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Portable Nanofiber-Light Addressable Potentiometric Sensor for Rapid *Escherichia coli* Detection in Orange Juice

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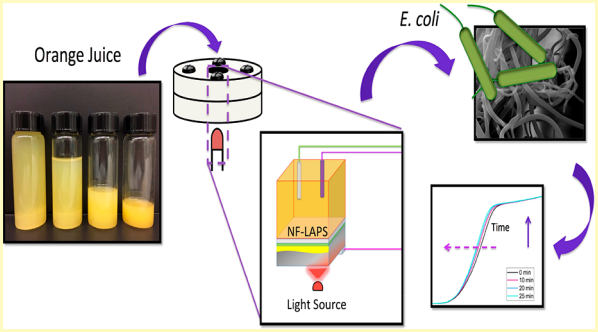
Supporting Information

ABSTRACT: The growing need to prevent pathogen outbreaks is irrefutable in the case of the food industry. Early detection in products, especially beverages, contaminated with bacterial strains is vital to avoid infected foods from reaching the consumer. If *E. coli* is pesent in such foods, it can cause infections. It can also be an indicator of the existence of other harmful coliforms. In this study, we have investigated the detection of *Escherichia coli* (*E. coli*) in orange juice using a portable nanofiber-light addressable potentiometric sensor (NF-LAPS). We have chosen electrospun pH-sensitive poly(vinyl alcohol)/poly(acrylic acid) (PVA/PAA) hydrogel NFs as the sensitive layer. The successful detection of *E. coli* was reported with the NF-LAPS in less than 1 h. The limit of detection (LOD) measured in the sensor is found to be10^2 CFU/mL. We have confirmed the selectivity of the biosensor toward *E. coli* by examining the response of the NF-LAPS against *Salmonella typhimurium* (*S. typhi*), also commonly found in orange juice. Despite the complex nature of orange juice, the response of the biosensor is in no way affected while orange juice is tested as is.



KEYWORDS: *potentiometric sensor, portable sensor, pH sensitive nanofiber, E. coli, orange juice, food safety*

Various reports of outbreaks linked to *Escherichia coli* (*E. coli*) from different food products have recently surfaced as a result of either minimally processed products or improper practices.[1−3] *E. coli*, as one of the leading causes of foodborne diseases, poses a serious danger to human health in developed countries.[4] Specific strains of *E. coli* can cause infections including diarrhea, bloody diarrhea, hemorrhagic colitis, or even kidney failure due to the production of Shiga toxin.[5] Among all fruit juices, orange juice is the most widely consumed beverage and demands high priority in the case of pathogen contamination prevention. Interestingly, acidic fruit juices were not considered as hosts for bacterial life until recent years.[6] However, the occurrence of outbreaks has validated the possibility of pathogen growth in such beverages. Accordingly, the United States Food and Drug Administration (FDA) has imposed a number of treatments, inactivation processes, and chemical sanitizations to ensure the safety of fruit juices available for consumption.[7]

During various stages of production, processing and disinfection treatment, it is imperative that the quality of the beverage be monitored for possible infection with pathogens such as *E. coli*. Traditional detection techniques for *E. coli* consist of plating and colony counting methods, polymerase chain reaction (PCR), and enzyme-linked immunosorbent assay (ELISA). The merits of these methods cannot be denied as they are reliable, accurate, and repeatable. On the other hand, these techniques require time-consuming measurements, intensive training of highly qualified personnel, and separate laboratory facilities. In the food industry, where products are manufactured, packaged or distributed in a short period, there is a critical demand for methods that require shorter detection time and are available on-site.

Recent developments in biosensing have aided the scientific community to invent sensitive and selective techniques that are suitable candidates to replace conventional detection tools. Examples of such methods are optical biosensors like surface plasmon resonance (SPR), chromatography, electronic sensors,[8] and microelectromechanical sensors (MEMS) such as microcantilevers.[9] The downside of these methods is their complexity and mostly slow responses. The typical use of antibodies in a variety of these tools renders their industrial application limited due to the short shelf time of the antibodies. Other shortcomings of these methods include cross-reactivity of the biological elements, a narrow operational pH range for

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antibodies, no capability to distinguish between live and dead cells, and lack of robustness.[9−12]

Electrochemical sensors offer a number of advantages over the above-mentioned techniques that make them the most suitable candidate for the production of benchtop sensing devices. First, the sensing mechanism is simpler in electrochemical sensors, and its simplicity does not compromise its level of sensitivity. Second, electrochemical platforms have the capacity to be miniaturized, and thus the instrumentation is easily implemented. Furthermore, electrochemical sensors have the capability to be integrated with optical methods to form photoelectrochemical sensors to achieve synergistic optical and electrochemical detection abilities.[13]

The light addressable potentiometric sensor (LAPS) is the most well-known photoelectrochemical sensor used in biological applications.[14−19] The feasibility of packaging LAPS into a portable device has led to the development of compact LAPS sensors for primarily electrochemical sensing.[20,21] A conventional LAPS device utilizes a working electrode (WE) that is considered the sensing heart of the system. This WE has a semiconductor base commonly made of Si. When light illuminates the sensor from the front side or the back in a modulated manner, electron−hole pairs are separated in the semiconductor substrate. Illumination with a designated frequency enables a sustainable photocurrent in the sensor. The use of light makes LAPS more sensitive as the location of the light governs the position of the generated photocurrent. Light can therefore be programmed to provide spatial information about changes on the surface of the sensor. LAPS is based on an electrolyte-insulator-semiconductor system. Depending on the electrical potential applied to the sensor, the charges might accumulate or deplete on a layer directly underneath the interface between the insulator and semiconductor base of the WE. This layer is known as the depletion layer when the applied voltage is higher than the flat band potential of the semiconductor (for p-type semiconductors). In a state of depletion, the width of the depletion layer is determined by the applied potential and the potential on the surface of the sensitive layer in the LAPS system. The potential on the surface is, in turn, dependent on the charges of the double layer of the electrolyte as explained in our previous work.[22] The pH of the solution is known to influence the amount of charges on the surface of the LAPS sensor and therefore affects the generated photocurrent. Any change in pH, i.e., the change in the concentration of H^+, is reflected by a shift in the photocurrent associated with the width of the depletion layer.[23]

The use of electrospun pH responsive hydrogel nanofibers (NFs) has enhanced the sensing properties of LAPS as reported earlier.[22] The nanometer size of the hydrogel NFs accelerates the swelling compared to their bulk form so that response time is shortest when hydrogel dimensions are smallest.[24] The physical swelling and deswelling of the NFs reflect the changes in medium pH and ultimately influence the LAPS photocurrent signal. Moreover, the increased surface area of the NFs amplifies the active sites available for interaction with biological agents when the NFs are functionalized for specific targets.

In this study, the application of a portable NF-LAPS system was reported to selectively detect *E. coli* in orange juice in less than 1 h. To the best of our knowledge, no such system has been developed that integrates hydrogel NFs with LAPS for biosensing in food safety applications. The sensing mechanism is based on the metabolic activity of *E. coli* toward sugar molecules and the production of acidic products such as lactates and acetates. By functionalizing the surface of the NFs with D-mannose, the need for antibody−antigen complex interactions which reduce sensor reliability over time is eliminated. A search in the literature reveals that the selectivity for nonfermenting bacteria has already been reported for NF-LAPS sensors.[22] In the present work, the selectivity of such sensors toward *E. coli*, *Salmonella typhimurium* (*S. typhi*), as a sugar fermenting bacteria, was investigated. *S. typhi* is a multiorgan pathogen that can be found in foods and fruit juices.[25,26] The results showed that the sensitivity of the sensor was not hindered by the presence of the opaque analyte with a variety of molecules in type and concentration. One of the advantages of the developed sensor was that the overall photocurrent was not interrupted by the opaque samples since the light was illuminated from the backside of the device instead of the front side. Furthermore, the developed photoelectrochemical sensor offers a compact and portable device for on-site biosensing applications. The portable nature of the device will potentially be of great value where detection is needed in the field where assuring food safety is paramount and time is of the essence.

MATERIALS AND METHODS

Chemical Reagents and Solutions. All chemicals including PVA, PAA, HCl, NaOH, phosphate buffer powder, and D-mannose were purchased from Sigma-Aldrich and used without further purification. Commercially pasteurized orange juice was purchased without additional preservatives and stored at 4 °C before use. A calibration curve for different pH values was obtained using phosphate buffer saline (PBS) solutions with HCl and NaOH for pH adjustments.

NF-LAPS Sensing Chips. PAA/PVA pH sensitive hydrogel NFs were fabricated using a homemade electrospinning setup as described in our previous work.[22] Si substrates of a p-type nature with low levels of B doping (resistivity of <100 Ω.cm) and a thickness of 525 ± 25 μm were used as the sensor chip substrates. The natural SiO_2 layer on the Si substrates was kept intact to ensure the insulation on the surface. After electrospinning, all sensor chips were annealed at 145 °C for 30 min before surface functionalization. The surface of the NF sensitive layer was functionalized with D-mannose according to previously established procedures.[22] The NFs were subsequently characterized by scanning electron microscopy (SEM VEGA-3, Tescan). The success of the functionalization and swelling characteristics were verified in our earlier study.[22]

Portable NF-LAPS Device. The NF-LAPS sensor chips were placed inside a 3D printed chamber. The back of the chamber was illuminated using a red LED with a wavelength of 635 nm and a frequency of 12 kHz. The WE was connected to a compact EMStat3 PalmSens potentiostat. DC voltage was applied through the potentiostat with 2 mV steps and a speed of 40 mV/s. The generated photocurrent is an AC current in nature and varies with respect to time. However, the potentiostat displayed the rectified values of the current at any given applied voltage since measurements were carried out in a linear sweep voltammetry (LSV) method. This means that every signal point showed a photocurrent value with respect to the voltage and not with respect to time. The measured current is sampled at the second half of each potential step. $I-V$ curve readouts were displayed on a generic tablet connected to the potentiostat. EMStat3 uses the PStouch application for measurements and analysis.

Bacterial Strain Preparation. *E. coli* strain ATCC 25922 and *S. typhi* strain ATCC 19585 from a −80 °C stock were cultured in Luria−Bertani (LB) broth overnight at 37 °C. The cultured cells were washed and suspended in 1×PBS for detection usage. Bacterial cells were used from storage for sensing purposes after cooling to room temperature.

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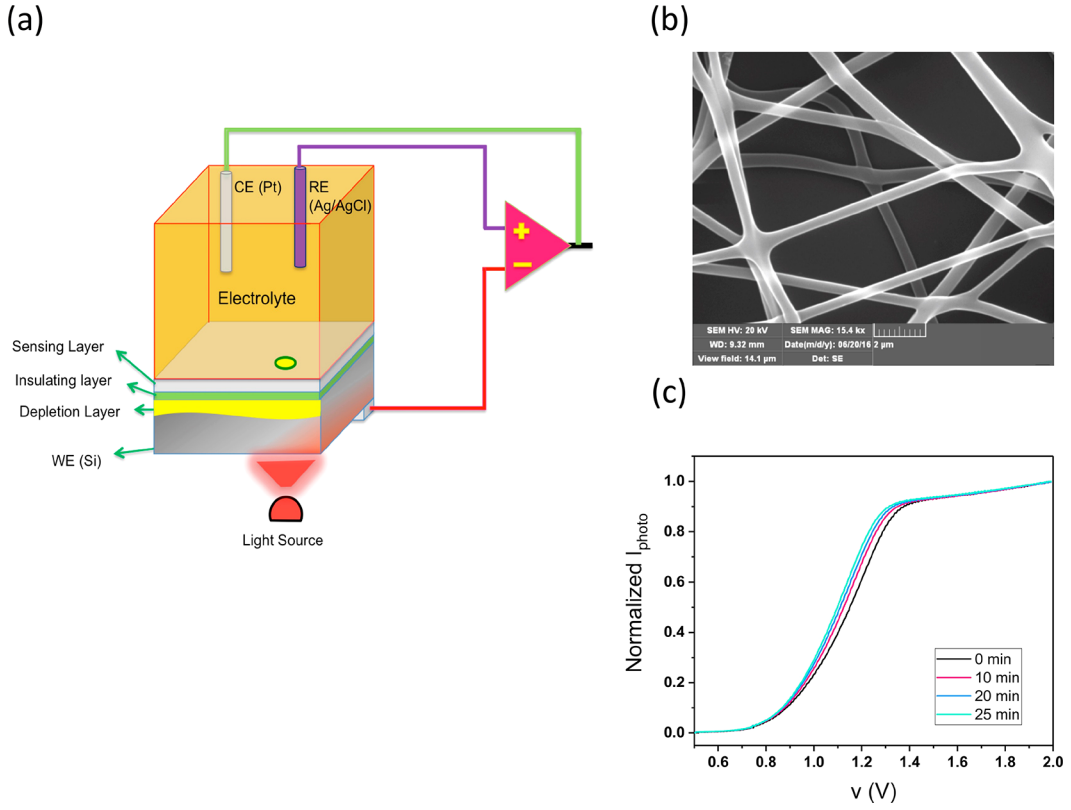

Figure 1. (a) Schematic representation of the NF-LAPS sensor comprising a three-electrode system. A Si-based sensor chip is acting as the WE with pH sensitive PAA/PVA hydrogel NFs, an RE comprising Ag/AgCl and a Pt CE. (b) Surface SEM image shows the hydrogel NF sensitive layer under magnification, with the scale bar depicting a 2 μm width. (c) NF-LAPS response with time for 10^6 CFU/mL *E. coli* in orange juice after signal stabilization. The photocurrent signal shifts toward lower applied voltage values, indicating a more acidic environment as a result of sugar fermentation by the bacterial cells.

NF-LAPS Signal Evaluation toward Acidic Environments. Before *E. coli* detection measurements in orange juice, the stability of the NF-LAPS was tested for acidic environments by adding different concentrations of HCl to tap water. In each step 20 μL of diluted HCl with a concentration of 3 μM was added to tap water.. To reach equilibrium with the swelling of the NF layer, 20 min runs were performed for each concentration. The acidity was then reduced by diluting the solution in two consecutive steps. Repeatability was evaluated by alternately making tap water acidic with 40 μL of the 3 μM HCl diluted solution and consequent dilution back to the original pH of 7. The pH values of all solutions were initially verified using a pH meter (Mettler Toledo FiveEasy). The stability of the system was also tested for a period of 125 min with a constant pH value of 7.2.

Detection of *E. coli* in Orange Juice with NF-LAPS. Orange juice was initially tested in the NF-LAPS with dilution factors of 1 (no dilution), 2, 4, and 6 to examine the response and stabilization time with respect to dilution. The pH of solutions was measured for comparison. Orange juice was then spiked with *E. coli* to achieve concentrations in the range of 10^2–10^6 CFU/mL. For all *E. coli* sensing measurements, glucose was added to the solution with a concentration of 80 μM to verify the fermentation of sugar and the subsequent pH change. Also, for all the experiments involving *E. coli*, glucose was added after 20 min to ensure that the pH-sensitive NFs had reached the swelling level associated with the initial pH of the solution. Each experiment lasted 1 h after adding the sugar.

A series of control experiments were also performed to verify the response of the sensor. Orange juice samples without any *E. coli* (blank sample) were tested for 1 h. The same experiments were then conducted on glucose/orange juice and *E. coli*/orange juice solutions. All experiments were performed at room temperature and repeated at least three times. Each test used 400 μL of solution. New chips were used every run to ensure reproducibility.

Selectivity Tests toward *E. coli*. To examine the selectivity of the sensor toward *E. coli*, *S. typhi* was used as a sugar fermenting bacteria. Orange juice was spiked with 10^6 CFU/mL *S. typhi* and tested with and without washing the surface of the sensor chip. The same procedure was followed for *E. coli* for comparison.

RESULTS AND DISCUSSION

Portable NF-LAPS Device. Figure 1a shows a schematic representation of the NF-LAPS system. As can be observed, the system consists of (i) a Si-based sensor chip that acts as the WE with pH sensitive PAA/PVA hydrogel NFs, (ii) an Ag/AgCl reference electrode (RE), and (iii) a Pt counter electrode (CE). The surface SEM image of the sensing layer confirmed the formation of cylindrical hydrogel NFs with an average diameter of 400 ± 50 nm (Figure 1b). Figure 1c depicts the photocurrent curves obtained with the NF-LAPS device for 10^6 CFU/mL *E. coli* in orange juice for the first 25 min after the addition of glucose. At lower applied potentials (0.6–0.8 V), where the sensor was in a state of accumulation, no photocurrent was generated. As the potential increased, a photocurrent was produced which indicated a state of depletion of the sensor. After initial stabilization where the NFs swell from a dry state to a wet state in accordance with the solution pH, *I*–*V* curves shift to the left toward lower applied potentials (non normalized curves are included in Supporting Information Figure S1) A possible explanation is that the pH of the medium was lowered as a result of the metabolic activity of *E. coli* and thus more photocurrent was generated at the lower applied voltage due to the higher number of positive charges present.

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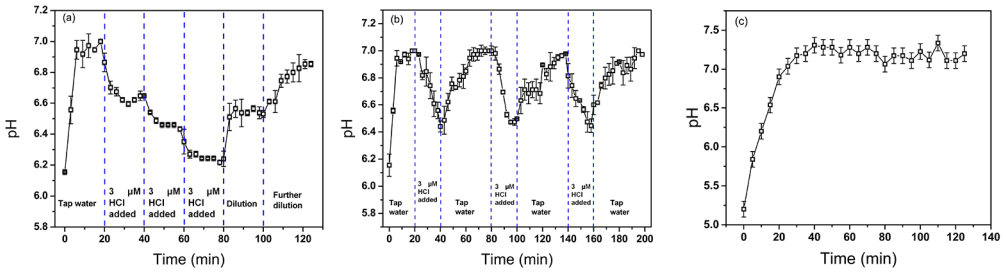

Figure 2. Response of the portable NF-LAPS system to (a) stepwise increase in the acidity of the medium and (b) cyclic acidification and dilution. Each cycle is 60 min long, 20 min for the acidification and 40 min for the dilution to increase the pH back to its initial value. All pH values are calculated based on the pH calibration curve of the NF-LAPS obtained previously with a sensitivity of 74 mV/pH.[22] (c) Stability of the NF-LAPS for over 100 min after reaching initial equilibrium in 20 min.

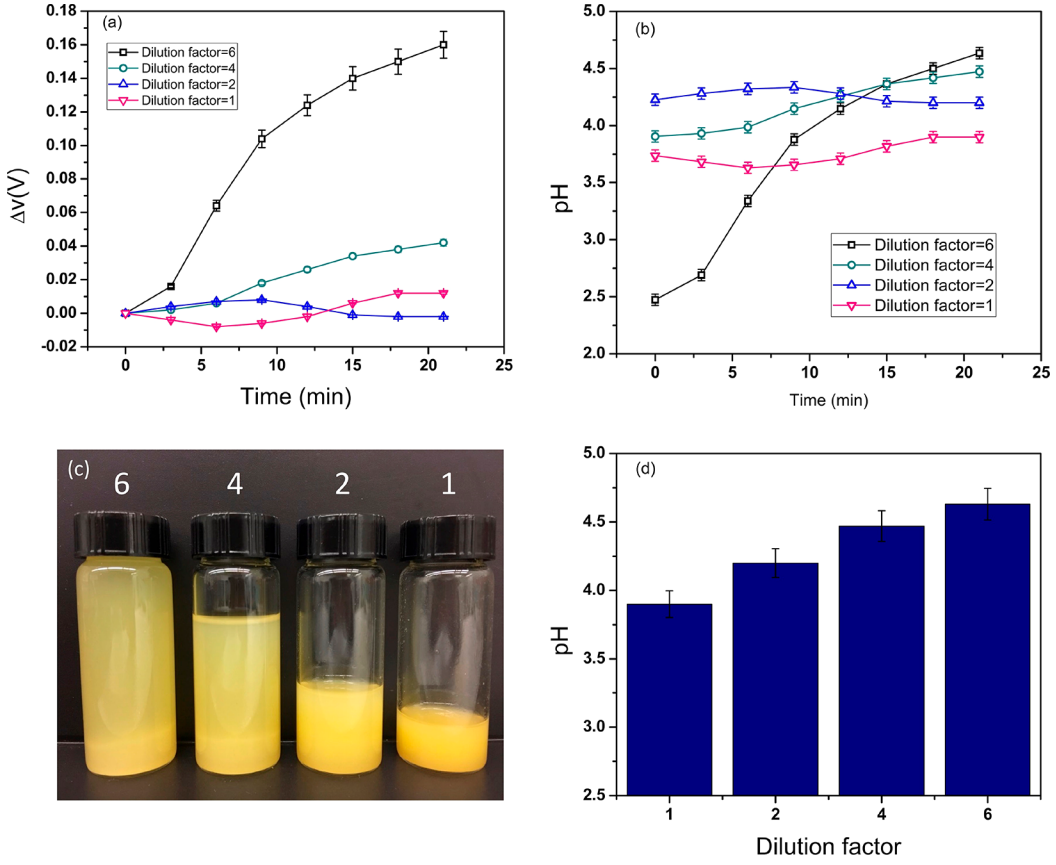

Figure 3. Effect of orange juice dilution on the NF-LAPS signal. (a) Shift in the inflection point of the $I-V$ curves with time for different dilution factors in orange juice. The signal is most stable where there is no dilution. (b) Calculated pH values with time for the various dilution factors. The graph shows the change in the pH values as a result of dilution. (c) Image of the orange juice with different dilutions, depicting the separation between the phases. Solutions without dilution proved to be the most reliable samples. (d) Final pH of the solutions after stabilization of the NF-LAPS. The measurements match bulk pH readings.

The inflection point of the curves was used to determine the pH value, based on previously obtained calibration curves.[22]

NF-LAPS Signal Evaluation in Acidic Environments. Response of the portable NF-LAPS system to stepwise increase in the acidity of the medium is illustrated in Figure 2a. When the sensor was exposed to tap water at pH 7, an initial jump was observed. This jump in pH during the first few minutes might be attributed to the swelling of hydrogel NFs from a dry state. The initial stabilization was achieved after 5 min; thereafter the pH values increased slightly and eventually reached pH 7 in 15 min. As the tap water was acidified in three steps with 3 μM HCl, the measured pH by NF-LAPS also decreased. This decrease is associated with the deswelling of the NFs at lower pH values. Following the last acidification step, the solution was diluted in two steps with the same volume as the added acid. The pH did not go back up to 7, as changes in pH were not linear with respect to volume.

Repeatability of the results is one of the most important evaluation parameters of sensors. Figure 2b shows the repeatability of the NF-LAPS response toward acidification. Each cycle consisted of acidification with 3 mM HCl solution and subsequent dilution back to the original pH value of 7. As can be observed, the developed NF-LAPS biosensor shows high repeatability and thus reliability in the measurement of pH. The results of the stability test can be seen in Figure 2c where after

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the initial equilibrium was reached the signal remained stable for the entirety of the experiment.

Detection of *E. coli* in Orange Juice with NF-LAPS. The effect of dilution of orange juice with the dilution factors of 1 (no dilution), 2, 4, and 6 was first examined. Figure 3a,b shows that the signal of the NF-LAPS device is more stable for no dilution case. A dilution factor of 2 also provides stable results; however, a drop in the voltage after 10 min indicates that swelling of the NFs might have been countered by a phase separation in the orange juice solution, as shown in Figure 3c. It must be noted that the measured pH of the diluted samples in Figure 3b does not represent the actual pH of the orange juice over time; it merely implies the delaying effect of dilution on the equilibrium in the system. For instance, without dilution, the calculated pH was very close to the actual pH value of orange juice (3.9). Table S1 in the Supporting Information shows the pH values in bulk measured using a conventional pH meter. The calibration curve used for diluted orange juice samples is included in the Supporting Information as well (Figure S2). Also, in this case, the variation in the measured pH was minor over time. Figure 3c clearly shows that there was a phase separation that occurred in diluted samples. From a microbiological point of view, Anvarian et al. showed that clarification and loss of the cloudy component of orange juice affects the viability of microorganisms in the beverage and may, in fact, increase the viability.[27] Despite the longer equilibrium times for diluted samples, the final pH values at 20 min were reasonable for all samples. The final pH values are shown in Figure 3d. As can be seen the pH of the diluted solution increased so that the orange juice with dilution factors of 1 (no dilution) and 6 had pH of 3.9 and 4.5, respectively. Taken together, these results suggest that the dilution of the orange juice sample is not required for the sensing measurements. Therefore, it is important not to interfere with the physiological interactions of *E. coli* and orange juice.

In the next step, the ability of NF-LAPS to detect *E. coli* in orange juice (without dilution), with 10^6 CFU/mL of the bacteria, was investigated. As can be seen in Figure 4, the pH value of the environment decreased by the addition of *E. coli*. Due to the presence of variety of sugars in the orange juice, fermentation occurred even when no glucose was added to the *E. coli* spiked orange juice. However, for real life applications, pH might have already been lowered if the beverage is contaminated beforehand with *E. coli*. Hence, it was paramount to add glucose to the mixture to explore whether further fermentation took place. The pH decline in Figure 4 implies that the incorporation of glucose has led to further extracellular acidification. It is worth noting that when there was no *E. coli* in the solution, pH did not show a significant change with time as is evident in Figure 4. It has to be mentioned that the successful incorporation of D-mannose on the surface of the NFs has been previously confirmed by Fourier-transform infrared spectroscopy.[22]

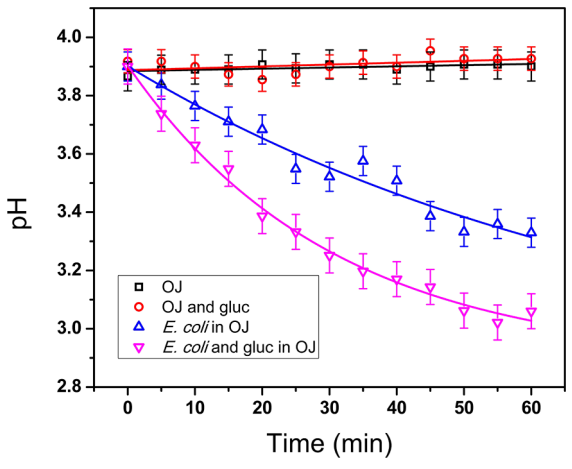

Figure 4. Drop in the pH of the media, as a result of the fermentation of sugars in orange juice spiked with *E. coli*. There is no noticeable change in pH with time in the absence of *E. coli*, with or without any sugar added. When *E. coli* is introduced, the media becomes more acidic due to the presence of a variety of sugars in orange juice; however, pH decreases even further when glucose is added to the medium.

To determine the sensitivity of the NF-LAPS device, a range of different *E. coli* concentrations from 10^6 CFU/mL down to 10^2 CFU/mL was tested. It is worth reiterating that the NF-LAPS signal was stable when glucose was added to the solution. Figure 5a displays the pH change with time for *E. coli* in orange juice after adding glucose. The higher the concentration of the bacterial cells, the lower the final pH of the media. The calibration curve was achieved for the device based on the overall change in the orange juice pH as a result of the metabolic activity of *E. coli*. The slope of the sensitivity line, depicted in Figure 5b, is 0.149 per CFU/mL of *E. coli* in orange juice with the regression coefficient of $R^2 = 0.989$. The slight deviation from the fitted line is associated with the lower concentrations of 10^2 CFU/mL and 10^3 CFU/mL, more likely because the amount of sugar available for the bacteria had been saturated. Hence, the change in pH is somewhat similar despite the fewer number of *E. coli* cells in the mixture. The measured limit of detection (LOD) of the NF-LAPS is shown to be 10^2 CFU/mL; considering the signal-to-noise ratio of the device, the theoretical LOD of the system is calculated to be 20 CFU/mL.[22] This measured LOD is comparable to the values reported by Jiang et al. where impedance spectroscopy was used to detect down to 10^2 CFU/mL[5] as well as Xue et al., who use a wireless microfluidic chip, with potential to be used outside the lab, and report a similar LOD.[28] However, in the work published in which Kim and colleagues utilized a microbial fuel cell, the lowest concentration tested was said to be 42 CFU/mL.[29] In this example, tests were not conducted at room temperature and there are no claims of the capability for the sensor to be used in the field.

Selectivity toward *E. coli*. Figure 6a illustrates the selective detection of *E. coli* in orange juice compared to the sugar fermenting *S. typhi*. When the surface of the sensor was unwashed, *S. typhi* resulted in a decrease in pH over time. The rate of pH decline was higher for *E. coli*, at the same concentration (10^6 CFU/mL), implying occurrence of more rapid acidification. When the surface of the NF-LAPS sensor chip was triple washed before adding glucose, the pH value dropped over time for *E. coli* while it increased slightly in the case of *S. typhi*. These results would seem to suggest that *E. coli* have a preferential affinity toward the D-mannose on the surface of the NFs as opposed to *S. typhi*. The difference in the specificity of various bacterial genera toward mannose has also already been established. Firon et al.[30] reported that the term mannose-specific bacteria is too general, and the combining lectin sites in *E. coli* differ significantly from those of *S. typhi*. In fact, all the *S. typhi* strains tested showed a different mannose binding behavior compared to *E. coli*.

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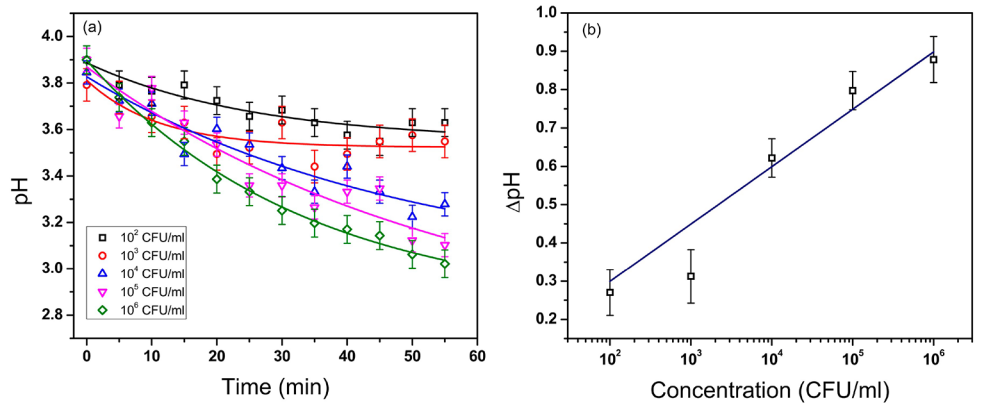

Figure 5. (a) Change in pH with time for various concentrations of *E. coli* in orange juice after adding sugar. Higher concentrations of the bacteria produce more acidic products, thus reducing the pH further. (b) Sensitivity of the NF-LAPS toward different concentrations of *E. coli* in orange juice. The fitted sensitivity line has a slope of 0.149 per CFU/mL of *E. coli* in orange juice and R^2 = 0.989. Lower *E. coli* concentrations of 10^2 and 10^3 CFU/mL do not show a significant difference in the final pH probably due to the fact that sugar may be in excess in the media and may have saturated the fermentation process.

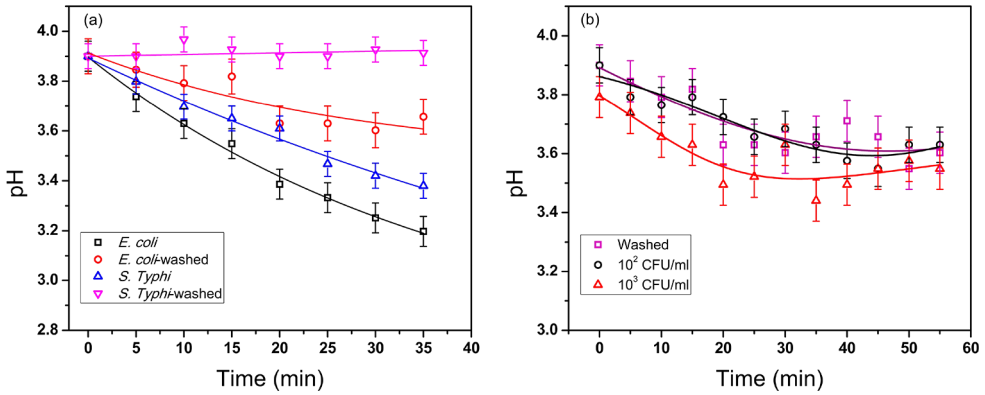

Figure 6. Selectivity of the portable NF-LAPS is tested against *S. typhi*. (a) Demonstration of the pH of the orange juice with time after the introduction of 10^6 CFU/mL of either *E. coli* or *S. typhi*. When the surface is not washed prior to testing, pH changes for both types of bacteria. However, pH becomes more acidic for *E. coli* in the same time period. When the surface of the sensor is washed before glucose is introduced, selectivity is achieved toward *E. coli* due to the higher affinity of *E. coli* to the D-mannose on the surface of the NFs. (b) Response of the NF-LAPS for *E. coli* after washing is compared to the known concentrations tested. The comparison and the sensitivity line of the NF-LAPS show that approximately 10^2 CFU/mL of *E. coli* remains on the surface after washing.

To further investigate the effect of washing, pH measurements were extended for *E. coli*. According to the obtained values established for different concentrations of *E. coli* in Figure 5a, approximately 10^2 CFU/mL of *E. coli* is estimated to have remained on the surface after washing as seen in Figure 6b.

The selectivity aspect of the NF-LAPS is a certain requirement when testing real samples, especially when a portable device is used. The evidence from this study suggests that the developed NF-LAPS sensor exhibits a high selectivity toward the target material, i.e., *E. coli*. This enhanced sensitivity and selectivity is predicted to eliminate the need for additional testing at sophisticated laboratory facilities. Additionally, to investigate the interference of other entities such as organic components in more complex media, real water samples were tested and the results shown in Supporting Information (Figure S3).

The sensitivity, selectivity, and compact nature of the entire NF-LAPS system render it the ideal candidate to be used in practical applications where portable sensors are in need and on-site detection is required. The relatively short detection time is also of value when safety tests are of the essence.

CONCLUSIONS

A successful and rapid detection of *E. coli* in orange juice was reported using a portable NF-LAPS device. The sensing measurements were performed by exposing the sensor to various solutions. The developed sensor was able to determine the presence of *E. coli* in less than 1 h. The sensitivity of the device toward *E. coli* was found to be 10^2 CFU/mL as compared to the calculated theoretical LOD of 20 CFU/mL. Furthermore, it was established that there is no need for dilution of orange juice prior to testing, which is an advantage for the developed sensor device. Repeatability of the responses has indicated the reliability of the sensor for applications outside the laboratory where reproducible results are desired. In addition, it was shown that the high selectivity of the NF-LAPS toward *E. coli* can be obtained by functionalizing the surface of the NFs with D-mannose that rules out the interference from the sugar fermenting *S. typhi*. Even without washing the surface of the sensor, the acidification of *E. coli* and *S. typhi* could be differentiated; however, for obtaining accurate results washing is recommended prior to testing. The developed portable NF-LAPS has great potential to be used as a monitoring device to

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ensure the safety of orange juice as well as other fruit juices at various stages of production, distribution, and consumption.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acssensors.8b00063.

Additional information on the $I-V$ curves provided in Figure 1c before normalization; additional information regarding diluted orange juice experiments; results from testing real water samples in the NF-LAPS system (PDF)

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Notes

The authors declare no competing financial interest.

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