) dependence of microwave reflectivity on concentration of the free charge carriers; (c) modulation of surface band bending by analyte–silicon interaction and applied voltage bias; (d) time-resolved photoconductivity decay due to recombination of the generated electron–hole pairs as a function of surface band bending.

pairs under illuminated area close to the front surface of a silicon wafer (penetration depth of the laser radiation in silicon is about 30  $\mu\text{m}$ ). Because the free electrons and holes recombine, their concentration and, consequently, conductivity of the silicon sample decreases exponentially in time after the action of the exciting light pulse. The decaying conductivity can be monitored by detecting microwave reflectivity ensured by the photogenerated free charge carriers as a function of time. The measured reflectivity decay is fitted with an exponential curve, and the obtained time constant ( $\tau_{\text{meas}}$ ) corresponds to the effective lifetime of the photocreated charge carriers in the given position of the silicon sample. Changing the illuminated zone of the studied sample allows the creation of its 2D map in terms of the lifetime values  $\tau_{\text{meas}}$ , which depend on silicon parameters and its surface chemistry. If a silicon sample is put in an intimate contact with a chemical substance (e.g., liquid), a new  $\tau_{\text{meas}}$  map induced by the substance molecules interacting electronically with the silicon surface will be obtained. Thus, one can establish a correlation between a surface distribution of the  $\tau_{\text{meas}}$  values obtained for the given substance/silicon interface and chemical composition of the liquid substance.

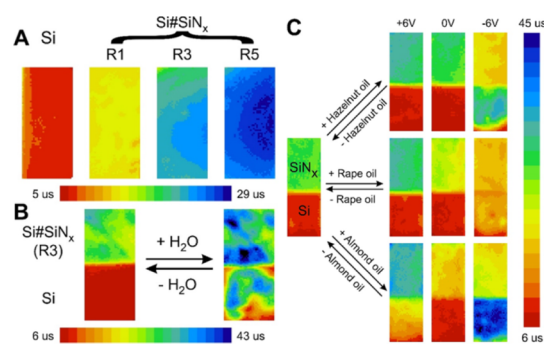
To increase reliability of liquid recognition, one can additionally record a set of  $\tau_{\text{meas}}$  maps by polarizing the liquid/silicon interface as it is schematically shown in Figure 1. Indeed, continuous tuning of the difference of electric potentials  $V_b$  applied to the interface will ensure modulation of the electronic bands bending in the near-surface region of the silicon substrate. The voltage dependent band bending leads to refilling of interface electronic states and significant voltage-induced modulation of the lifetime maps formed by the studied liquid sample onto the silicon surface.

A typical Si-based structure used in this work is schematically represented in Figure 2. A half of the structure was covered with an ultrathin  $\text{SiN}_x$  layer (10–15 nm). The  $\text{SiN}_x$  films are well-known to be widely used in the industrial silicon solar cell fabrication process.<sup>39</sup> Indeed, chemical composition of the  $\text{SiN}_x$  layers ensures an excellent electronic passivation of various defects localized at the surface of bulk silicon substrates.<sup>40</sup> As a result, lifetime of charge carriers photo-generated in Si substrates covered with a  $\text{SiN}_x$  layer increases



**Figure 2.** Typical Si–Si/ $\text{SiN}_x$  sensitive structure used in this work. A half of the structure is covered with an ultrathin (non continuous)  $\text{SiN}_x$  layer passivating the silicon surface and characterized by increased lifetimes of photogenerated charge carriers.

significantly. One can compare 2D maps of the  $\tau_{\text{meas}}$  values obtained for the Si samples covered with  $\text{SiN}_x$  layers deposited at various silane-to-ammonia gas flow ratios  $R = \text{SiH}_4/\text{NH}_3$ . As shown in Figure 3A, the higher the  $R$  value is, the higher the



**Figure 3.** (A) Characteristic lifetimes of a bare Si substrate and Si substrates covered by  $\text{SiN}_x$  films obtained at various silane-to-ammonia gas flow ratios ( $R$ ). (B) Impact of water molecules on the 2D  $\tau_{\text{meas}}$  map of a Si–Si/ $\text{SiN}_x$  substrate. (C) 2D  $\tau_{\text{meas}}$  maps of a Si–Si/ $\text{SiN}_x$  substrate put in contact with almond, rape, and hazelnut oils at different polarizing voltages  $V_b$ .

corresponding lifetimes ( $\tau_{\text{meas}}$ ) values are. In particular, the  $\tau_{\text{meas}}$  value (about 30  $\mu\text{s}$ ) obtained for the Si-rich  $\text{SiN}_x$  layers deposited at R5 is much higher than the  $\tau_{\text{meas}}$  value for a bare Si surface (5  $\mu\text{s}$ ), for example.

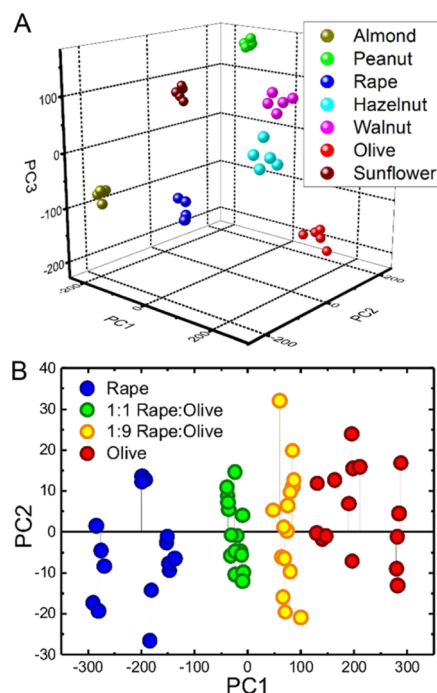
In our work, extremely low deposition time (about 10–20 s) of the  $\text{SiN}_x$  layers was used in order to avoid complete capping of the silicon surface by a continuous film of silicon nitride (as sketched in Figure 2). This is especially important to allow the studied liquids to get at least partial access to the Si surface semipassivated by the  $\text{SiN}_x$  films. Indeed, being able to penetrate through the numerous subnanometer pores, grooves and/or gaps present in the  $\text{SiN}_x$  ultrathin layers, the studied liquid will modulate significantly the initial lifetime values measured in air. As one can see in Figure 3B, water molecules influence strongly the  $\tau_{\text{meas}}$  map of a Si sample having two regions: (i) the bare Si surface and (ii) the  $\text{SiN}_x$  ultrathin layer deposited on the Si substrate at R3. In general, the lifetime values are considerably enhanced for the both regions exposed to water. After water removal, the 2D distribution of the  $\tau_{\text{meas}}$  values was completely reset.

To demonstrate high applicative potential of the optoelectronic tongue for the oil recognition purpose, a set of various oil samples has been used. The most representative 2D  $\tau_{\text{meas}}$

maps of the photoelectric fingerprints created by almond, rape, and hazelnut oils on the Si–Si/SiN<sub>x</sub> sensitive structures at 0 and ±6 V of the voltage bias  $V_b$  are shown in Figure 3C. Here, positive sign of the voltage corresponds to accumulative band bending of the p-type silicon wafer. As one can see, the most important difference between the  $\tau_{\text{meas}}$  maps formed by the studied oils corresponds to  $V_b = -6$  V. Thus, one could use this  $V_b$  value for rapid qualitative recognition of the tested oils. At this value of the polarizing voltage (−6 V), the most pronounced electronically passivating impact on the silicon surface is provoked by almond oil, while distribution of the  $\tau_{\text{meas}}$  values (17–22  $\mu\text{s}$ ) in the Si/SiN<sub>x</sub> region remains almost insensitive to the oil samples. In particular, almost no difference between the  $\tau_{\text{meas}}$  values for both Si and Si/SiN<sub>x</sub> regions can be observed for the case of the rape oil polarized at  $V_b = -6$  V. As for the inhomogeneity of lifetime distribution over the sample surface, which was observed for water, is almost negligible for the all the studied oil samples. Indeed, a slight distribution of the lifetimes within a given zone is much less than the changes of the lifetimes for various oils exposed to various voltages.

At the same time, the most significant voltage-induced evolution of the recorded  $\tau_{\text{meas}}$  maps corresponds to the interface between the Si–Si/SiN<sub>x</sub> substrates and almond oil. The evolution kinetics is extremely slow. Once a voltage value is set, the corresponding voltage-induced changes of a  $\tau_{\text{meas}}$  map can take up to 30 min. Finally, it is worth to note complete reversibility of the initial 2D  $\tau_{\text{meas}}$  maps in air after the washing out of the oil samples. Indeed, as one can see in Figure S1 (shown in the Supporting Information), the  $\tau_{\text{meas}}$  maps recorded in air after each exposition of the Si–Si/SiN<sub>x</sub> substrate to an oil sample are identical and correspond to the map of the original substrate before its first use.

To ensure a high discrimination capability level of the proposed sensing system, the data can be visualized in more illustratable manner. In our work, principal component analysis (PCA) method was applied.<sup>41</sup> This multivariate analysis approach enables to find a set of linearly uncorrelated variables so that the first has maximum variance, and each next variable is orthogonal to the previous and has maximum possible variance with this constraint. This method is widely used for dimension reduction because most of the information is concentrated in the first components and commonly employed in various combinatorial platforms for discrimination purposes.<sup>42–44</sup> As a result, Figure 4A shows a plot of three first principal components (with corresponding scores of 26.8% for the first principal component, 22.6% for the second principal component, and 14.7% for the third one) containing clouds of points for each of the tested oils. In particular, because the distance between the points reflects their nonsimilarity, clear clustering of the data illustrates a perfect degree of discrimination capability of our analytical system to recognize among the studied oils. With only two kinds of regions (bare Si and Si covered with SiN<sub>x</sub> islands) and three values of the polarizing voltage  $V_b$  (0 and ±6 V), we are already able to observe a significant degree of discrimination between the different oils. Natural small variation of the lifetimes within a single zone is taken into account and reflected by distance between the points at the PCA diagram in Figure 4A for each oil sample. Because of the distance between the points within the same cloud is less than the distances between the clouds, the recognition of the oil samples can be evaluated as a reliable one.



**Figure 4.** (A) 3D PCA plot corresponding to the electronic fingerprints of the tested oils formed on the Si–Si/SiN<sub>x</sub> substrate at different polarizing voltages  $V_b$ . (B) 2D PCA plot corresponding to the electronic fingerprints of olive and rape oils as well as their mixtures. The units of the principal components on the axes are microseconds.

Virgin olive oil possessing unique qualities is highly appreciated by consumers because of its exceptional nutritional, medicinal, cosmetic, and even ceremonial value. As a result, it is one of the most expensive and also one of the most adulterated food products. In particular, its adulteration with low-grade olive oils or cheaper vegetable oils (e.g., such as rape oil) could potentially be very profitable for sellers or raw material suppliers and may yield large economic profits. Thus, authentication of virgin olive oils has become an interesting subject from both commercial and health perspectives. Figure 4B demonstrates the ability of the optoelectronic tongue based on the Si–Si/SiN<sub>x</sub> sensitive structures described in this letter to recognize mixtures of olive and rape oils. Indeed, Figure 4B shows a plot of two first-principal components (with corresponding scores of 67.7% for the first principal component and 16.6% for the second principal component) containing points for initial pure olive and rape oils as well as their mixtures (all samples measured three times). The first mixture (1:9) was selected as a minimal (from commercial view point) amount of rapeseed added to olive oil. The second mixture (1:1) is intermediate between the pure rapeseed and olive oils. As one can see, the cloud corresponding to the olive oil with 10% of rape oil is clearly different from the cloud related to the sample of the pure olive oil. It is worth noting that the cloud corresponding to the 50% mixture of the oil samples is localized on the PCA plot in the middle between the clouds corresponding to the pure oils.

The described approach is based on a holistic measurement approach. Indeed, a complex and simultaneous impact of all oil components forms the sensor signal in each zone. In other words, all the molecules constituting the given oil physically impact the electronic surface states initially governed by the

surface chemistry of each zone (bare c-Si or SiN<sub>x</sub> film) and by bias voltage. Unlike other known sensing systems, specific sensitive materials and components are not necessary to be used. Moreover, certain chemical indicators are not determined, such as peroxide value, concentration of additives, etc. Even so, our optoelectronic tongue is able to distinguish different classes of oils and their mixtures using PCA.

## CONCLUSIONS

In conclusion, the optoelectronic tongue based on the Si–Si/SiN<sub>x</sub> substrates can be efficiently applied for a label-free and nonspecific recognition of vegetable oils. In particular, the lifetime of photogenerated charge carriers is confirmed to be an extremely efficient physical parameter ensuring high sensitivity of silicon-based structures to play a role of an electronic screen reflecting the complex physicochemical interaction between the bare or partially covered silicon surface and the studied chemical substance. This specific kind of electronic tongues is an inexpensive and environmental friendly combinatorial electronic-sensing platform that is able to create characteristic electronic fingerprints of liquids, detect, and recognize them.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.9b03196>.

Fabrication of the Si–Si/SiN<sub>x</sub> samples, cleaning of the Si–Si/SiN<sub>x</sub> samples, and mathematical treatment of the obtained data (PDF)

## AUTHOR INFORMATION

### Corresponding Author

**Anton I. Manilov** – Institute of High Technologies, Taras Shevchenko National University of Kyiv, 01033 Kyiv, Ukraine; Corporation Science Park Taras Shevchenko University of Kyiv, 01033 Kyiv, Ukraine; [orcid.org/0000-0003-2225-2754](https://orcid.org/0000-0003-2225-2754); Email: [anmanilov@univ.kiev.ua](mailto:anmanilov@univ.kiev.ua)

### Authors

**Bohdan V. Oliinyk** – Institute of Analytical Sciences (ISA), UMR CNRS 5280, UCBL, University of Lyon, 69100 Villeurbanne, France; OlisensTech, 69006 Lyon, France; [orcid.org/0000-0002-1895-3980](https://orcid.org/0000-0002-1895-3980)

**Karyna Isaieva** – IADI, Université de Lorraine, INSERM U1254, Nancy F-54000, France

**Tetyana Nychporuk** – Nanotechnology Institute of Lyon (INL), UMR CNRS 5270, INSA de Lyon, University of Lyon, 69621 Lyon, France

**Alain Geloën** – CarMeN Laboratory, INRA UMR1397, INSERM U1060, INSA de Lyon, IMBL, University of Lyon, 69621 Lyon, France

**Florent Joffre** – Institut des Corps Gras (ITERG), 33610 Canéjan, France

**Valeriy A. Skryshevsky** – Institute of High Technologies, Taras Shevchenko National University of Kyiv, 01033 Kyiv, Ukraine; Corporation Science Park Taras Shevchenko University of Kyiv, 01033 Kyiv, Ukraine

**Sergii V. Litvinenko** – Institute of High Technologies, Taras Shevchenko National University of Kyiv, 01033 Kyiv, Ukraine; Corporation Science Park Taras Shevchenko University of Kyiv, 01033 Kyiv, Ukraine

**Vladimir Lysenko** – Light-Matter Institute (ILM), UMR CNRS 5306, University of Lyon (UCBL), 69622 Lyon, France

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acsomega.9b03196>

## Notes

The authors declare no competing financial interest.

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